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The long term stabilization of uranium mill tailings

Final report of a co-ordinated research project 2000–2004

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International Atomic Energy Agency

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FOREWORD

The IAEA attaches great importance to the dissemination of information that can assist Member States with the development, implementation, maintenance and continuous improvement of systems, programmes and activities that support the nuclear fuel cycle and nuclear applications. This includes managing the legacy of accidents and past practices, including that from uranium mining and milling.

A comprehensive IAEA programme of work covers multiple aspects of environmental remediation:

- technical and non-technical factors, including costs, that influence environmental remediation strategies and pertinent decision making;
- site characterization techniques and strategies;
- assessment of remediation technologies;
- techniques and strategies for post-remediation compliance monitoring;
- special issues such as the remediation of sites with dispersed radioactive contaminations or mixed contamination by hazardous and radioactive substance and of uranium mining and milling sites.

In the past, often little or no care was taken to isolate uranium mill tailings from the environment. In order to address the specific problems surrounding the disposal of uranium mill tailings, the IAEA developed a co-ordinated research project (CRP) in this area. CRPs are intended to bring together researchers from different Member States with the view to share and disseminate the experience in solving problems of common interest.

The aim of the CRP on the long term stabilization of uranium mill tailings was to contribute to the development of conceptual, technical, and management solutions that:

- render tailings more inert over prolonged time-spans;
- render impounded materials and engineered structures stable;
  over prolonged time spans;
- minimize the need for active maintenance;
- can be applied in a remediation context; and
- support solutions that are technically and economically feasible.

The IAEA wishes to express its thanks to all Chief Scientific Investigators and other contributors to the work. Special thanks are due S. Needham, Australia, who under contract provided a seed review of tailings disposal techniques, and to H. Cotzee and R. Gatzweiler, who reviewed an early draft.

The IAEA officer responsible for this technical publication was W.E. Falck of the Division of Nuclear Fuel Cycle and Waste Management.

EDITORIAL NOTE

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1. INTRODUCTION

1.1. Background

Large volumes of low activity milling residues, such as mill tailings, are produced – sometimes exceeding millions of tonnes at a single uranium mining/milling facility, in particular, when uranium is only a by-product. The common mode of disposal is in near-surface impoundments in the vicinity of the respective mine or mill [1]. Such impoundments were often arranged in a haphazard fashion, utilizing geomorphological depressions or by filling-in valleys. As a result, there was (is) little or no care taken to isolate the tailing materials from their environment.

While geomechanical aspects, such as the stability of pile slopes, dikes and retaining dams, are standard engineering problems, for which in most countries provisions are made in the relevant building or mining regulations, environmental and radiological impacts have often been neglected. It should be mentioned, however, that mill tailings as such can pose serious engineering challenges, owing to the geomechanical and physico-chemical characteristics of the sediments.

Fig. 1. Typical uranium mill tailings pond (Urgeiriça, Portugal).

Typical environmental problems arising from mill tailings are radon emanation, windblown dust dispersal, and the leaching of contaminants, including radionuclides, heavy metals and arsenic, into surface and groundwaters. Radon (Rn) emissions are due to exhalation from the waste materials and the Rn can reach the ambient atmosphere when free circulation of air in the material and its cover is possible. Emissions to water bodies occur when infiltration of precipitation is unhindered, bottom-liners are absent, and no collection of drainage waters is installed. The leaching of contaminants is usually exacerbated by acid formation from pyrite oxidation under conditions of varying degrees of saturation with water. Additional effects from acid rain have also been observed. In many instances contaminants other than radionuclides may be the real problem, and a comprehensive and holistic assessment of the impoundment inventory and all processes may be necessary.

A range of technical measures can be employed to prevent or reduce the extent of these processes. Capping can be used to control radon emanation, moisture infiltration and chemical reactions that may promote leaching. The physical and chemical properties of the tailings can be improved *in situ* or by reprocessing to enhance long term stability. Containment structures can be improved to meet the minimum factor of safety. Tailings drainage canbe collected and treated in the short term, until the discharge standards set by the appropriate regulator(s) are met. If such measures are determined not to meet long term objectives, relocation of tailings may be considered.

Any engineering solution has a finite life-span, which may be shorter than desirable from a radiological or toxicological safety point of view. Apart from the structural degradation and/or weathering of the material impounded, failure of retaining structures, such as dams, must be considered. Erosion of cappings and other engineered structures may be a problem in certain settings. Engineering solutions, therefore, may need to consider long term care and maintenance as an integral part of planning and design. In turn, this may require active institutional control and stewardship over very long periods of time. Engineering solutions, long term care and maintenance and institutional control should together strive for an optimization of economic, technical, risk reduction and societal factors.

The aim of searching for long term solutions is to limit risk to future generations and minimise the commitment of future resource requirements [2]. Design requirements for disposal longevity generally range from a few hundred to a thousand or more years. For example, the USA EPA promulgated standards for long term stabilization and control of uranium mill tailings [3] require that the remediation: “Be designed to be effective for up to 1000 years to the extent reasonably achievable, but at a minimum for 200 years.”

Based on the objective to keep environmental emissions to a minimum over long times, the task, therefore, is to find conceptual and technical solutions

- that render tailings more inert over prolonged time-spans,
- that render impounded materials and engineered structures stable over prolonged time spans,
- that minimize the need for active maintenance,
- and that are technically and economically feasible and acceptable to society.

The emphasis of this CRP is on technical solutions that can be applied in a restoration/ remediation context. Of crucial importance in this particular context are costs, as these frequently have to be borne by the taxpayer and can no longer be included in the product price. Any proposed expenditure has to be carefully balanced against the likely benefit from such measures, implying that a comparison of forecast environmental and radiological impacts with and without the measure is to be undertaken beforehand.

1.2. Objectives

1.2.1. Overall objective

Dozens of uranium mining/milling sites have been shut down over the last few years. To ensure long term stabilization and isolation of residues is but one element in sustainable and environmentally responsible plant operation schemes. However, legal requirements, environmental targets and standards, economic resources available, and hence the actual management and remediation/restoration practices may vary considerably from Member State to Member State. This CRP is proposed as one step towards raising the awareness of potential problems and assisting Member States in the development of efficient procedures and processes for the sustainable long term management and, if deemed appropriate, remediation of uranium mining/milling waste sites, and to encourage a harmonized and systematic approach where feasible.

1.2.2. Scientific research objectives

The overall objective of stabilization and isolation of mill tailings and other uranium mining residues is to minimize exposure of target groups from radiation and contaminants in the various environmental media. This can be achieved by creating conditions resulting in low source terms for solid, aqueous, and gaseous releases, and by designing disposal facilities resistant to failure.

Long term stabilization and isolation of mill tailings is an active R&D area, covering *inter alia* the development of new techniques for tailings deposition, the geomechanical and geochemical stabilization of waste materials, and the design of advanced barriers, both at the bottom and as cappings. A closely related field that has seen rapid technological advances over the past decade is the restoration/decontamination of contaminated land, and the remediation of engineered landfills.

It is recognized, however, that the above objectives cannot exclusively be achieved by engineering design, but must involve also adequate management and planning procedures. Hence, the long term stabilization of uranium mill includes, *inter alia*, the following topical areas:

Planning and management

- Site characterization;
- assessment of likely and probable environmental impacts due to radiological and non-radiological contaminants;
- identification of processes relevant to the long term performance;
- design features that improve long term performance;
- conceptualization of time-frame for closure;
- conceptualization of remediation goals and techniques;
- definition of factors affecting long term care and maintenance and the need for institutional control;
- methodologies for quality control and quality assurance (QA/QC);
- design of cost-effective long term surveillance and monitoring programmes for
  - environmental performance;
  - geotechnical performance.

Technologies

- identification of properties relevant to the long term environmental and geotechnical performance of tailings and structural materials;
- structural integrity of impoundment, *viz*.
  - design features controlling the long term stability of engineered structures, e.g. dams;
  - techniques for *ex post* improvement of isolation, e.g. bottom seals;
  - design features controlling erosion resistance;

    - *in situ*/on site techniques for *ex post* treatment of existing tailings, e.g. solidification, de-watering, capping;
    - techniques for (*ex post*) improvement of the long term geotechnical performance of waste materials, *viz.* biochemical and geochemical resistance of sealants/additives with respect to structural degradation;
- techniques for cost-effective characterization of radionuclide inventory, *viz.* determination of source term characteristics;
- techniques to minimize long term contaminant release and to improve geochemical stability of tailing materials including *in situ*/on-site techniques for *ex post* treatment of existing tailings, i.e. to reduce leacheability and/or permeability, or to reduce Rn emanation;
- Low maintenance/cost or maintenance-free drainage systems and drainage treatment systems for removal of radionuclides and other contaminants;
- tools (models) for the assessment/prediction of long term environmental and geotechnical performance;
    - mechanistic models
    - systems analyses
        - fault tree analyses
        - incident sequence analyses;

Institutional, legal and economic aspects
- site release criteria and use restriction criteria;
- applicable legislative and regulatory regime for radiological and non-radiological issues;
- funding of and liability for remediation/restoration activities.

1.3. The focus of the CRP

The projects for this CRP were selected to provide a number of focal areas and clusters of related projects. The main emphasis, however, was on the technological aspects and the design aspects as relevant for the development of appropriate technologies.

The influence of institutional, legal, management and socio-economic aspects on decision making in remediation/restoration projects and the problem of site and source-term characterization is being addressed by other IAEA projects [4][5], while the environmental issues in uranium mining and milling in general are discussed in joint reports by OECD/IAEA [6][7].

It is expected that this CRP will contribute to the transfer of technologies and know-how within the international (uranium) mining/milling, waste disposal and contaminated land communities. The specific problems arising from the properties of relevant radionuclides and the properties of tailing materials have to be addressed. Special emphasis has been given to the development of innovative methods and techniques for stabilization.

The objective of the proposed CRP was to encourage the sharing of practical experience (adaptive research) and (applied) R&D work by Member States on topics relevant to the long term stabilization/isolation of mill tailings.

1.4. The structure of the CRP

The projects composing the co-ordinated research project were grouped into four subject areas (Table 1). Projects whithin these subject areas are intended to complement each other. The final reports on the individual projects are given in Annexes I to XIII.

Table 1. The project composing the CRP

Project Title	Principal Investigator	Country
Subject area I: Tailings remediation case studies		
A study case on the uranium tailings dam of Poços de Caldas uranium mining and milling site	H. Fernandes	Brazil
Cameco Research and Development Projects for Tailings Disposal Technology	P. Landine	Canada
Subject area II: Capping of tailings		
Studies of bentonite and red soils as capping of the uranium mill tailing impoundments	Z. Wen	China
Development of Method of Covering Raising Dust Beaches of Radioactive Wastes Storage Out of Operation	A. Gagarin	Kazachstan
Polymeric Coats for Contaminated Surfaces Localization	S. Mikheykin	Russian Federation
Improvement of Soil Properties Applied to Capping and Multi-Layer Barriers	J. Koszela	Poland
Subject area III: In situ conditioning of materials		
Remediation of Uranium Mill Tailings Using Natural and Organo-Clays	S. Choi	Korea
Development of Technologies for In-Situ Remediation of Contaminated Sites by Directed Formation of Naturally Occurring Slightly Soluble Minerals	G. Ziegenbalg	Germany
Room Temperature Ceramics, the Breakthrough Material for Long Term Stabilization and Isolation of Low-Waste Uranium	A. Piestrzynski	Poland
Research and Development of Measures to be Taken for Long Term Stabilization of Uranium Liquid Wastes	G. Maslyakov	Ukraine
Subject area IV: Management of tailings in remediation situations		
Predicting the Long –Term Stabilization of Uranium Mill Tailings	J. Trojacek	Czech Republic
Harmonization of Radiological Impact Assessment Methodologies of Uranium Mill Tailings Repositories	A.-C. Servant	France
Holistic Approach to Remediating Uranium Mill Tailings and Contaminated Groundwater	D. Metzler	USA

2. HISTORICAL PRACTICES

2.1. The ‘ages’ of uranium production

The special risks that were associated with uranium mining were implicitly evident as long ago as the early 16th century, when in central Europe workers in silver mines, where in our days a uranium mineralization was recognised, appeared to be more susceptible to pulmonary disorders than workers at other mines. In 1879 such diseases were diagnosed as lung cancers [8]. Concerns regarding operational practices are therefore commonly greater at uranium mines than elsewhere, although the focus was on situations where workers were in immediate contact with the ore and processing streams. These concerns for worker health and safety have been the driving force for gradually tighter controls and improved practices over decades. Only in relatively recent times have concerns developed for impacts upon public health and on the natural environment from the full range of operational activities related to uranium mining and the remainder of the nuclear fuel cycle. The concerns for the natural environment include risk of environmental degradation, contamination, reduced ecosystem viability and biodiversity, aesthetics, public amenity, access to land, and quarantining of land for future beneficial land use.

Uranium mill tailings are of particular environmental concern because they:

— retain the majority of the radioactivity of the ore from which they are derived;
— their radioactivity is very long lived;
— contain a range of biotoxic heavy metals and other compounds
— may contain sulfidic minerals and thus prone to generate acid mine drainage
— their granular to slime constituency makes them readily leachable, erodable or collapsible under different conditions;
— the common method of surface disposal exposes a large surface area to the natural elements and thus increases the risk of release of radiation flux, radioactive and geochemically toxic dusts, and interaction with surface water systems;
— the large surface area of these generally thin tailings deposits (or ‘piles’) adversely affects large areas of land and renders potentially valuable land unfit for other uses.

The history of uranium mining can broadly be divided into the following ages, reflecting the main use for which the uranium was being mined, the urgency of this task, and the evolution in understanding of the character, risk levels and governmental and societal responses to the hazards associated with this activity [9].

Before the 1940s many areas around the world were worked on a small scale to produce radium for medical purposes, luminescent material for the manufacture of luminous dials etc, and material for research into radioactivity. The same areas commonly also yielded quantities of uranium used as a bright yellow pigment in glass making and ceramics. Ore was commonly hand-sorted and no tailings fitting the definition above were produced. Several of these areas became significant uranium mining area in later times.

From the mid 1940s to the mid 1960s a concerted attempt was made to discover and develop uranium resources to supply feedstock uranium oxide for the development of military weapons. Owing to the newly discovered military and strategic significance of uranium, exploration and mining of uranium by the private sector was banned in Canada, USA and the UK from 1943 so that the industry was totally government-controlled [6] (the ban was lifted in 1948). Development commenced during the Second World War continued through to the

1960s, demanding continued supply and acquisition of stockpile material. This urgency fuelled the first "uranium rush"; for example, the governments of the USA and UK stimulated exploration for uranium by cash rewards for new discoveries. Commodity price played no part, as many mining contracts were on a "cost-plus" basis to ensure a reliable supply to those governments regardless of cost. In other countries, exploration as well as development was largely in government hands in order to match the build-up of uranium materials and weapons stockpiles for strategic military purposes. Notable in this period were the:

— rapid development of many new mining districts (or enlargement of some areas previously worked for small quantities of uranium and radium);
— development of regulatory frameworks focussed on worker health; and
— tailings disposal practices that generally saw tailings placed adjacent to mills using practices common to other metal mill tailings of the day — i.e. no design features to improve containment security, or reduce radon flux, or isolate the tailings from wind or water erosion or interaction with surface or ground water systems;
— the small size of many mines, and the level of government control, saw many of the mines serviced by central mills, thus focussing tailings disposal problems in a relatively few areas that (for reasons of transport, employment, accommodation and servicing) were often close to urban centres.

The first uranium rush had subsided by the end of the 1960s, but the potential of nuclear power for peaceful energy generation had been realised. Concerns over the limits of oil supply for the World's energy needs led to projections for substantial increases in the then depressed price of uranium, so that from the mid 1960s to mid 1970s a second 'uranium rush' eventuated. This differed significantly from the previous rush in that:

— the impetus was driven by market forces and the World commodity market;
— a range of much larger uranium deposits was discovered;
— economies of scale at these larger mines and extended profitable mine lives allowed more thorough planning of facilities including tailings facilities and integration of mines with dedicated mills;
— lower production costs in association with the downturn in uranium prices at the end of this period forced the closure of many of the smaller mines developed in the 'first rush'.

The last 'age' of uranium mining coincides with the period of environmental enlightenment ushered in since the mid-1970s. This enlightenment was born out of an awareness of the level and types of impacts that human activity, including mining, was having on the environment. In particular, this period coincided with:

— development of the first set of national environmental regulations in several key uranium-producing countries;
— introduction of legislation for and conduct of the first environmental impact assessments for major mining operations (such as the Ranger Uranium Environmental Inquiry in Australia [10];
— the public health risks from the nuclear fuel cycle were becoming widely known;
— concern for possible impacts upon the natural environment were developing;
— environmental impacts from the previous generation of uranium mines were becoming understood.

This 'age' has also been a period of major research efforts into the environmental issues related to uranium mining, and to uranium mill tailings in particular. The largest single incentive for research was related to passage of the Uranium Mill Tailings Radiation Control Act 1978 (USA), under which the US Environmental Protection Agency established standards to be used for remediation of 24 sites across ten USA States, where the health of the public and the environment had been put at risk from inappropriate placement, isolation and security of uranium mill tailings [11].

Research has continued at a high level since the late 1970s. Advances have generally been uneven between different countries because of the economic value of the uranium resource; the level of public and regulatory concern; and the levels of funding made available for technology development [8]. The two main research thrusts today are:

(1) into remediation technologies for those countries where the impetus for remediation action has begun only recently. For instance, in Eastern European countries since the fall of the Soviet regime and sufficient funds have only recently been appropriated through programmes such as PHARE [12];

(2) into technologies for improved placement, containment and isolation of tailings at new and currently operating mines, in order to avoid the failures of the past and to provide successful long term containment that will not impose significant financial, health or environmental liabilities on future generations.

It must be said that the 'ages' described above have not occurred over the same periods in different countries. The varying levels of strategic importance given to uranium as an energy and weapons feedstock; varying times of opening up to and participation in a free market for uranium; and varying levels of secrecy and centralised control of the uranium mining and nuclear sectors, has meant that many countries have progressed through these "ages" at considerably slower rates. Most notably the Soviet block countries were only able to progress after the disintegration of the USSR. Uranium mining in eastern Germany continued under tight military conditions and total secrecy until 1990, and was not subject to control by national regulatory authorities even after 1962 when radiation protection standards had been introduced [13].

2.2. Age, number, geographic and climatic distribution of uranium mill tailings piles

The geographic distribution, age, number and climatic distributions of uranium mines and mills is summarised in Table 1. Some information is also listed on the quantum of tailings for some countries. The data are incomplete and approximate only, but indicate that uranium mining and milling have been widespread and that the issues of remediation, health risks, environmental impact, financial costs and future liabilities are shared by every continent and almost every major climatic zone (i.e. except for the polar and mountain zones).

With the advent of a free market for uranium and it being removed as a strategic stockpiled commodity by most countries, many smaller, high-cost mines have become uneconomic and have closed down. Many mines that produced uranium as a byproduct have abandoned uranium production and closed their plants. Therefore the number of countries faced with the need to develop techniques for modern operating mines is relatively small and obviously limited to those endowed with lower-cost deposits — generally large unconformity deposits or small-medium sandstone deposits amenable to *in situ* leaching (ISL).

Table 2. Age, number and size of closed and operating uranium mines and tailings piles by country and climatic zone, and general status of current activities. Main information sources: [7] [14] [15] [16] [17]

Continent	**Country**	**U mining since**	**No. hard-rock mines**	**No. U mills**	**Climatic zone**	**Tailings – no. piles**	**Tailings – volume [10^6 m^3]**	**Tailings – area [ha]**	**Country status**
AFRICA	Gabon	1956	5	2	tropical	7			Mining and milling ceased 1999, rehabilitation due for completion in 2000
	Namibia		1		hot desert				
	Zaire		3		subtropical				
	South Africa		34	3	dry grassland				
NORTH AMERICA	Canada	1933	24	14	taiga	34	ca. 30	ca. 300	Old small sites mostly rehab'd. Major current producer from 3 mines + 3 mills
	United States	1898	3900	< 100	dry grassland, hot desert	> 52	120		Major rehab program (UMTRA) completed 1979-99
SOUTH AMERICA	Argentina	Early 1950s	11	9	dry grassland, hot desert	8			Mining and milling ceased, planning for rehabilitation in progress
	Brazil	1981	1	1	tropical	1	2.17	86ha	Mining/milling ended 1997, restoration pending
AUSTRAL ASIA	Australia	1930s	25	8	tropical, dry grassland, hot desert	10	48.6	620	Old small sites mostly rehab'd. Major current producer from 2 mines
ASIA	China		8		subtropical, dry grassland				
	India		4	3	subtropical				
	Japan	1957	1	1	temperate	1	0.03	12.5	No mining, mill dismantled 1981
	Kazakhstan	1955	19	3	dry grassland	3	209	1733ha	Only ISL continues – hard-rock mining ceased
	Krgyzstan		4	1	mountain, hot desert	32			Many mines closed in early 1990s, dumps unremediated. Radioactive tailings still produced as uneconomic (Th) by-product of Au Ag Pb REE mining
	Uzbekistan		4	1	hot desert	1	30.0	600	

Continent	Country	U mining since	No. hard-rock mines	No. U mills	Climatic zone	Tailings – no. piles	Tailings – volume [10^6 m^3]	Tailings – area [ha]	Country status
EUROPE	Bulgaria		20	2	mediterranean	3	18.5		
	Czech Republic	1948	13	3	temperate	21	46.8	638	Reduced production matched to national energy needs, mining expected to cease 2002. Rehab of closed sites underway
	Estonia		1	1	taiga	1	8.0		
	Finland	1958	1	0 (ore exporte d)	taiga	1	0.04		No mining since 1961
	France	1950s	180 (some very small)	8	temperate	19	47.3	256	Mining and milling stopped, old sites being progressively rehab'd since 1990
	Germany	1946	9	4	temperate	15	161	727	Production ceased 1990, major Wismut clean-up in progress
	Hungary	1956	1	1	temperate	2	20.4	163	Production ceased 1997, rehab planning in progress
	Poland					1	0.114		
	Portugal		55	1	mediterranean	2	3.5	10	1 mine operating, minor production. Rehabilitation of old mines since 1990
	Romania		10		temperate	>3	4.5	37	Production from several sites to end in near future, rehab planning underway
	Russia	1950	21	2	dry grassland, taiga	3	54.1	457	Production continues, old workings rehab'd, rehab and decommissioning plans for operating plants being reviewed
	Slovenia		1		mediterranean	1	0.7		
	Spain	1959	21	2	mediterranean	>3	2.4	25	1 mill operating at 1/3 capacity. Old site progressively rehabilitated since 1991 and continuing
	Sweden	1965	1	1	temperate	1	1	25	Production ceased 1969, site rehabilitated 1990-93, ongoing leachate treatment, monitoring & maintenance
	Ukraine	1950s	6	2	dry grassland	3	130	686	Production continues from one mill. Restoration activity started in 1991 has paused owing to economic difficulties
TOTALS			4196	163		186	908	5769	

Available information does not permit data on the age of tailings piles to be tabulated. However it is clear that many tailings piles are not yet fully remediated, and that several of these date back to the 1950s. Some that have been remediated may require further attention as the effectiveness of isolation becomes evident — for example at Rum Jungle and the South Alligator Valley in Australia where remedial work undertaken in the 1970s and 1980s has proven not to meet standards for environmental protection expected today, and planning is now commencing for a second programme of remediation works.

2.3. Relationship of tailings piles to mine and mill facilities

In general terms, older uranium mining involved ore extraction from a large number of small workings, usually from shallow excavations clustered in ‘uranium districts’. It was common practice for there to be only one mill processing the ore from each district, and in some situations a mill would also treat ore from outlying mining districts as they were discovered. This trend was reinforced by individual mining companies commonly being too small to afford to build a processing plant of their own; of mines not having sufficiently long lives to warrant the cost of constructing dedicated processing plants; and tight government control on the production of uranium oxide because of its high strategic importance from the 1940s to the 1960s, and later in some countries.

It was common for these ‘central’ mills to be built close to towns in order to benefit from transport and industrial infrastructure, and to access workers and suitable accommodation for them. Grand Junction, Colorado, is a good example of a central mill constructed in a previously established town [18]. Some metallurgical processing plants in major towns were modified to allow processing of uranium, such as at Port Pirie in South Australia, where ore was trucked 300 km from Mount Painter/Radium Hill for treatment.

The practice at almost every uranium processing plant is to dispose of the tailings at the nearest convenient place. Therefore the approach of processing uranium ore at central mills close to towns had the unfortunate consequence of placement of tailings close to populated areas, where risk of exposure to and interference from people was highest. In some instances, mills originally built for processing other types of ore were converted to uranium processing plants; at Uravan in Colorado USA, the vanadium processing plant was converted to a uranium mill in 1948 and treated ore from over 200 small mines in the region [19].

Over time the central mill philosophy has still been used for good economic reasons, but as more sophisticated exploration and mining techniques resulted in the discovery of larger uranium deposits, commonly with lower grades or with ores less amenable to simple on-site high-grading by hand picking etc, mills tended to be built to serve a single mine, or within a cluster of mines (the clusters of mines reflecting how uranium deposits often occur in ‘uranium fields’). Examples include the Elliot Lake district in Ontario, Canada; and Rum Jungle in northern Australia. These mills were built away from established towns, and consequently posed less risk to the health and welfare of people. However, because the same ‘convenience’ approach to placement of tailings applied, the tailings piles still posed a considerable hazard to the environment.

Where uranium was discovered in remote and undeveloped areas, the mines and mills became the focus of development and towns were built within easy commuting distance to the mills. Early examples are Beaverlodge in northern Saskatchewan, where Uranium City was sited about 6 km from the mill, and Rum Jungle in northern Australia, where the township of Batchelor was built about 8 km away. At other remote sites no permanent facilities were constructed so that the local population at potential risk from the tailings piles was at or close to zero (e.g. the Rockhole tailings deposit in the South Alligator Valley of northern Australia [20]). Construction of mills away from population centres significantly reduced the level of impact upon human health relative to mills constructed in or on the fringes of established settlements. However, the same range of approaches was used for siting, containing and depositing the tailings, such that similar impacts upon the environment eventuated wherever tailings piles were made.

In areas where uranium was produced as a by-product on existing mines, e.g. the gold mines of the Witwatersrand, uranium tailings and plant wastes were often mixed with far larger volumes of tailings. Furthermore, in some mines, the tailings contain significant concentrations of uranium and other radionuclides.

2.4. Historical approaches to tailings placements

Past practices for placement of uranium mill tailings are summarised as:

- no effective containment
- topographic depressions
- within a custom-built ring-dyke or turkey nest dam
- returned to an underground mine
- low embankments
- returned to a mined out pit
- in a valley, usually behind a dam or dyke
- into a deep lake or river.

This list includes effectively all of the options available today for effective containment of tailings, but is also contains options that are now considered unacceptable because of the high probability of containment failure. The main difference between historical placement and that practiced today is that in the past no risk or impact assessments were done and little or no regard was given to selecting the option that would impact least upon human or environmental health. Indeed in the 1950s when many mines and mills began operation, uranium mill tailings were not considered to be problematical, and techniques little different to those used for non-radioactive tailings were employed. The drivers for choosing a disposal site were convenience and cost.

In the following historical examples are given for each of the types of disposal.

2.4.1. No effective containment

In Kyrgyzstan at Mailii Su, 23 separate tailings piles are situated in deep ravines and on river banks falling in to the Mailii Su River and its tributaries; at Kaji-say a tailings pile lies on the banks of a wadi (dry river bed) less than 3 km upstream of a large lake important as a tourist destination [14]. In Slovenia the disposal site near Borst consists of tailings with dams on a sloping area of ground [21]. In Australia, tailings from the Rockhole mine in the South Alligator Valley were placed on a flat area in the floodplain of the South Alligator River and immediately adjacent to the river bank; the river system is now part of the World Heritage listed Kakadu National Park [22]. Some tailings at Grand Junction, Colorado USA were placed on flat ground adjacent to the mill in the centre of the city [22]. Further south at Uravan in Colorado, the tailings were placed on a bench along a canyon wall with no provision for physical stability or containment to prevent cascading to the canyon floor and river below [19].

2.4.2. Low embankments

At Rum Jungle and Moline in northern Australia, tailings were deposited behind low bunds constructed on gentle slopes that were prone to over-topping by supernatant water in the tropical wet season and uncontrolled discharge into local creeks and rivers [6].

2.4.3. Topographic depressions

The mill at the Rabbit Lake mine in northern Saskatchewan deposited tailings into a tailings basin constructed by damming a natural depression in the bedrock [23], and some of the tailings generated at Elliot Lake in Ontario were also placed in topographic depressions [24][25]. In Grand Junction Colorado, some tailings were placed in shallow depressions behind river bank levees along the river flats with only minor additional earth works to aid containment.

2.4.4. Valley fill

This approach is more common in mountainous areas owing to the scarcity of flat ground for other forms of construction. In Kyrgyzstan at Ming Kush tailings were deposited in a permanent stream valley and the stream flow diverted around the pile by two bounding canals [14]. At Jaduguda in India, the cyclones sand fraction of the tailings are used as underground backfill, and the slimes are pumped to a natural valley site with decant allowed to flow over an earthen dam [26]. At Nejdek in the Czech Republic tailings were placed behind a dam on the Rolava River, which receives flows of supernatant water [7]. Tailings at the Priargunsky operations near Krasnokomensk in Russia are held in two valley-fill dams [7].

2.4.5. Ring dyke or turkey nest dam

One of the earliest dams of this type was built in 1958 at Grants, New Mexico USA, where tailings from the Kerr-McGee mill were totally surrounded by a high embankment built in several 'lifts', with each lift being constructed of the cycloned coarse tailings fraction [22]. This approach was taken owing to the absolute flatness of the landscape and the absence of any natural depressions, embankments or levees. Whilst statistical information on construction methods of tailings impoundments is only fragmentary, it is probable that this method is now a common form of tailings facility construction [7]. It had become a popular technique by the 1970s, for example in France where many tailings storages constructed in this decade are of this type (e.g. Gueugnon, Rophin, Bois Noirs Limouzat, Ecarpiere, Lavaugrasse, Jouac).

2.4.6. Mined out pit

This approach is confined to situations where the mill is in an area of previous extractive mining or quarrying, or where the deposit being mined is amenable to progressive excavation and back-fill – for example where the deposit is made up of a series of ore lenses. At Ranstad in Sweden, tailings were back-filled into worked-out parts of the shallow open-pit [22]. Tailings from the Cellier, Brugeaud, Montmassacrot, Bellezane, Lodeve and St. Pierre du Cantal mines in France are held in old open pits [7]. One of the three impoundments at Ukraine's Zhovty Vody mill is in an old iron ore open pit [7]. Two old open pits were used to dispose of uranium mill tailings at Seelingstädt in Germany [15]. At Nabarlek in Australia, tailings were pumped directly to the pit from which the uranium ore was extracted, facilitated by the high-grade ore being mined out within a year and stockpiled at the beginning of the project; milling then took place over ten years as the stockpile was depleted [7]. The same was undertaken at Rabbit Lake, Key Lake, and Maclean Lake in Sasketchawan, Canada.

2.4.7. Underground mine back-fill

Some tailings from the Gunnar mill in Saskatchewan, Canada, were returned into the underground workings [7]. It is also considered as an option for the remediation at the Zirovski Mine in Slovenia [21] and some mines in the Ukraine (see Annex XII) and in some cases for the Wismut mines.

2.4.8. Deep lake or river

Tailings disposal from the Port Radium mill in Canada's Northwest Territories included placement in several small lake basins and also deep-water discharge into Great Bear Lake [7]. Lake disposal was also common in northern Saskatchewan in Canada: most of the tailings from the Gunnar mill were deposited into Mudford Lake about half of the Beaverlodge mill tailings went into Fookes and Marie Lakes [23]. Tailings were also discharged into lakes in some mines operating under the jurisdiction of the former Soviet Union [7].

This list of examples demonstrates that the natural features of the landscape in the vicinity of the mills determined how the tailings were placed. Steep valleys in mountainous areas, lakes in the glacial till-covered Canadian shield and northern Russia, mined-out pits in areas of previous mining activity, and opportune natural depressions were all obvious and convenient features. Where such features were not present, low bunds or ring dyke structures were sometimes constructed where the developers had some concern or forethought for possible health or environmental effects. However these were commonly inadequate in their design to provide service for more than the short term, and commonly failed. In other situations no provisions for containment were made at all, and the tailings were accessible to people and fauna as well as to dispersion through wind, surface water erosion, and seepage to groundwater.

2.5. Classification of uranium mill tailings

The volume of tailings piles varies from several hundred to several tens of millions of cubic metres, depending on the size, nature and duration of the operation. The activity of the tailings depends on the grade of ore mined and varies from less than 1 Bq/g to more than 100 Bq/g. The grain size distribution plays a major role in the physical stability, consolidation and hydraulic properties of the tailings. It is related to the nature of the host rock, the texture of the mineralization and the crushing and grinding processes applied in extraction. A classification based on various geotechnical and phenomenological criteria has been proposed [27] (Fig. 2).

The chemistry of tailings depends mainly on the leaching process (acid or alkaline) and the mineralogy of the ore. Wastes from the mining of sulphidic ores tend to produce acid leachate, which can mobilise radionuclides and other hazardous components of the tailings.

Trace element concentrations in the ore and process chemicals may have a major influence on the tailings chemistry and its total environmental impact.

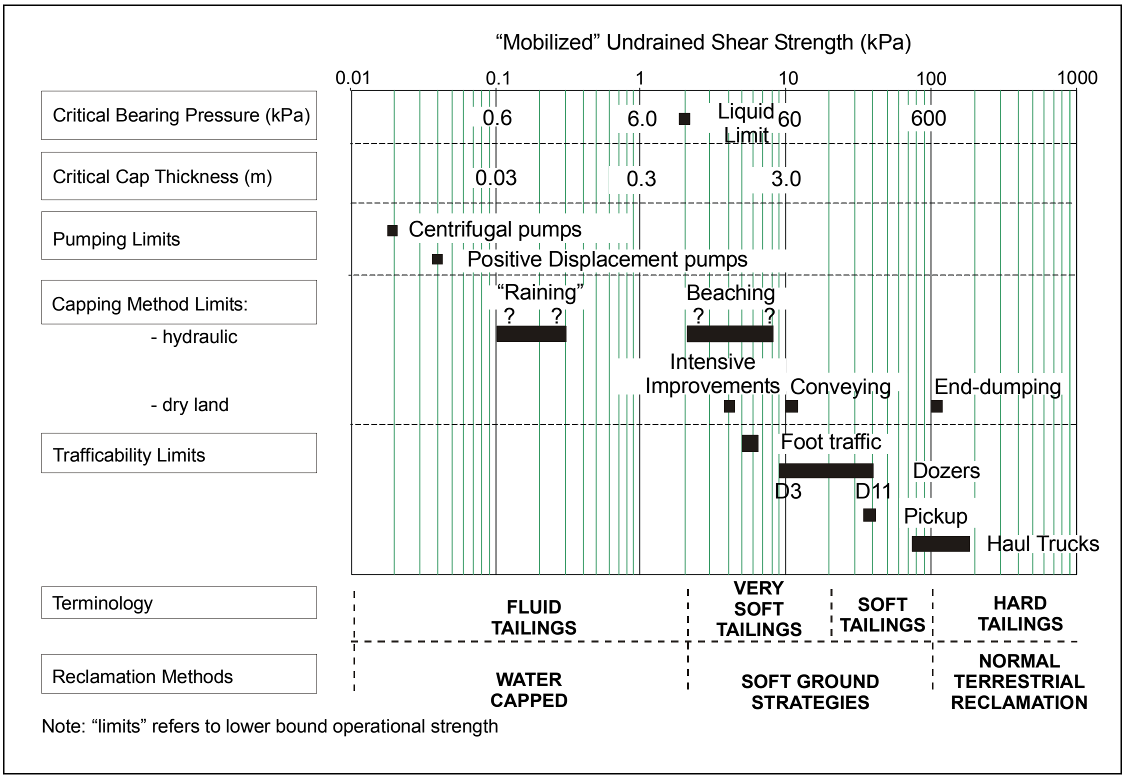

Fig. 2. Proposed classification of tailings for remediation purposes [27].

2.6. Inappropriate uses of uranium mill tailings

The coarse, sandy nature of some mill tailings makes them an attractive resource for re-use as a building material, mainly as sand for concrete making. Re-use was a feature in areas where tailings piles were in or close to urban centres and where controls on access and removal were low or absent. Perhaps the most extensive re-use was in Grand Junction, Colorado, where tailings were used as sand for concrete manufacture, mortar, backfill around foundations, and as fill under pavements and streets etc [18] [28]. Over 4050 properties contaminated by uranium tailings have been cleaned up in the 'UMTRA Vicinity Property Program', with over 1.65 million cubic metres of contaminated material removed from, around or beneath civic buildings, private homes, footpaths, drainage dykes, industrial sites, police stations, jail, sewer system and railway line beds [29].

Similar use of tailings was made in other uranium milling districts, including the Saxony and Thuringia areas of Germany [22] [30] Lower Silesia in Poland (see Annex X) and Port Hope in Ontario, Canada [25]. Tailings that flowed into the local river after failure of the containment structure in 1962 were recovered by the local people of Ak-Tuz in Kyrgyzstan and used as a building material [14].

3. ENVIRONMENTAL IMPACTS

3.1. Events that have led to environmental impacts

The types of events that have led to environmental impacts can be broadly categorised into chronic and acute failure [31]. Acute failures involve sudden physical failure of the containment structure, and are listed in Table 3.

Table 3. Examples of tailings (including uranium) dam failures, data from [32] [44] [45]

Date	Location	Parent company	Type of Incident	Release	Impacts
(1994)	Zirovski vrh, Slovenia	Rudnik Zirovski vrh, Gorenja vas	ongoing slippage of the slope (7 million t) with the "Borst" tailings deposit (600,000 t) on the top, at velocity of 0.3 m per year	*n/a*	*n/a*
1994, Feb. 22	Harmony, Merriespruit, South Africa	Harmony Gold Mines	Dam wall breach following heavy rain	600,000 m^3	tailings travelled 4 km downstream, 17 people killed, extensive damage to residential township
1994, Feb. 14	Olympic Dam, Roxby Downs, South Australia	WMC Ltd.	leakage of tailings dam during 2 years or more; for details see SEA-US, NIC	release of up to 5 million m^3 of contaminated water into subsoil	*n/a*
1979, Jul. 16	Church Rock, New Mexico, USA	United Nuclear	dam wall breach, due to differential foundation settlement	370,000 m^3 of radioactive water, 1,000 tonnes of contaminated sediment	Contamination of Rio Puerco sediments up to 110 km downstream
1979, Mar. 1	Union Carbide, Uravan, Colorado, USA	Union Carbide	two slope slides, due to snow smelt and internal seepage	*n/a*	*n/a*
1977, Feb. 1	Homestake, Milan, New Mexico, USA	Homestake Mining Company	dam failure, due to rupture of plugged slurry pipeline	30,000 m^3	no impacts outside the mine site
1977	Western Nuclear, Jeffrey City, Wyoming, USA	Western Nuclear	dam failure, due to melting of snow	40 m^3 of tailings and 2.3 million gallons of liquid	"no offsite contamination"
1976, Apr. 1	Kerr-McGee, Church Rock, New Mexico, USA	Kerr-McGee	dam failure, due to differential settlement of foundation soils	'minor quantity'	*n/a*
1971, Mar. 23	Western Nuclear, Jeffrey City, Wyoming, USA	Western Nuclear	dam failure, due to break in tailings discharge line	*n/a*	"no offsite contamination occurred"
1967, Jul. 2	Climax, Grand Junction, Colorado, USA	Climax	dam failure, due to unreported causes	12,000 m^3	Effluent release into an adjacent river
1963, Jun. 16	Riverton, Wyoming, USA	Susquehanna Western Inc.	The dam was intentionally breached and a 2-ft depth of effluent was released to prevent uncontrolled release of the impoundment contents during heavy rain	*n/a*	*n/a*
1962, Jun. 11	Mines Development, Edgemont, South Dakota, USA	?	dam failure, due to unreported causes	100 m^3	Tailings released reached a creek and some were carried 25 miles to a reservoir downstream
1961, Dec. 6	Union Carbide, Maybell, Colorado, USA	Union Carbide	dam failure from unreported causes	280 m^3	Effluent released did not reach any stream
1960	Gunnar mine, Beaverlodge area, Saskatchewan, Canada	Gunnar Mines Ltd.	dam failure	*n/a*	Tailings release into Lake Athabasca, creating Langley Bay tailings delta

Note: n/a – no information available

Causes for acute failure in uranium tailings dams are no different from those at other mill tailings dams. The causes, earthquake-induced instability and failure owing to cracking or liquefaction; physical weakness of the embankment leading to breaching; erosion from heavy rain or adjacent waterway leading to thinning of the wall or overtopping by decant water; cracking induced by settlement; piping; lateral instability (movement) of the wall caused by insufficient mass or lubrication by saturated weak foundations; slumping of material into the containment, causing overtopping and/or erosion of the wall or its physical destruction; and spillway collapse caused by heavy overflow of decant or slurry after severe rainfall [33].

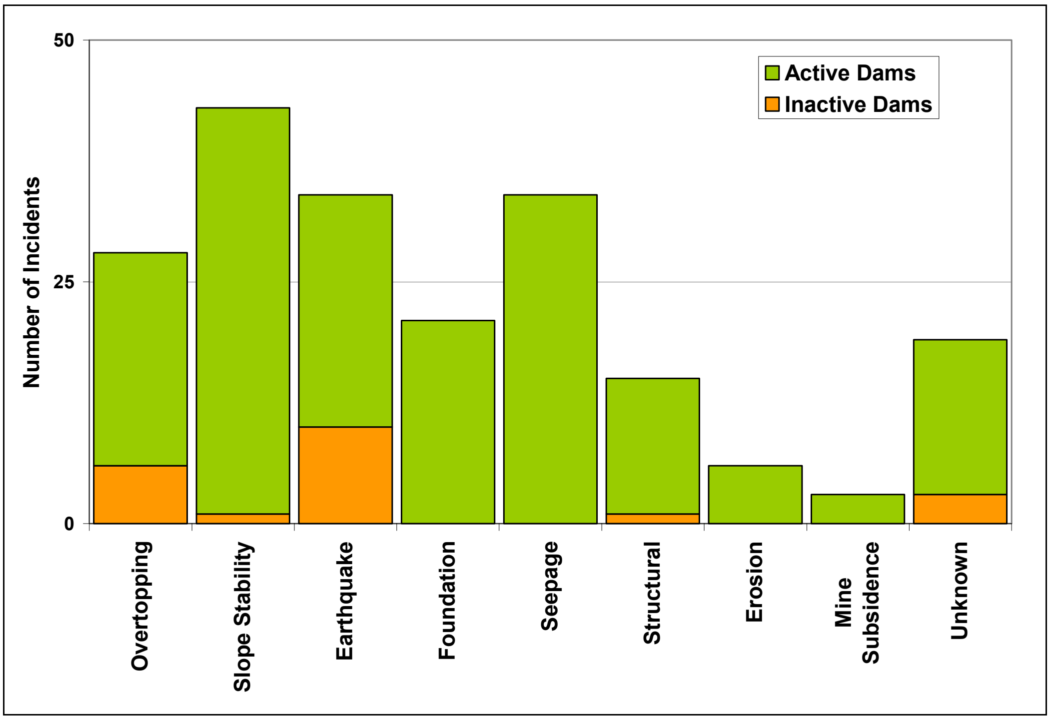

Fig.3. Tailings dam incidents (a total of 199) classified by cause of failure and active/inactive facility [34].

The various causes of tailings dam incidents (ie acute events, and chronic events indicating performance inconsistent with design at all tailings dams including non-uranium tailings facilities) are tabulated for 199 incidents in Fig.3. At inactive dams the main causes are earthquake and over-topping; for active dames the leading causes are slope instability and seepage, followed by earthquake, over-topping, foundation conditions (including mine subsidence), and structural [34]. About 90% of the documented incidents were associated with active impoundments where the dams contained surface water as decant liquor or flood water, indicating that tailings pore water pressures and water management practices encountered during operation of the facility are major risk factors, which become much less significant after closure (Fig.4). The data also suggest that dams built by the upstream construction technique (see Section 6.3) are prone to the most failures, but this also be due to the fact that the majority of tailings dams in the data set are of this type.

A review of the literature suggests that tailings containments have been subject to most of these types of failure, although the details of the cause of failure are rarely reported. Examples — not necessarily only uranium tailings — include Church Rock USA 1979 [36], Moline Australia between 1973 and 1980 [22] [37], Rum Jungle, Australia (various times) [22]; Schneckenstein in Germany [38], Merriespruit in South Africa (Fig.5) [39] [40]. Notably,

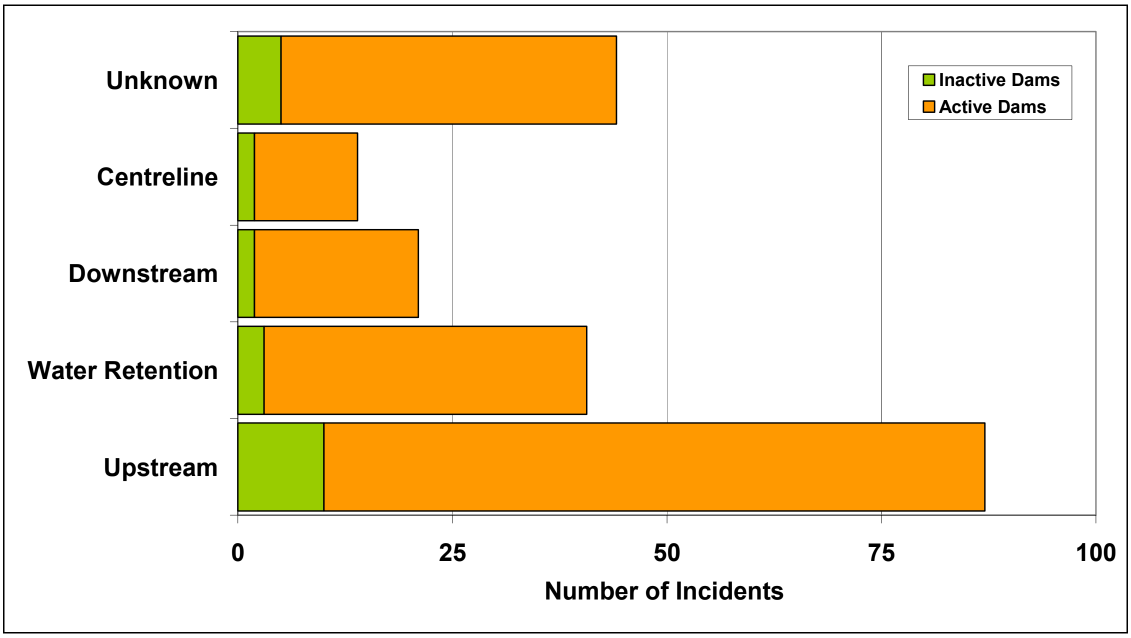

Fig.4. A selection of 199 tailings dams incidents classified by embankment construction type and as active/inactive facilities [34].

Fig.5. The Merriespruit (South Africa) tailings dam failure in 1994 [35].

whilst most of these dams and embankments were not purpose-designed or built, existing simply of waste rock or local soil or even tailings being roughly bulldozed to form a bank, the Church Rock containment was hailed in its day as the most carefully engineered tailings dam in the State of New Mexico. However, engineering assessment after the failure pointed to multiple failures in design, construction, and management. Differential settling was caused by construction over inhomogeneous foundations of solid bedrock and weak soils and cracking was visible for two years before the actual failure. Furthermore, the drain zone had not been constructed according the approved plans, no tailings beach had been formed against the wall as recommended, and the level of water was 0.6m higher than design limits at the time of failure [36].

There are many forms of chronic failure. These include:

- Dispersal of radioactive dust: tailings surfaces allowed to dry out, followed by wind-blown distribution of dust. This is reported as a problem in almost every area of historic tailings storage, e.g. Durango in Colorado [22], Wismut operations Germany [30], West Mecsek Hungary [41], Bukhovo Bulgaria [36], Kyrgyzstan [14], Rum Jungle Australia [22], Ulba, Kazakhstan (see Annex VII);
- Erosion of tailings from outer surfaces of the containment, e.g. Rum Jungle Australia [22], Wismut operations Germany [30];
- Seepage through the floor and/or walls of the containment – many early containments were constructed with no seal or seepage collection system in the base (e.g. Mecsek in Hungary [41], Schneckenstein Germany [38], Kyrgyzstan [30]. On some of the Far West Rand mines in South Africa, tailings dams were sited on karst features in dolomite to promote drainage. At Anhua in northern China seepage from the tailings dam resulted in significant uranium and cadmium contamination of a lake used for agricultural irrigation [42]. In Estonia seepage from the tailings pile near the coastline has resulted in measurable contamination in the Baltic Sea up to 300m from shore [36];
- Effluent discharge as decant allowed to drain or overflow from the containment directly or indirectly into natural waterways, for example at Jaduguda in India [22] [26] and several Wismut operations in Germany [38]. At Elliot Lake in Canada, effluent discharge resulted in significant radiological contamination and acidification of the 300 km^2 Quirke Lake [24].

Both acute and chronic events are commonly associated with non-radiological impacts related to heavy metals and other toxic compounds in the tailings, pore water, and decant water. Of particular concern is the generation of acid conditions from tailings derived from sulfidic ores. In the past un-neutralised tailings with acid-producing potential were deposited. These tend to exacerbate the probability of contamination because of the solubility (and hence greater mobility) of many metals under acidic conditions. It may also affect the bioavailability of toxic metals and compounds [43]. The neutralization of tailings prior to deposition, when required, has become common practice in more recent times. Therefore the level of impact caused by tailings and tailings waters released through containment failure may vary significantly due to the chemical nature of the material as well as the sensitivity and condition of the receiving environment.

3.2. Impacts on human health

Sudden failure of any large structure may cause death through drowning, crushing or suffocation, and the larger uranium mill tailings containments are the same as tailings from other types of mines or from water containment dams in that regard. Landslides involving precariously placed uranium mill tailings at Mayluu-Suu in Kyrgyzstan over a 30 year period, killed people and destroyed buildings [14]. However, the failure of uranium tailings containments does not feature highly on the list of dam failures to have caused death or major destruction [32] [46], perhaps because dams at uranium mines are a small proportion compared to the total number of mine dams. They are also are generally smaller than the very large dams at many large scale. Low tonnage, gold and base metal mines developed around the world over the last 20 years.

Although other types of mill tailings may also contain radionuclides, the particular human health risk that is associated with uranium mill tailings is the risk from radioactivity. In turn, uranium mill tailings may also pose risks associated with their inventory of heavy metals and other chemical elements, such as arsenic. It is not possible to determine the actual risk or the level of harm caused by tailings distinct from other causes, because people affected by radiation in uranium mining districts are potentially exposed to radiation doses from mining, milling, transport of radioactive materials, radioactive dust and contaminated water and foodstuffs, at it is not practicable to distinguish between this mixture of point and diffuse sources.

Tailings in themselves may pose a significant proportion of the health hazard because of the manner in which they are commonly disposed of. The disposal at the surface over relatively large areas allows significant flux of radioactive gasses and interaction with surface water. In many historic uranium mining areas both have constituted a pathway from the immediate surrounds of the mine site and impacted upon the general public. However, the information on the relative and absolute impact can be unclear or confusing – for example, an analysis of the downstream impacts from the Church Rock tailings spill (New Mexico USA) in 1979 concluded that "there were no demonstrable health effects" and that groundwater contamination was no greater that pre-spill conditions [47]. However, it is probable that the 'pre-spill conditions' against which contaminated levels were compared were themselves elevated as a consequence of long term seepage from the mining operations [32], indicating that long term assessment of effects may be poorly founded.

The long half-life of radiation from uranium tailings and the demonstrated risks association with them have given rise to levels of high concern in the general public and in government – in some places exacerbated by the high levels of secrecy and lack of data on health impacts such as in the Saxony-Anhalt/Saxony/Thuringia areas of Germany [6]. Here, people in the 'immediate vicinity' of (unremediated) tailings from the Wismut operation were subject to additional doses of 1-2 mSv/year and in special cases up to 6 mSv/year [15].

Perhaps the most direct implication of tailings as a radiation source sufficient to cause human health impacts relates to the re-use of tailings for building materials. Whilst this has happened in many places around the world (see Section 2.6), it is best documented in Grand Junction, Colorado, in the Unites States of America. It became known in the mid-1970s that leukaemia rates in the county including Grand Junction city were twice the average for the State. This fact, and the concern it caused, was the driver for the development of state legislation for radiation control and protection measures, which led to the development of the Uranium Mill Tailings Remediation Control Act 1978 (UMTRCA Act) [18].

The body of research which indicated that direct impacts on human health were resulting from exposure to uranium mill tailings gradually grew, with the realization that radon concentration in houses built with mortar sand derived from uranium tailings, or over tailings-derived fill etc. could reach dangerous levels (e.g. 200 000 Bq/m^3 at Schneeberg in Germany [30] [36], radon and dust derived from tailings were calculated to result in between 0.3 and 1.0 death per 2600 residents of Eleshnitza in Bulgaria, and radium concentrations were found in cereals being grown in tailings-contaminated soils [48].

These concerns have resulted in a large body of research being undertaken on the mechanisms of transfer of radiological dose to humans via a variety of pathways, and the development of models to determine biological as well as non-biological pathways (see for example BIOMOVS [49]). Uranium mill tailings are one of a number of potential sources for uptake, e.g. [50].

An evaluation of the completed US American UMTRA programme calculated that 1289 lives will be saved over 100 years (using a linear model with no threshold values) as a consequence of removal of uranium mill tailings and placement in purpose-designed containment structures removed from populated areas [51]. The UMTRA programme serves to demonstrate that uranium mill tailings were identified in the USA as a significant health hazard. Whilst this assessment may overstate the numbers of lives saved because of the methodology used for the calculations, it demonstrates that the risk is sufficient to warrant major expenditure to clean up old sites where tailings have been disposed of inappropriately, and to develop regulations to ensure that similar hazards do not eventuate from modern uranium mining and milling activities.

3.3. Potential radiological impacts upon the natural environment

There are many articles in the literature that purport to describe impacts on the natural environment from uranium mining in general, and the placement/disposal and management/mismanagement of mill tailings in particular. However, the description of environmental impact that most of these articles present is limited to a qualitative description of impact in terms of contaminated soil, surface waters, groundwaters, lake sediments etc (e.g. [14] [15] [33] [36] [37] [42] [52]), any quantitative description is usually given in terms of the area/volumes affected and concentrations of radioactive elements (e.g. [14] [15] [36]), or activity levels and doses (e.g. [17] [53]. A body of literature exists on the contamination of environmental media directly linked to consumption and therefore potential uptake of radionuclides by man: principally soils (e.g. [14] [53]), and water, e.g. [54]. In contrast, there are relatively few articles that set out to quantify the level of harm to the biosphere caused by uranium mill tailings.

Research into the biological impacts from the nuclear fuel cycle is a relatively new science, and is strongly biased towards research on the risks, hazards and impacts to humans from the fuel fabrication, power generation and fuel reprocessing/disposal parts of the cycle. However, interest in research into the biological impacts from the uranium production part of the nuclear fuel cycle has grown from:

— A realization that fisheries in the vicinity of some uranium mines were deteriorating, e.g. [24] [25] [55] [56];
— A realization that radionuclides could be taken up by plants, thereby posing a potential risk to vegetation as well as animals higher in the food chain including humans, e.g. [14] [57] [58];
— A realization that radionuclides could be taken up by animals, thereby posing a potential risk to the health of individuals, communities and ecosystems as well as animals higher in the food chain including humans, e.g. [47] [53] [59];
— Documentation on the concentration of radionuclides in bottom sediments and their long term availability to bottom feeders and potential for multiple cycling through the aquatic food chain over the long term, e.g. [60];
— Development of transfer pathway models for the transfer of radiological dose through the food chain, e.g. [49];
— Concern that tangible impacts on human health from radioactive contamination associated with uranium mines and tailings piles may also be having significant impacts on the environment [11?]; and

— Questions over the validity of ICRP's 1977 statement [61] that:
"although the principal objective of radiation protection is the achievement and maintenance of appropriately safe conditions for activities involving human exposure, the level of safety required for the protection of all human individuals is thought likely to be adequate to protect other species. The Commission believes that if man is adequately protected then other living things are also likely to be sufficiently protected".

At the time the available evidence suggested that humans were indeed the most radio-sensitive species. Some recent discussion includes suggestions that significant evidence of environmental damage from 'controlled practices' is hard to find [62], and that concerns over impacts from uranium mining, particularly in remote areas such as in Canada, have been over-stated [63]. However, since the late 1970s there has been a general increase in concern for the environment, presumably due to evidence that the actions of humans are causing visible and significant environmental changes, for example the effects of gaseous effluents on sensitive ecosystems through acid rain and on the ozone layer and the pollution of rivers as a result of pesticide application to farm land. Furthermore, close attention s now being paid to the larger implications of harm being caused to elements of particular ecosystems with consequent impacts on biodiversity. It is perhaps for these reasons that the issue of environmental protection in the context of ionising radiation is being addressed in many countries [62]. A recent IAEA review of practices for close-out of uranium mines and mills observed that:

> *"In recent years more attention has been directed to the impacts that the nuclear fuel cycle may have on the environment. When environmental media (soils, surface and ground waters, and air) have increased contaminant levels, biota may also take up and accumulate contaminants. Such uptake may pose a threat to ecosystem vitality in the shorter term and to the biological community structure in the longer term. As humans ultimately depend on the environmental media of soil, water and air for survival, and also occupy the top of the food chain, there is an inescapable link between human health and safety, and the health of the environment"* [31].

This concern is demonstrated through a series of conferences addressing 'Protection of the natural environment from ionising radiation' (Stockholm, May 1996; Ottawa, May 1999; Darwin, July 2002). Several papers in these conferences document changes in biota directly attributable to ionising radiation, e.g. lower fecundity, more genetic diversity and DNA strand breaks in mosquito fish in ponds near the Oak Ridge National Laboratory in Tennessee USA [64]. Whilst most papers are related to effects of ionising radiation from nuclear power plants etc., some papers are reporting investigations into areas contaminated by uranium mining and milling. In that, meaningful comparisons can be made with data such as those prepared by the US National Council on Radiation Protection and Measurements on the effects of ionising radiation on aquatic organisms, which relate to dose levels mostly much higher than those encountered in situations at mine, mill and tailings pile sites [65]. Notably, one paper presented at the Darwin conference in July 2002 assesses the impact of radionuclide releases from Canadian nuclear facilities on non-human biota. It concludes that whilst releases from uranium refineries and conversion facilities, power reactors and associated waste management facilities and research reactors are not entering the environment in concentrations likely to have a harmful effect, the same is not true for releases of radionuclides from uranium mines and mills and waste management areas. Releases from mines, mills and waste facilities are assessed as to be in sufficient quantities or concentrations or conditions that have or may have a harmful effect on the environment [66].

3.4. Toxic and hazardous compounds in tailings and their potential impacts

The impacts on the environment from uranium mill tailings are not all related to the radionuclide content alone. The presence of other contaminants can exacerbate the availability of the radionuclides to the environment, and in some cases the other contaminants have direct harmful effects in themselves. In this regard, the potential for uranium mill tailings to cause environmental harm is little different from other forms of mill tailings, and the resultant impacts may be very similar. Indeed it is not adequate to consider the radiological impacts from uranium mill tailings that relate only to the effects of ionising radiation.

The other effects may include:

- the chemical toxicity of the radionuclides, including uranium;
- the chemical toxicity of heavy metals and metallic compounds;
- the chemical toxicity of non-metallic minerals and compounds in the ore or introduced during processing (e.g. sulfuric acid, kerosene);
- acidity, resulting from sulfidic (ore) minerals or acid introduced during milling;
- turbidity;
- salinity.

The types of non-radiological contaminants that may cause harm are dependent of the nature of the mineralization in the ore body, including the gangue mineralogy, and the processing technique used in the mill; choice between acid and alkali leach, and whether to neutralise the tailings prior to deposition, has marked effects on the levels of contaminant concentrations in the slurry. It also affects the bioavailability of many compounds.

Some historic impacts have been improperly ascribed to uranium, such as at Rum Jungle in Australia, where 15 km of river and riparian ecosystem downstream of the mine and tailings pile was killed following discharges of copper-rich acidic drainage; effluent from the tailings is estimated to have contributed less than 5% of the total copper load, and uranium is not considered to contributed to the impact to any significant degree [7][22]. Arsenic is a significant constituent of Schlema-type ores in the Ronneburg district of Germany, and contributes significantly to environmental impacts, particularly ground water quality [15]. Cadmium has contaminated farmland and polluted a lake critical for agricultural irrigation near the Xiushui mine in China [42].

Elevated acidity plays a major role in increasing the mobility of metals in aqueous solution, including uranium as well as copper, arsenic, cadmium and other metals. Impacts on vegetation in the Uranium City district of northern Saskatchewan are a result of tailings salinity, acidity, and low fertility [25]. Acid drainage has led to serious contamination of ground water systems in several countries, notably Germany [15] and USA [54] (note that severe ground water contamination in the Czech Republic relates to ISL mining, and no mill tailings are implicated).

As reported in a recent review on mine and mill close-out procedures by the IAEA:

> *"Acid drainage ... can be a prime environmental concern. Low pH waters may dissolve minerals containing radioactive elements and heavy metals. Acidity combined with deposited salts and heavy metals can prevent plant growth. In addition, downstream surface water bodies may become contaminated and affect the health of the ecosystem. Stream banks may become destabilised as the vigour of vegetation supporting the banks*

is decreased. Dissolved uranium and daughter products can reach levels in water leaching from tailings and uranium waste rock piles which pose a direct chemical toxicity threat to ecosystems." [31].

The potential for impacts from non-radiogenic contaminants has been investigated in modern Environmental Impact Studies for uranium mine development proposals. Perhaps the first was for the Ranger mine in Australia [10], where loads, concentrations and possible effects of base metals and sulfates on the environment were assessed prior to approval for mine development.

It is interesting to note that the same principle espoused by ICRP concerning man as a suitable indicator species for environmental health does not apply to non-radiological contaminants, because in the aquatic environment some elements are more toxic to aquatic organisms than to man [43]. The effects of heavy metal pollution are complex and strongly dependent on local geographic and climatic factors, the mix of chemical constituents present, and on the nature of the affected organisms. Effects can be lethal or non-lethal. Chronic sub-lethal poisoning can affect growth, reproduction, behavioural patterns, and result in lower resistance to disease, as well as causing the organism to have body concentrations of elements above standards permitted for consumption. In some instances the non-radiological hazards have a much larger effect than the radiological pollutants [53]. Non-radiological components are reported as more limiting on operations at the Ranger mine in Australia than for the radiological contaminants [67].

4. REMEDIATION PROGRAMMES TO REDUCE HAZARDS FROM HISTORICAL URANIUM MILL TAILINGS

4.1. Driving forces for remedial work

The fundamental reason to remediate tailings piles is to reduce the level of hazards and thus lessen the risk of significant impacts upon human health and the environment, or to reduce ongoing levels of impact where effects are already apparent. Whilst the objective may be altruistic, that is the work is undertaken because of a concern for human and environmental well-being, the objective typically is also based upon concerns about the potential cost of not undertaking remedial works. These costs may be in the form of:

— Compensation payments, where a case is made that inadequate or improper measures were taken to safeguard people or the environment;
— Lost productivity from contaminated and quarantined land or fisheries;
— Development costs for constructing water wells or sophisticated water treatment plant to replace water previously supplied from now contaminated surface and ground water resources;
— The larger financial consequences from greater areas of contaminated land and water resources and continued contamination or air sheds should the remedial work not be undertaken.

Regulatory and legal frameworks pose significant potential (financial) risk to organizations holding liabilities in the form of current or past mine, mill and waste storage facilities. Increased knowledge of the harm caused by and the potential risks associated with such facilities, more open arrangements to access information and to challenge responsible agencies, greater accountability required of government and private enterprises, and increased public concern, combine to provide powerful incentives for reducing current and potential future liabilities through design and execution of remediation works.

4.2. Remedial work undertaken

The incentive for remedial work to be undertaken by the private sector is to comply with regulatory requirements. Examples of remediation of historic sites by the private sector are limited. Many of the mines concerned, which began operations in the 1940-1950s, were operated by government agencies or were subject to tight government control through to their closure. A few continued operating through to the 1970s and later, when regulations requiring remediation of tailings piles were in place in some Western countries. Therefore upon closure the private operator was compelled to conduct remedial works consistent with the regulations of the day. The number of private sector entities, where liabilities to remediate historic uranium mill sites could be identified, was however further reduced owing to many going out of business during the years of low uranium prices in the 1980s. Examples of remediation of historic sites by the private sector include the Beaverlodge area in Saskatchewan, and Mary Kathleen in Australia.

The Eldorado Beaverlodge operation was shut down in 1982 after 29 years. Decommissioning objectives were set by a panel of regulators in radiation protection, safety and environment. Physical decommissioning at Beaverlodge, which included a number of satellite mines, was done in three years. Monitoring continues to this day and is undertaken by government agencies. When stable conditions are evident, the company will apply to relinquish the leases and turn the property over to the government [63].

At the Mary Kathleen mine in Queensland Australia, mining and milling took place in two periods between 1954 and 1982. By the time operations ceased, regulations requiring remediation of the tailings pile had come into force, and the operator was required to design and complete remedial works consistent with these requirements [68].

Several governments around the world in the last twenty years have implemented major remedial works. The timing and type of programme has been influenced by several different factors. These include access to information on health and environmental effects, changes in government agencies' attitudes towards making information available to the public, access to sufficient funds, and the type and extent of operations, and their impacts on the environment. Funding is not necessecarily an impediment in Western countries, provided that the problems translate into political debate and pressure. However, some other countries have been unable to implement remediation owing to lack of funding or political willingness. The breakdown of the Soviet Union made public discussion on impacts from uranium mining and milling possible for the first time in many countries. German unification led to sufficient funding for dealing with the impacts from uranium mining and milling in the former GDR. Plans for extension of the European Union to include Central and Eastern European (CEE) countries led to development of a funding scheme for remediation planning and execution in those countries under the PHARE programme [69]. Similar assisstance is being given be the EU to former Soviet Republics under the TACIS (Technical Assisstance to the Commonwealth of Indipendent States) programme. Some examples of government remediation programmes are given in the following subsection.

4.3. Examples from the USA

A suggested link between leukaemia rates and uranium mill tailings in Grand Junction, Colorado was the driver for development of legislation for the Uranium Mill Tailings Remediation Programme (UMTRA) [18]. The UMTRA programme in the USA focussed specifically on uranium tailings piles accumulated at 22 sites mainly in the central western states [70]. In many places tailings had been placed in unbounded or poorly bounded piles on level to gently sloping ground. Several piles were within city limits and along or close to river banks. Dust dispersion of the tailings, and re-use by the community as construction sand and fill, gave rise to significant health risks. The programme resulted in tailings being moved from the remnant piles, and reclaimed from thousands of urban sites, for relocation in purpose-designed and built repositories. The repositories generally consisted of lined storage cells with rock-armoured embankments designed to last at least 1000 years. Capping to minimise the risk of exposure of the tailings and reduce radon flux was placed on top. The containment designs were based on extensive research funded for the purpose by the US Government.

4.4. Examples from France

Milling residues were stored at 22 locations across France, and their remediation is part of a programme to rehabilitate mines, mills and waste storages in accordance with regulations under the Mining Code, Registered Facilities for Environmental Protection, and general regulations dealing with water, air, water, noise and landscape protection [71]. Except for the Forez site (where residues are stored under water), the tailings are above ground level, covered with clean waste rock from the site to provide mechanical protection, resist erosion and intrusion, and to reduce radiological flux and exposure. Drainage works are included, to minimise the volume and concentration of contaminated surface water.

4.5. Examples from Germany

Remediation of the WISMUT sites in the states of Saxony and Thuringia entails a range of stabilization measures, such as mine flooding, reshaping and covering of waste rock dumps, pit backfilling and *in situ* consolidation of the tailings ponds [72]. Owing to the climate, intensive land use and regulatory conditions, the water pathway is most important in the evaluation of remediation options. The goal is not to stop all releases of contaminants, but to reduce loads to levels that fall within the natural assimilative capacity of the surrounding environment.

4.6. Examples form the Czech Republic

Uranium mining and milling are controlled by the state in the Czech Republic. Due to declining needs, increases in production cost, and low world market prices a step-wise reduction of uranium production was decided at the end of the 1980s. The state-owned enterprise DIAMO is now responsible for a large scale closure programme, including technical, social, and environmental aspects. The first decommissioning and remediation works of exhausted mines were undertaken in the 1950s, but these works are sometimes considered inadequate, resulting in 'legacy sites'. As part of the policy of continuing improvement of the environment, the government is funding the remediation of such 'legacy sites' [206]. Between 1993 and 1997 an inventory of these 'legacy sites' was drawn up. A respective remediation project was initiated in 1998, and by its completion in 2007 is estimated to have cost 232.3 M CZK (8.3 M USD). Overall, the DIAMO remedial activities concern conventional as well as *in situ* leaching sites, tailings ponds, and long term water treatment facilities. Environmental activities will have to continue until about 2040 and are estimated to cost above 60.0 bill. CZK (2.2 bill. USD). The predominant part (> 90%) of these activities is being funded through the state budget of the Czech Republic.

4.7. Examples from Australia

The Rum Jungle mining area was operated by a private company under contract to the government and remained unrehabilitated after milling ceased in 1972. A small clean up programme in 1977 dealt mainly with aesthetic matters, but failed to prevent occasional breaches of the dam and continued releases of tailings and liquor to the Finniss River. The government funded a remediation programme aimed at reducing hazards in 1985–86, in which tailings and contaminated soil were placed in one of the open pits without any preparation. The tailings were covered with a geomembrane and a 1 m rock blanket, then alternating layers of contaminated subsoil and copper heap leach material. The area was then covered with topsoil and revegetated, and drainage diversions installed [22]. Hazard reduction works were also undertaken in the South Alligator region, where tailings were removed for reprocessing for gold, and the tailings pad ripped and revegetated.

Concerns over the continued hazards posed in these areas — mainly from open pits and waste rock piles, and requests from indigenous land owners for improved access and use — has lead to funds being provided for studies into performance of the existing remedial works and options for improvement. The tailings areas may be further remediated as part of this programme.

4.8. Evaluation of the remedial works at historic tailings piles

Many remediation programmes of historical tailings piles are not yet complete, or have not been completed long enough to enable meaningful assessments of their effectiveness to be made. A few qualitative assessments are available for some sites, and high level assessments of cost and effectiveness (in terms of lives saved) have been made.

At ***Rum Jungle*** in Australia, the post 1982–86 remediation programme monitoring regime indicates that aquatic life has returned to the Finniss River in the 15 km stretch downstream of the tailings pile, which had been killed as a result of copper and uranium contamination. Results from the groundwater monitoring programme suggest that it will take another 15 years before contaminant concentrations leaving the waste heaps drop significantly [7]. However, the level of environmental protection afforded generally by the 1980s remediation works is considered inadequate by today’s standards, and funding has recently been provided by government for planning further remediation work. One aspect requiring attention will be landform design and capping design to reduce the impacts of natural erosive and vegetation processes. The present design requires annual removal of vegetation to avoid capping degradation through root penetration. Clearly this high-maintenance approach is not consistent with objectives for long term stability and reliable stewardship.

Monitoring at ***Port Pirie*** in South Australia, where tailings were covered by a 1.5 m layer of zinc slag, indicates that the level of dust generation has been reduced very significantly, and that radon emanation has dropped to below limits recommended by the USEPA for rehabilitated uranium tailings piles [7].

Design of containments built to accommodate tailings relocated by the ***UMTRA Programme*** in USA was initially based on steady state equilibrium conditions with covers constructed to limit infiltration into the pile to no greater quantity than seepage rates out of it. However, revised water quality standards required more emphasis on limiting infiltration in order to avoid or minimise groundwater contamination. Other considerations requiring revision of early design concepts and standards were redesign of covers that beneficially exploit ecological changes on and in the cover over time, instead of concentrating solely on physical design parameters that are liable to degrade over time as a consequence of natural erosional, climatic and vegetative processes (see Annex XI).

Attempts have been made to evaluate the effectiveness of remedial work done to date in terms of expected environmental impacts and conditions arising from closed out uranium mine and mill facilities by relative cost and calculated lives saved. An attempt in 1997 to collate such information for the medium to long term was of very limited success [31]: only eight countries responded to this survey question. The information provided was quite different from country to country, and no comparisons or overall conclusions were possible.

Relative costs of remediation of mines, mills and ancillary facilities including tailings piles are reported for individual countries or remedial programmes in various articles, and information has been recently tabulated for a wide range of types of mine and mill and ancillary facilities around the world (Tables 2,3,4 in [7]). The tabulated information does not discriminate costs of tailings remediation from other operational components, but it does indicate which remedial programmes included tailings piles as part of the works conducted. These comprise sites in eight countries (Australia, Canada, Czech Republic, France, Germany, Spain, Sweden, and the USA). The discussion points out that a comparison based on aggregated cost figures would not be objective, because the scope and extent of works will vary considerably for several reasons, even for similar sites and projects. Observations made on the data compilation for mines and mills are equally valid for tailings piles. The costs vary greatly as a factor of the amount of planning and preparation done for remediation and closure during the operational phase, local climate, condition of the tailings pile, degree and area of contamination, population density in the area, land use, labour costs, standards and objectives for remediation, etc. The single undisputable conclusion is that the costs of remediation are very significant, and place a significant budgetary burden on governments.

For this reason an assessment of the effectiveness of remediation in terms of benefits to society is warranted. A good example is the assessment of lives saved as a result of reduced public doses related to removal of tailings in the UMTRA programme [51] [73]. Whilst the basis for calculation may be subject to discussion, this sort of analysis presents information of a type readily understood by government officials and decision makers.

5. PRESENT DAY PRACTICES

5.1. Objectives

Practices for the placement and disposal of uranium mill tailings have evolved to where they are today because of several factors:

- An appreciation of the extent of the impacts caused by historic practices and their costs in human safety, social, environmental and budgetary terms;
- The large body of information generated from monitoring and research on the types of impacts and how they occur;
- Better understanding of environmental systems and the sensitivity at the ecosystem and individual level;

- Development of regulations and standards prescribing limits on the types, volumes and concentrations of contaminants;
- Development of guidelines on design, construction, management and performance of tailings dams etc.;
- Increasing public concern on the safety of tailings impoundments and improved organization by NGOs on the topic, as well as increasing levels of expertise available to them;
- The efforts by mining companies, in most countries, towards best practice, regular and more open environmental reporting, and a sense of environmental responsibility as a response to expectations from all stakeholders of improved accountability.
- The need to include conceptual decommissioning plans in environmental impact studies and mine plans.

Adoption of new practices, and achieving conformity with tighter performance requirements, is much easier for new operations. Conversely, achievement of these new requirements is much harder to 'retrofit' to older mines. Remediation of an old mine, where no provision had been made for appropriate stabilization of tailings, will be difficult. The task is likely to be more costly, to take more time, and to result in a poorer outcome than if appropriate planning had been done at the outset.

5.2. Standards for radiological and environmental protection

Standards for radiation protection relating to uranium tailings impoundments effectively interpret ICRP recommendations for radiation safety of humans. Long term exposure to low levels of radiation and possible synergistic effects of radiation dose and exposure to other agents or stresses are issues of discussion. This latter issue is of particular relevance at uranium tailings facilities, where effluents comprise a cocktail of metallic and other toxic compounds in addition to radionuclides.

The IAEA has recently issued guidance for the safe management of radioactive waste from mining and milling of ores [121] and for the monitoring of such residues [203].

The variability between mine and mill types and the processes used, size and type of tailings facilities, climate, and the character, quality and sensitivity of the receiving environment strongly argue that methods to improve environmental protection would need to be chosen on site specific grounds. It is also evident that regulations and standards for environmental protection will not themselves prevent further environmental impacts from occurring. Developing an adequate protection of the environment involves:

- an awareness of the character and sensitivities of the environment to be protected;
- site specific analyses to determine a design appropriate to the location;
- adequate planning and design of the tailings facility to avoid events likely to lead to environmental impacts;
- assurance that the facilities are constructed and managed consistent with approved requirements (QA/QC);
- monitoring and analysis of the tailings facility's operating system, management performance, contingency preparedness, discharges, and environmental effects;
- continuous research into operational effectiveness and closure planning.

In many Member States standards for environmental protection have already been in place for several decades. These typically concern aqueous or gaseous contaminant releases from facilities such as mines or mills into the environment and the engineering stability of tailings impoundments. A license for controlled discharges of (treated) effluents and drainage waters may also be required. Such licenses may take into account maximum concentration levels, total mass/activity of contaminant discharged, as well as the total load on the surface water stream. The latter takes into consideration other dischargers within a catchment. Re-injection of effluents or drainage waters into groundwater aquifers is prohibited in various Member States. In some countries, legislation and regulation regarding radiation protection and environmental protection have been separated, sometimes leading to confusing and contradictory regulatory regimes. Where such problems have been identified, it is hoped that efforts are made to harmonise regulations and avoid contradition and confusion.

5.3. Member States' regulations, standards and guidelines

In many Member States, no new uranium mine could commence operations without a comprehensive tailings disposal plan having first been designed to conform to international standards and agreed to by the relevant authorities. Previous bad practices and growing public concern have led to the introduction of regulations in all jurisdictions, where uranium mining and milling is undertaken.

The USA has perhaps the most extensively documented procedures for dealing with uranium mill tailings. Many of the regulatory procedures around the world draw heavily from the American codes and regulations, and so they will be examined here. It should be noted, however, that these regulations are rather prescriptive in nature. Depending on the circumstance, performance or risk based regulations might be more suitable.

Deposition and disposal of tailings from uranium mills is governed by the provisions of the US Code of Federal Regulations (CFR), specifically chapters 192 and 264 of 40 CFR [3], the US Nuclear Regulatory Commission (10 CFR) [74], and local State mining ordinances and laws. The regulations focus on reducing emanations of radon from the tailings, preventing the spread of tailings through erosion of the containment structure, reducing contamination by seepage, and setting out requirements for thorough assessment of risk to the public and the environment including the risk of ground water pollution. Code 40 CFR [3] stipulates

- "a design for 1000 year disposal life, where reasonably achievable and minimally for 200 years. Design considerations cannot rely on active maintenance to ensure disposal integrity;
- the use of natural materials to construct the disposal cover;
- that the concentration of ^{226}Ra in land averaged over any area of 100 m^2 shall not exceed the background level by more than 5 pCi/g, averaged over the first 15 cm of soil below the surface, and 15 pCi/g, averaged over layers of soil 15 cm thick at a depth exceeding 15 cm below the surface;
- that in any occupied or habitable building, the objective of remedial action shall be, and a reasonable effort shall be made to achieve, an annual average (or equivalent) radon decay product concentration (including background) not to exceed 0.02 working levels. At all times, the radon decay product concentration (including background) shall not exceed 0.03 working levels, and the level of gamma radiation shall not exceed the background level by more than 20 μR/h.
- that the cover be designed to perform under steady-state conditions such that moisture infiltration is limited to a rate, or flux, that will not allow long term seepage to migrate out of the disposal cell into groundwater and exceed the established groundwater

standards for constituents common to uranium mill tailings at a given point of compliance".

The application of supplemental standards are allowed under Code 40 CFR in site specific cases, where the prescriptive standards would be too strict and result in unreasonable costs. Conditions for application of supplemental standards can be summarized as follows [3]:

(1) When r*emedial actions would pose a clear and present risk of injury to workers or to members of the public, not withstanding reasonable measures to avoid or reduce risk.*
(2) When the remedial action would, notwithstanding reasonable measures to limit damage, directly produce environmental harm that is clearly excessive compared to the health benefits to persons living on or near the site, now or in the future.
(3) When the estimated cost of remedial action at a vicinity property site is unreasonably high relative to the long term benefits, and the residual radioactive materials (tailings and other radioactively contaminated material) do not pose a clear present or future hazard. (The likelihood that buildings will be erected or that people will spend long periods of time at such a vicinity property should be considered in evaluating this hazard.) Examples are residual radioactive materials under hard surfaces, such as public roads and sidewalks, around public sewer lines, or in fence post foundations.
(4) When the cost of a remedial action for cleanup of a building is clearly unreasonably high relative to the benefits. Factors that should be included in this judgement are the anticipated period of occupancy, the incremental radiation level that would be affected by the remedial action, the residual useful lifetime of the building, the potential for future construction at the site, and the applicability of less costly remedial methods that remove residual radioactive materials.
(5) When there is no known remedial action.

Other jurisdictions are less prescriptive. Guidelines provided to proponents indicate that design, systems, and performance must reflect the particular characteristics of the site, and currently accepted 'best practice'.

Many guidelines have been promulgated for the design and construction of tailings dams by the professional geotechnical community (e.g. [75] [76] [77] [78] [79] [80]) addressing design, construction, operation, remediation of tailings dams and transport, placement and decantation of tailings. Guidelines have been developed in several countries; examples include:

— ***Canada*** – Mining Association of Canada has produced "A guide to the management of tailings facilities" in three languages – English French and Spanish [81]; the Canadian Dam Association produces updates to safety guidelines including information specific to tailings dams;
— ***Australia*** – a series of booklets produced by Environment Australia on Best Practice Environmental Management in mining includes a title on "Tailings Management"
— ***South Africa*** – the South African Bureau of Standards has developed a code of practice for mine residue deposits, and guidelines are being developed under the Mine Health and Safety Act;
— ***Peru*** – the ministry of Energy and Mines has engaged international expertise to develop a comprehensive national regulatory system for tailings impoundment system.

Lessons learned through analysis of tailings dam failures is a significant stimulant for operators of current tailings dams to re-assess the safety factors of their dams and review operational procedures, contingency measures and emergency response plans. These analyses also provide a sound empirical knowledge base for the design and specification of new dams [80] [82].

The regulatory frameworks for tailings impoundments in some countries, and availability of guidelines produced by government bodies, mining industry bodies, and professional organizations in others, provide a sound information set for selection and construction of an impoundment design suited to site and climatic variables. There is relatively little guidance from regulations or standards or guidelines on the placement of tailings within the impoundments. Information is growing from experience at mines, mainly aimed at increasing settlement densities to improve physical stability of the tailings mass and reduce the potential for instability of impoundment embankments.

5.4. Current approaches to tailings containment

5.4.1. Less-favoured options

In broad terms, today's options for containment for uranium mill tailings have narrowed compared with the range of methods that were described in Section 2.4. The key difference is that tailings must now be securely contained. Options like stacking on flat surfaces, or in-filling of shallow depressions with no or minor bounding, or any arrangement that allows for uncontrolled discharge of tailings liquor, are not considered to be acceptable practice anymore.

Valley (or ***gully***) ***containment*** (Fig. 6) may no longer considered to be consistent with best practice, although in steep mountainous terrain there may be few or no alternatives. Valleys naturally are *foci* for concentrated water flow, such that water management is a major challenge, especially effective diversion of extreme flows way from the impoundment. High level flows from flood events and extreme rainfall into the impoundment increase the volume of contaminated water to be dealt with, and are a significant cause of catastrophic dam failure [32] [82]. Other arguments against valley for tailings containment are that they commonly alienate some of the best quality agricultural land in the area. Foundation conditions may be weak and heterogeneous and in the event of catastrophic failure the resulting pulse of water and tailings may be funnelled down the valley system for significant distances, causing major damage, environmental impacts, and potential loss of life.

Marine disposal is an option not described in Section 2.4, but a method used for disposal for tailings from some other mines – notably some gold mines in the southwest Pacific region. IAEA guidance on this subject [83] uses as the basis for quantitative radionuclide limits the protection of humans from ionising radiation. However, it was recognised that in the case of ocean disposal, the general tenet that protecting humans would automatically protect other species would not hold. This is because comparatively large doses could be delivered to marine species without approaching the human dose limit, owing to the remoteness of any humans from the deep ocean environment. The final prohibition of sea dumping of radioactive wastes was agreed by Contracting Parties to the London Convention in 1972 [84] that came into effect as amended in 1994.

Therefore the options for containment available today can be summarised as follows:

- Above ground
    - Custom-built ring dyke (or turkey nest, or paddock) dam
    - Side hill containment

— Below ground level
  — Returned to a mined out pit
  — Returned to an underground mine
— Subaqueous
  — Tailings placed into a deep lake.

5.4.2. Above ground disposal

Most of the world's uranium mill tailings are contained above ground, i.e. above the natural land surface. It is commonly the most economical option, although in-pit and sub-aqueous disposal may be cheaper in some instances. The topography of the area around the mill will usually determine, whether valley dam, side-hill or ring dyke forms of construction are viable.

Table 4. Comparison of embankment tailings dams, modified after [87]

Embankment type	Mill tailings requirements	Discharge requirements	Water storage suitability	Seismic resistance	Raising rate restrictions	Material required	Relative cost
WATER RETENTION	Suits any type of tailings	Any discharge type suitable	Good	Good	In most cases entire embankment constructed in one operation	Soil, rock, clay	High
UPSTREAM	Sand fraction of tailings > 40-60% sand, low density slurry to promote sand segregation	Peripheral discharge and well controlled beach development	Unsuitable for significant water storage	Poor in high seismic areas	Less than 5-10m/year desirable	sand tailings or soil, rock, mine waste rock	Low
DOWN-STREAM	Suits any tailings type	Varies according to design details	Good	Good	None	Sand tails or waste rock if production rates sufficient, or natural soil, rock	High
CENTRE-LINE	Sand or low-plasticity slimes	Peripheral discharge and at least some beaching	Not recommended for permanent water storage. Temporary flood storage acceptable if proper design	Acceptable	Height restrictions for individual raises may apply	Sand tails or waste rock if production rates sufficient, or natural soil, rock	Moderate

All above ground level options have the disadvantage that the long term containment of the tailings will be at risk through erosive forces, and therefore they present inherently greater risk for long term physical isolation than for below- ground level options (cf. Table 4).

5.4.2.1. Ring-dyke containment

This is the most widespread method of containment construction today in terms of number of sites and total volume of tailings contained. This type comprises a single, enclosing embankment, on more or less level ground (Fig. 6). Whilst they can be any shape, rectangular and circular are the most popular. The type of construction varies. Some are built as 'water-retaining structures' with zoned construction and rolled clay cores. This type is usually built to full height in one operation. Others are built similar to agricultural dams, simply by pushing up locally available fill — commonly overburden stripped from above the ore body in the case of open pit mining. Most of this type are built as a series of 'raises', with the walls being increased in height in stages as the dam fills up with tailings. This reduces the capital cost of works in the pre-production stage of mine life, and helps to match construction to periods when raw materials are readily available. These 'raises' may be built of locally available fill, waste rock or sub-economic ore. Where this is not available, particularly where underground mining techniques are used, then the raises are commonly built from the coarse fraction of the tailings, won by cycloning.

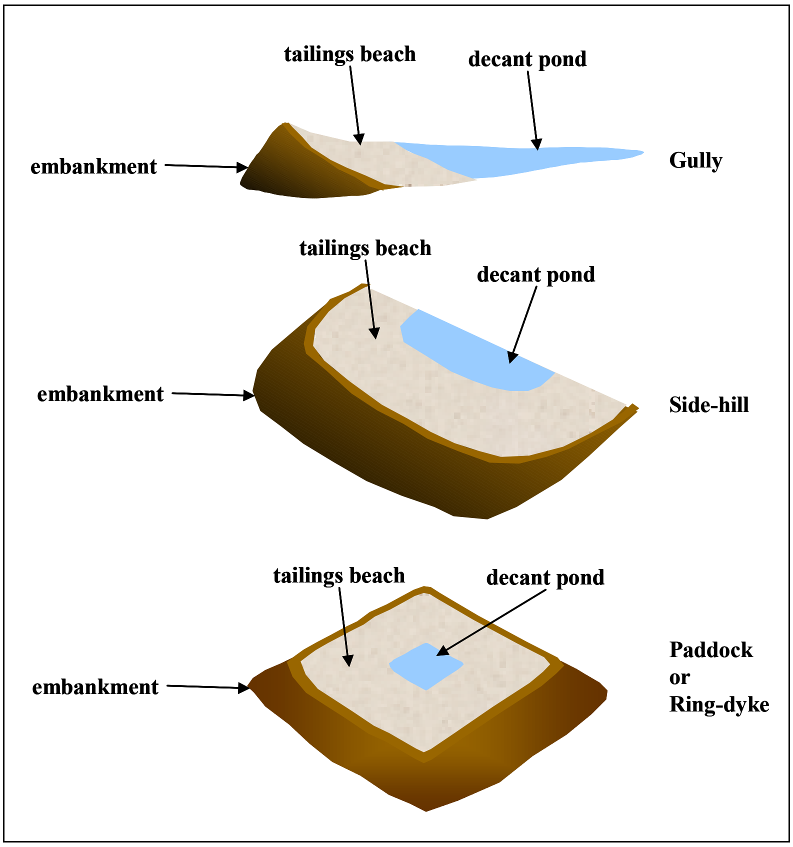

Fig. 6. Types of above ground impoundments, after [85].

The raises may be constructed by three different methods – upstream, downstream, or centreline (Fig. 7). The various requirements for and features of each are compared in Table 4, which also compares these types of construction against water retention type embankments.

The upstream technique is the cheapest method as the amount of material needed is the least. However, it requires that the tailings beach over which the raise is to be constructed has a high sand content, to provide a high-strength, well-drained foundation. This may be provided, if the tailings slurry has a high water content so that there is good hydraulic sorting on the beach, that is the sand fraction deposits near the head of the beach where the spigot is mounted, and the fine fraction/slimes run down to the pond. Examples of upstream-constructed tailings dams include Kerr-McGee and Homestake, both in New Mexico.

The phreatic surface (i.e. top of the saturated zone) expected to develop in each type of dam is different. If the phreatic surface emerges on the external face of the dam wall, seepage will result, with major implications for dam wall stability. The upstream method is the worst in this regard. Special internal drainage systems may be required to improve embankment stability to within acceptable levels. Raises constructed of cycloned tailings result in the embankment having a higher permeability than the tailings. The phreatic surfaces shown in

Figure II-5 (left) are modelled on that assumption. If the raises are built of material less permeable than the tailings, then other drainage provisions need to be considered.

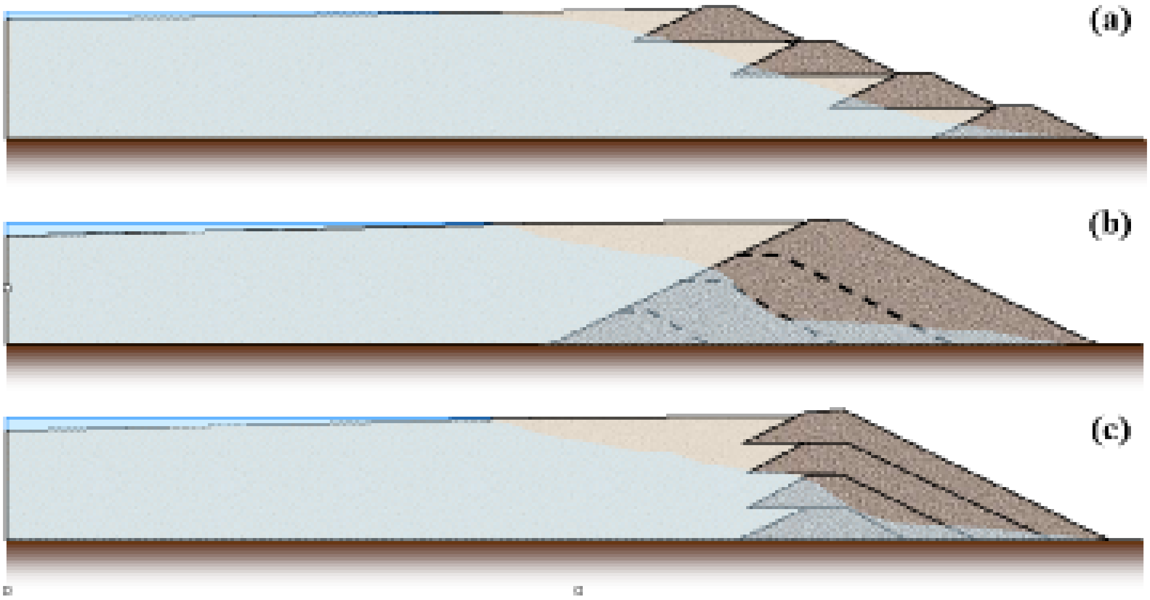

Fig. 7. Cross-sectional views of different techniques for constructing 'raises' to embankment dams, after [86].
a= upstream raises; b = downstream raises; c = centreline raises;
Note: pale blue indicates saturated zone.

Some embankment dams are constructed as water containment structures, and whilst many may be constructed in their entirety at commencement, it is possible to build them in 'raises' too — the tailings dam at Ranger mine, Australia, is built like this with each raise requiring the impermeable clay core to be exposed, keyed in and extended as the rock inner and outer ramps are raised. In this type, the phreatic surface runs down the inner face of the clay core and passes under the base of the embankment foundations, providing high security against seepage and improving stability.

Another important strategy for maintaining a low phreatic surface in the embankment is for the tailings beach to be as wide as possible — that is the decant pond should be as far away as possible from the embankment. This may require careful design and operation of the water management system for the facility, especially in higher rainfall areas. The disadvantage, however, of a wide beach may be the segregation of tailings into different size fractions:

– highly permeable sands close to the discharge point that allow easy percolation and contamination of rainwater;
– a fine tailings sludge further away that may require many decades to consolidate. It would also be subject to uneven settlement and there will on-going release of porewater.

As a consequence, operators take care to prevent segregation in tailings, unless segregation is needed for providing a safe foundation of up-stream raises.

When the containment is full or the facility closes, a number of problems for satisfactory management and remediation arise. Access onto the surface to spread cover materials can be difficult, time-consuming, and expensive due to the unstable nature of the tailings material.

For instance, an estimated cost of US$ 42.4 million for construction of a geotextile/rock platform across the 100 ha Ranger tailings dam contributed to the decision to relocate the tailings to a worked-out pit, rather than rehabilitate *in situ*. A variety of techniques to accelarate dewatering have been developed, such as wick drainages and vibrasonic or seismic shockwaves. Surface runoff can be problematic if erosion is to be avoided. The long term erosion stability of the relatively steep embankment batters poses a problem. Seepage may be a problem and will require special infiltration capping if geotechnical problems are to be avoided.

A concern for all types of embankment dams that are extended in increments is that the raises are commonly constructed by mine personnel who may lack specialised engineering expertise. Also the materials available when a lift is due may not meet specification. In some cases the life of the mine and mill may be extended, and the dam design modified by more raises than those originally intended, so that its original design parameters are exceeded.

With respect to uranium mill tailings dams, using the sand fraction or sub-economic ore to construct raises leads to some issues related to the radioactivity of the material. These issues include the possibility of contaminated runoff from the outer wall of the dam, and the need for the final cover to extend across the entire dam including the outer walls to provide sufficient long term stability and to reduce radon flux. In contrast, embankments built with non-radioactive material, such as rock overburden, may be constructed to provide enough post-closure stability without modification, and the original batter may form part of the post-remediation landform.

Examples of ring dyke type impoundments for uranium mill tailings include Ranger and Olympic Dam mines, Australia; Key Lake in Canada; and Homestake and Kerr McGee and Ambrosia Lake in USA. The Ranger impoundment is constructed as a water containment structure throughout the 4 km length of its wall. The Key Lake impoundment is constructed on a slightly inclined surface, with embankments on all sides built from compacted till in one construction campaign. Discharge spigots were placed along the inner side of the upstream embankment to encourage beach formation adjacent to it to improve stability, and formation of the decant pond against the opposite side of the containment. This opposite side is built to water containment standard to avoid the risk of seepage through the embankment [88].

5.4.2.2. Side hill containment

Dams of this type share the same design and construction characteristics and variables as ring-dyke impoundments (Fig. 7). The only significant difference is that they are constructed on generally planar sloping ground. Unlike valley or gully dams, they do not cross an area of concentrated runoff, so that problems associated with storm events, such as flooding and over-topping are less likely. Similarly, in the event of failure, potential contamination is less likely to form confined and fast-moving flows and consequently the risk of catastrophic damage is more remote. An example of a side hill dam is the Cluff Lake Waste Management System [23], where an embankment was built with a compacted soil/bentonite layer beneath and parallel to the inner wall surface that was keyed into the foundation material.

5.4.3. Below ground containment

This approach provides high long term security for the isolation of tailings, but is only a viable option where mined-out pits or underground workings are within pumping distance from the mill. Alternatively, if the mill is not yet built, it may be economically feasible to site the mill close to the respective underground workings or pits.

Whilst tailings have been placed in pits or underground workings made from mining non-uranium ores, such as at the Zhovty Vody mill in the Ukraine, where one of the three impoundments is in an old iron ore open pit [7], most examples are of uranium tailings being returned to uranium mine openings made in the same or earlier phases of mining activity.

Placement of tailings below ground, where applicable, has the following advantages:

— Reduces the amount of land needed for tailings storage. Storage of uranium mill tailings in above ground containments quarantines land effectively for perpetuity, or at least reduces the range of options for future use of that land. The possibility of reducing the amount of respective land-used is seen to be of significant environmental and social benefit – particularly where the land has desirable environmental attributes, is valuable agricultural land, or has the potential for other social or economic beneficial use.
— Effectively eliminates the potential for catastrophic failure of the containment, provided that the underground workings or pits have no drainage adits exiting to the surface, or such openings are plugged with high degrees of reliability.
— Greatly reduces the potential for erosion and dispersion of the tailings pile, even in the very long term.
— Places the tailings into a geological situation similar to conditions prior to mining (particularly where the tailings are placed in old uranium mine openings).
— Allows thick capping with benign materials to maximise isolation of tailings from the surface and optimise options for beneficial land re-use.
— Maximises the probability of remediation and landform construction outcomes that harmonise with the natural landscape and ecosystems.

Where tailings have been placed below ground during milling operations, or at the time of decommissioning, there is likely to be little further scope for and need of remediation [7]: *"The placement of tailings below or under ground is likely to provide the best long term management solution from the point of view of both reducing potential liability, and providing the greatest long term environmental safety. However, the possibility of leaching and suffusion by permeating ground waters has to be considered. Possible options for prevention and remediation include the sealing of open mine workings and the creation of underground barriers by injecting grouts etc."*

5.4.3.1. Tailings deposition in mined out pits

Examples where uranium mill tailings have been returned to exhausted pits include:

Ranger mine, Australia, where tailings from the #1 pit were temporarily stored in a ring dyke impoundment; when the pit was exhausted and mining began in the second pit, tailings were then discharged directly into the now exhausted #1 pit [89];

Nabarlek mine, Australia, where the entire orebody was mined in a six month period and stockpiled for milling over the following ten years. Tailings were deposited directly into the pit from whence the ore came [90];

Rabbit Lake, Canada, where tailings arising from mining and milling of the Collins Bay B-Zone orebody, Canada, are placed in the mined out Rabbit Lake pit [23];

Key Lake, Canada, where tailings arising from the Key Lake mill are placed in the mined out Deilmann pit [91].

Seelingstädt, Germany, where tailings were deposited into mined out open pits at Trünzig and Culmitzsch [15].

Spook, USA, where dry tailings were placed back in the open pit [92].

West Wits, South Africa, where cycloned tailings are discharged into an open pit after reprocessing for gold.

Many of the pits and underground shafts etc are below the water table, and prevention of groundwater contamination has become a significant issue, particularly in cases where sulfidic tailings or wall rock increase acidity, and therefore the mobility of radionuclides and toxic metals.

Available pits may be of insufficient volume to accommodate all of the tailings, including cases where tailings are being returned to the same pit, from which the ore was mined and overburden and waste rock ratios are low. In this case it is important to maximise settled densities by removing as much water as possible. The relatively low and confined surface of the tailings mass reduces evaporation levels, so that in cases where evaporative loss is important to improve settled densities, ancillary pumps and evaporation pans may be required, or tailings thickened prior to deposition.

5.4.3.2. Tailings deposition in underground mines

The use of mill tailings as backfill in underground mines was first recorded in South Africa in the early 1900s [93], and it is now in widespread use. Most operations use cycloned coarse fractions from the tailings to make cemented paste backfill, which is placed in open stopes to provide stability for the removal of adjacent ore blocks. However, this left the problem of slimes disposal. In 1990 high-density backfilling with whole tailings was introduced at the BBU Bleiberg/Kreuth lead-zinc mine in Austria. This was done in response to regulations prohibiting the further surface disposal of slimes [22].

If cycloned, uranium mill tailings are to be used as backfill, issues to consider include:

— Emanation of radon and radon daughters will continue. If the mine is to remain active the mine ventilation system will need to cope with increased radon levels.
— Most of the radioactive components are in the fine or slimes fraction. Therefore, the radionuclide concentrations of slimes to be disposed of on the surface will be increased.
— An above ground facility disposal facility is still required to dispose of slimes.
— The slimes are virtually impossible to consolidate and remain highly susceptible to erosion.
— Underground tailings placement can lead to groundwater contamination, especially if the tailings are not neutralised, the wallrock is highly fractured, and the tailings and/or host rock are sulfidic.

Research into paste tailings backfill technology for non-uranium tailings is now focussing on incorporating the whole-tailings. This would avoid the problem of slimes disposal on the surface. Results from this research will be of potential benefit to the uranium mining industry, if similar techniques can be developed for uranium tailings.

It may be noted that frequently the objective of the deposition of tailings underground is primarily mine stabilization and not tailings disposal. Generally only the cycloned fraction of a small proportion of the total volume of tailings was involved in this practice.

5.4.4. Deep lake

The practice of uranium tailings disposal in lakes is basically confined to Canada, where the great number of lakes, and their remoteness make them obvious candidates for tailings containment. In the 1950s the Lorado and Gunnar mills discharged tailings to lakes, mainly the Nero and Mudford Lakes, with no controls or treatment. In the late 1970s control structure and chemical treatment were introduced, with the use of barium chloride and ferric sulfate to remove radium [23]. The practice is no longer used at active mines, nor for the decommissioning of old tailings facilities. The possibility of relocating all the Elliot Lake tailings to Quirk Lake was considered, but rejected due to the high estimated capital cost and negative short term environmental effects on fish habitats and downstream residents [94].

5.4.5. Purpose-built containment

Another option for tailings disposal is the excavation of purpose-built underground openings for the disposal of uranium mill tailings. There are two examples of where this option has been considered, but to date the approach has not been applied.

At Elliot Lake in Canada one disposal option considered was the relocation of tailings to underground workings [94]. As this would accommodate only about 25–30% of the total volume, new underground voids would need to be created to accommodate them all. This option was rejected on the basis of cost and the need to dispose of the waste rock so created in an environmentally considerate manner.

The Jabiluka mine proposal in Australia was modified during the Environmental Impact Assessment process to include purpose-built underground ‘silos’. This was to accommodate tailings that would not fit into the stope space. The proposal was to emplace the tailings as cemented paste to provide stability for the removal of adjacent stope blocks. Whilst the proposal received government approval on this basis, the project has not gone ahead because of the failure to negotiate land access and use agreements with indigenous land owners.

The major benefit of this option is the ability to choose low-permeability, benign rock into which the tailings can be deposited, and to engineer the void to optimise placement and containment. Also, the potential risk of dispersal due to surface erosion would be avoided. However, as in the Elliot Lake case, the big disadvantages are the cost of construction and the need to rehabilitate the waste rock dump created by the excavation. Another consideration is the potential contamination of groundwater, and possibly surface water, due to leaching, when no lining and surface cap prevents the infiltration and migration of atmospheric precipitation. At Jabiluka, however, this was found to be insignificant owing to the low permeability of the host rock. Uranium and radium are predicted to travel less than 50 m in 1000 years [95] [96]. Following this assessment, the mining company tendered an alternative method of storing the excess tailings in specially excavated pits instead of in underground silos.

5.5. Current approaches to stabilise and isolate uranium mill tailings

5.5.1. Design objectives

The designs for tailings containments described in the preceding section represent different ways of stacking tailings so that their interaction with and impact upon the environment is kept within acceptable limits. The performance of a tailings impoundment can be maximised and the risk of failure reduced,

- if careful thought is given to siting dam-like containments,
- by careful supervision to ensure construction is to specification,
- by identification of the potential risks and development of appropriate 'normal' and 'contingency' management strategies and operational plans, and
- by implementing appropriate quality control mechanisms.

However, the history of failure of tailings containments over the last 40–50 years, shows that containment design and management themselves may not be enough to provide a high level of certainty that the tailings will remain properly isolated from the environment for indefinite periods. This is particularly true when we consider that uranium mill tailings need to be effectively isolated for a long period to provide a high degree of certainty that the public and environment will not be negatively affected. The short duration of baseline and performance monitoring results in uncertainty in any long term predictions.

A number of design steps and procedures are required to assure the effectiveness of isolation of tailings. These can be divided into containment preparation, tailings preparation, tailings discharge and deposition, tailings dewatering, tailings surface treatment, decant water treatment, seepage control, and capping. They usually require ancillary infrastructure in addition to the containment structure itself, and are best managed as an integrated operation.

5.5.2. Containment preparation

Containment preparation involves work to reduce the permeability of the structure in order to reduce the ingress of groundwater, and egress of tailings porewater, thereby reducing the potential for groundwater contamination. Examples, where unlined containments have led to groundwater contamination, include the two tailings dams near Pécs in Hungary. There seepage through the base has raised the water table and threatens to contaminate two aquifers that provide more than 60% of the drinking water for the city [41]. At Olympic Dam in South Australia, sudden seepage from the tailings retention system in 1993-94 caused a localised rise in the water table, but as the aquifer is 60 m deep, highly saline and unfit for human or lifestock consumption, and no significant change in water quality was detectable, a subsequent government inquiry did not require recovery or treatment of the seepage [97].

In above ground level tailings facilities the floor and walls may be sealed with a low-permeability membrane, or the floor may be covered with a rolled clay layer and the walls may incorporate a clay core or clay layer. For example, the Ranger mill tailings dam has a clay floor and a clay core in the walls. As tailings are compacted under their own weight by the addition of more tailings, their permeability will decrease. However, the seepage rate may stay the same since, of course, the head also increases as more tailings are added. The Key Lake tailings facility has an underseal beneath the tailings basin and the inner part of the embankments. US regulations now require all above ground containments not managed as part of the UMTRA programme to be lined with a double-skinned liner with a seepage collector system between the two skins; the lower skin can be compacted clay provided it is no less than 0.91 m (3 feet) thick and has a vertical permeability of no more than $1 \cdot 10^{-9}$ m/s.

In below ground containments, the most common technique to reduce permeability is rock grouting into fracture zones, lining of porous faces with an impermeable membrane, or application of a cement-based low-permeability layer. Careful geotechnical mapping of the containment walls is required to identify areas requiring treatment. In some mineralised systems, fracture zones may be accompanied by certain mineral alteration assemblages; these should be tested to ascertain possible reactive relationships with the grouting to be used.

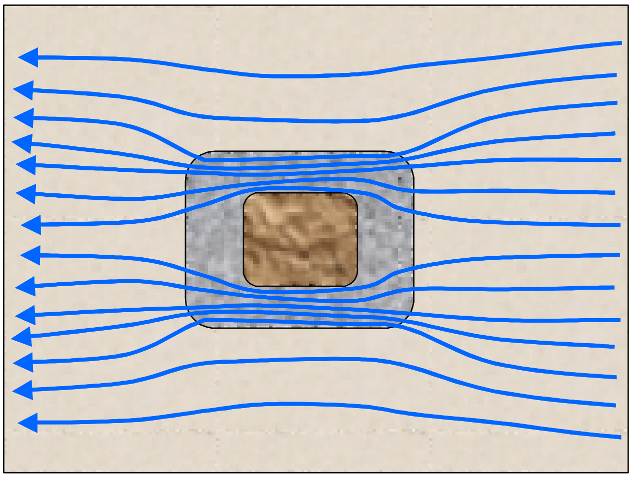

Fig. 8. The concept of pervious surround (W.E. Falck, IAEA).

A recent innovation in some below ground containments is the construction of a highly permeable surround placed around the outside of the tailings mass. The concept is that regional groundwater flow will pass preferentially through the highly permeable gravels instead of through the much less permeable tailings, thereby reduce the source term to a diffusion controlled process (Fig. 8). The Rabbit Lake disposal pit is an example of the pervious surround method. The Ranger mine has a partial pervious gravel blanket at the base of the tailings mass. Water is drawn from the gravel bed via an adit and well pump and is returned to the decant pond above the tailings for evaporative loss; this system assists in achieving the settlement density of > 1.2 g/cm^3 required under regulation.

5.5.3. Tailings preparation

It is common practice to neutralise tailings prior to disposal. The effect of adding neutralising agents, however, has to be investigated carefully, as various constituents may have varying solubility ‘windows’. Adding carbonate compounds might have the unwanted effect of increasing the uranium solubility due to the formation of highly mobile uranyl-carbonate complexes. Circumneutral to slighly alkaline pH-values would reduce the solubility of uranium. The addition of buffering capacity would also reduce the potential for acid generation within the tailings mass and, hence, the potential source term for contaminants. The choice and quantity of neutralization agents needs to match the predicted long term conditions within the tailings deposit as well as the conditions at the time of deposition.

Adding chemical binders can have a triple effect on the tailings properties: a chemical bond is created between constituent particles, some of the porewater may be consumed in the ensuing chemical reactions (e.g. as crystal water), and the pH may be raised (e.g. by adding cement).

Typical additives are ordinary Portland cement, or $BaCl_2$ (see Annex VI) where sulfates are available. Using residues or waste products with suitable hydraulic properties, such as fly-ash, may be of (economic) advantage. The prepared tailings material then can also have further constructive uses.

For instance, it is common practice in underground base metal mining to prepare tailings into a cementiferous paste suitable for return to underground workings to stabilise stopes so that adjacent ore blocks may be removed. Fly-ash based grouts are used for this purpose on a large scale in coal mining. In the uranium mining industry its use appears to have been limited to the Grants mineral Belt in New Mexico [98]. The technique formed part of the Jabiluka mine proposal, in order to reduce the surface impact from mining to as low as practicable owing to the proximity of the mine area to Kakadu National Park [96]. The proposal was to return 75% of the tailings to the underground workings as cemented paste. The remainder was to be placed in either purpose-built pits or underground stopes in benign rock. The potential for groundwater contamination from this underground disposal of cemented tailings was given special scrutiny in the environmental impact assessment process. To be suitable for the application of paste technology, tailings should contain at least 15% by weight of particles of less than 20 μm diameter to avoid segregation [99]. At Jabiluka, the tailings have more than 30% of this particle size range. Permeability varies according to stress, i.e. as overburden pressure increases, permeability decreases. A permeability of less than 10^{-9} m/s was recommended for tailings placed underground in order to reduce the potential for groundwater contamination. Whilst it was calculated that this permeability was impossible to attain in uncemented tailings, addition of 4% cement to the tailings would lower permeability by three orders of magnitude and ensure that potential for groundwater contamination is minimal [95]. In addition, reduction of water content by dewatering the tailings before cement addition will improve cohesiveness and reduce the likelihood of segregation [96].

The rather uniform grain-size distribution and gel-like physico-chemical properties lead to poor dewatering behaviour under gravity only. Some physical treatment to accelerate the process may be needed. Such processes include cycloning, centrifuging, filtering, evaporation in open pans, and more novel techniques such as electro-osmosis. The outcomes range from a percentage reduction in water content, through to the production of filter cake. The various techniques are applied widely in the mining industry, particularly in preparation of tailings to specified consistencies for making cemented paste for back-filling of underground workings, and for preparation of tailings to a consistency suitable for dry stacking.

A terminology has been developed to distinguish the different states of dryness of dewatered tailings [100]:

- Slurry tailings – the typically segregated mass of tailings that is in a fluidised state for transport by conventional distribution systems (e.g. pumping);
- Thickened tailings – partially dewatered slurry that has a higher solids content by weight than the basic tailings slurry, but is still pumpable; chemical additives are often added to enhance thickening;
- Paste tailings – thickened tailings with some form of chemical additive (typically a hydrating agent, such as Portland cement);
- wet cake tailings – a non-pumpable tailings material that is at, or near, water saturation;
- Dry cake tailings – a water-unsaturated tailings product that cannot be pumped.

Development of large capacity vacuum and pressure filter technology has provided the opportunity for storing tailings in a dewatered state as a dry cake. The technique has not been applied in the uranium mining industry, but is used in several base and precious metal mines in arid or in very cold climates. In arid areas the tailings are placed as a dry tailings stack without containment. Advantages the technique offers are:

- (process) water conservation in arid areas;
- enhanced metal recovery by tailings filtration;
- reduced risk of liquefaction etc. in areas of high seismicity;
- avoidance of having to handle drainage waters in very cold regions during winter;
- minimised entrapment of frozen porewater in very cold regions;
- reduced volume of tailings.

Accelerated dewatering and thickening of tailings under controlled conditions will help to reduce grain-size segregation during disposal and, hence, improve the further dewatering under gravity or overburden. For instance, tailings discharged at Key Lake contain 35–40% solids. The same solids to solution ratio is achieved at Ranger mine in northern Australia without thickening. Although rainfall is high (1,565 mm annually), evaporative loss is much higher (around 2,500 mm), and so evaporation is a major factor in achieving water loss after tailings discharge. An old tailings pond is now used as a large evaporation pan and the tailings are deposited in a worked-out pit.

The geomechanical and dewatering properties of tailings can also be improved by mixing them with other mining residues, albeit at the price of increased volumes. Mixing the tailings with material with a broader grain-size distribution is conducive to better settlement and improved dewatering.

In the USA, regulations require that wet tailings be dewatered to meet consolidation requirements, if capped in place, or that the moisture content be less than the specific retention capacity of the tailings material, if relocation is chosen [3].

5.5.4. Tailings discharge and deposition

An important objective of tailings discharge and deposition is to optimise the use of available area and volume, while maximising the physical stability of the impoundment. The method of tailings discharge is important, as it affects grain-size segregation and the final density. In turn, these determine the impoundment stability and accessibility of the tailings surface for further work. These factors also relate to other considerations, such as potential liquefaction during earthquakes, or flow behaviour in the event of an embankment failure. Deposition of denser tailings will reduce the requirement for dewatering in the later stages of remediation and decommissioning.

Greater physical strength of the overall structure of the impoundment can be obtained by isolating the coarse fraction of the tailings. This is done for example when the coarse fraction is cycloned out, so that the sand-grade material can be used for construction of embankment raises. The sorting can also be achieved by discharging from spigots and allowing the tailings to form and then flow down a slope, whereupon the coarse fraction settles first (i.e. closer to the spigots) to form a beach. It is now common practice in the case of above ground level impoundments for spigots to be placed along the inner top side of the embankment so that coarser fraction tailings build up adjacent to the embankment, with the slimes settling out in the centre of the dam (i.e. a single spigot may be periodically moved to different points around the dam wall, or the tailings discharge line may encircle the dam and have several discharge points, which may all discharge at the same time, or which may be switched on and off). This approach significantly improves the stability of the structure, as the beaches provide physical support to the embankments. Stability generally improves over time as densities increase under the weight of additional tailings deposition. It also removes the finer fractions, with greater potential to liquefy, away from the walls of the impoundment. At the Key Lake above ground facility, discharge took place along one side of the dam, causing beaches to form against the embankments, which on this side were constructed of compacted fill. The slimes and the supernatant pond formed on the other side of the dam that was constructed to water retention standards [23].

This approach does not necessarily guarantee that settlement and consolidation will take place to desired levels. At the Ranger mine (Australia), various discharge arrangements were tried, including subaqueous discharge, central discharge, and perimeter discharge. Perimeter discharge became the preferred approach, and wide beaches developed against the embankments all the way around the dam. However, the densities in the tailings were extremely low in places, with the tailings forming a gel-like matix. These very low densities contributed to the decision to relocate the tailings into a worked-out pit, as access onto the tailings for capping and remediation would have been difficult and expensive.

Tailings discharge control is also critical for in-pit containments. It is desirable to use techniques that will maximise the final density so that more tailings will fit into the available space. Denser tailings will also tend to have a lower permeability reducing release of contaminants into the surrounding groundwater. Avoiding grain size segregation is important to achieve a good consolidation behaviour..

5.5.5. Tailings consolidation

The main mechanism to achieve consolidation of tailings is their dewatering. Dewatering can be undertaken prior to discharge into the tailings containment as discussed above, or *in situ*, after the tailings have been placed into the impoundment. The various technical options available are summarised in Table 5.

An under-drain gravity system provides an effective means of dewatering tailings. For impoundments already constructed without provision for gravity drainage horizontal drains, a single-well, or well-point systems are recommended for reliable dewatering [101]. Limited recharge rates may restrict the use of a single well. A multiple well system with variable pumping positions may be more effective. The settlement of tailings may be accelerated by stacking additional material and by loading due to cover construction. Dewatering can cause settlements of magnitudes equal to or greater than those caused by placement of the cover material. Therefore dewatering during the operational phase would reduce the need for post-closure dewatering prior to placement of the cover.

Most of the *in situ* dewatering techniques listed in Table 5 are used at various uranium mine and mill sites around the world. However, information on dewatering techniques is rarely presented in the published literature. Certainly under-drainage and evapo-transpiration techniques are widely used (e.g. Ranger, Key Lake), and wicks have been applied to dewater tailings prior to closure at several mines. At the Atlas mill near Moab, Utah USA, wicks on a 3 m pattern were used in an above-surface, unlined ring-dyke impoundment to consolidate the tailings mass and to reduce seepage to the groundwater system [102].

Table 5. Comparison of *in situ* dewatering techniques for tailings (after [101])

Technique	Advantages	Disadvantages
Under-drains	Economical construction Minimum complexity Existing technology Stabilise tailings during operation	Must be placed before tailings discharge starts Potential clogging from poor construction of precipitated salts Only gravity drainage
Horizontal drains	Drain existing impoundments Supplemental advantage with vacuum application	More expensive Requires long drains
Single-well system	Technology well developed Dewater existing impoundments	More maintenance & equipment Limited recharge may restrict recharge radius of influence Requires stable access
Well points	Same as single well May be used on perimeter to avoid interior access problems Flexibility in pumping positions	Maximum 7.6 m (25') depth More expensive than single wells
Jet pumping	Same as well points Increased effective depth to 30 m (100') or more	Increased equipment costs
Electro-osmosis with well points	Greater depth than well points alone Applicable to low permeability soils	Expensive Limited effectiveness in acidic pore water
Evapo-transpiration	Passive system	Limited depth Adverse impact on cover integrity
Sand drains/wicks	Dissipate pore water pressures to increase consolidation	Little dewatering capacity
Pervious surround	Three-dimensional drainage	Requires an open pit

The target of 90% consolidation was not achieved within the requisite time, as only a fraction of the design surcharge load was applied because of lack of funds. However, the degree of dewatering by the wicks was greater than that predicted.

At the Nabarlek mine in Australia, wicks were inserted in a 3 m grid pattern to a depth of 33 m (100 ft) into the tailings (<60 m thickness) in the mine pit to assist in settlement and dewatering [22]. Water expelled from the wicks collected on the upper surface of the tailings and was evaporated through sprinklers placed around the top of the pit walls. The tailings were covered with contaminated material from mill and pad demolition, and capped with stockpiled rock overburden (totalling about 15 m). Subsidence of the cap has been slightly less than the predicted 1.5 m and has been uniform across the cap with only minor cracking in some areas above the edge of the pit, but no subsidence is evident over the tailings mass.

At Seelingstädt and Helmsdorf tailings ponds in Germany (WISMUT), wick drains (Fig. 9) are used on a large scale for the construction of the intermediate layer in remediating the large tailings impoundments (2.5 m 2.5 m grid and 5 m depth). Deep wick drains have also been employed in the Trünzig tailings pond to assist in dewatering of finer parts of the tailings pile.; Loading by placing a temporary embankment over this area of tailings assists in achieving the desired amount of settlement within five years [104].

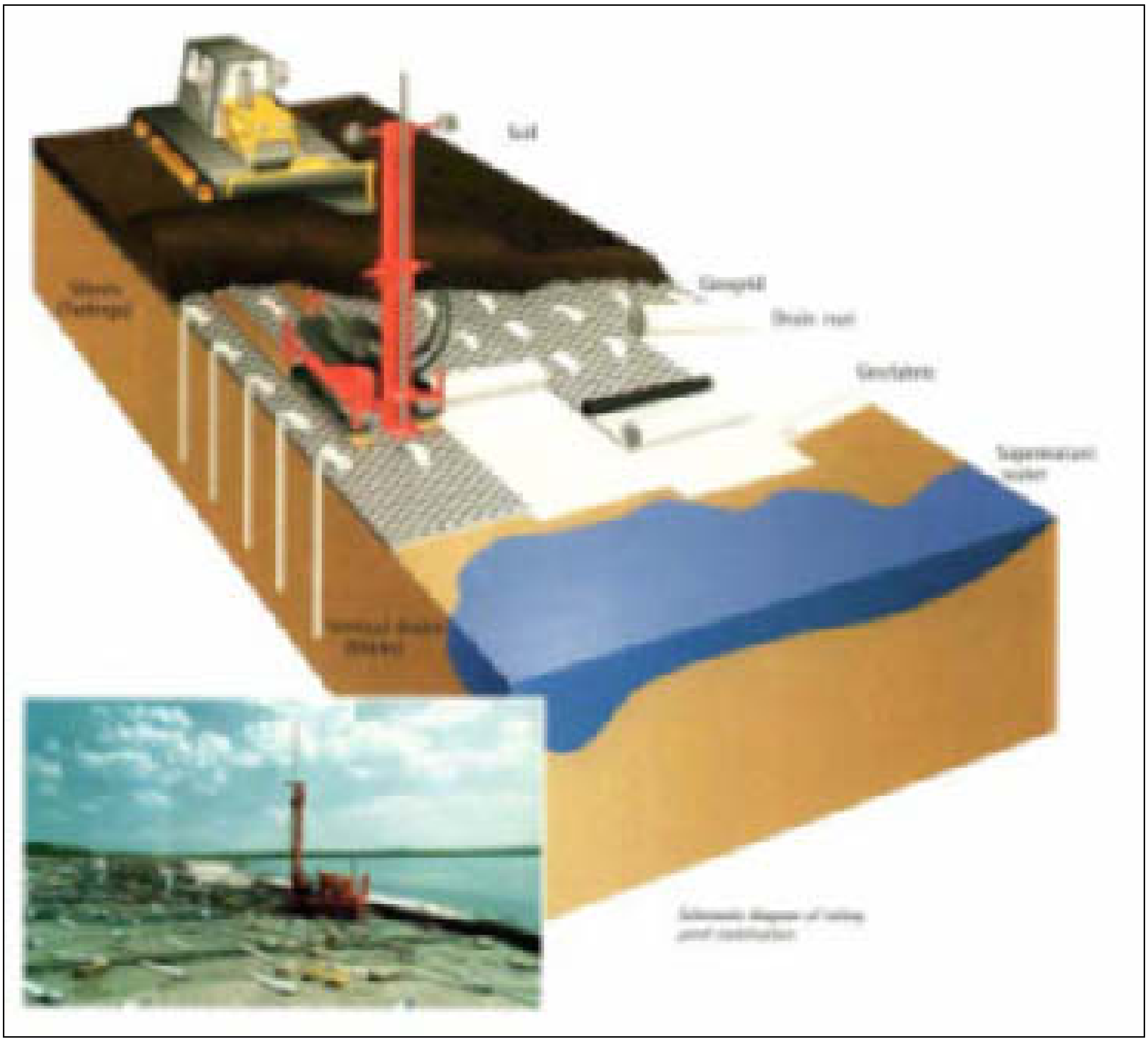

Fig. 9. Construction of wick drainage system on a Wismut tailings pond [103].

5.5.6. Tailings surface treatment

Treatment of the surface of tailings within containments is designed to temporarily reduce radon flux and the generation of fugitive dust prior to final covering. Treatments include the maintenance of a water cover. Depending on the circumstances, this may be incompatible with tailings dewatering and improved settlement through evaporative water loss. Careful management is required to avoid possible instability of embankments as a consequence of wave erosion, or high phreatic surfaces leading to piping or seepage through the walls.

Where beaches are allowed to form continual wetting of the surface to prevent dusting and assist evaporative water loss may be needed. At Ranger mine, for instance, tailings were initially deposited subaqueously to reduce radon flux. However, this approach was replaced by beaching and continual wetting by sprinklers following research that demonstrated that the rate of radon flux was not significantly different between the two techniques.

Another form of surface treatment involves the application of sealants or stabilising agents to reduce the potential for dust generation from dry surfaces, improve erosion resistance, or reduce water infiltration. This approach may be particularly useful at the stage between completion of tailings deposition in a surface impoundment and the removal of decant water, to reduce the risk of surface failure prior to commencement of final covering (e.g. when the facility is kept on a care and maintenance basis in between operational periods). The use of certain sealants is being investigated and developed *inter alia* in Russia (see Annex XI) and Kazakhstan (see Annex VII). However, like the use of bitumen or molasses, the use of other types of organic binders usually can be viewed only as an interim solution due to their biodegradability.

5.5.7. Decant water treatment

Decant water (supernatant water) contains most or all of the suite of contaminants in the tailings. It may readily escape to the environment in the case of overtopping, breaks in pumping lines, or failure of the drainage system. The decant water is either returned to the mill as part of the take-up water, thus reducing the draw on uncontaminated water, or is treated and released to the environment.

In arid climates, the standard practice for decant water management is the evaporation of water in ponds, sometimes with the assistance of spray systems.

5.5.8. Seepage control

Most tailings impoundments by their very nature will produce seepage at some stage during their life. Seepage will continue after closure due to local groundwater in-flow or surface infiltration unless adequate measures are taken to control this. The long term performance of liners is not guaranteed, and seepage may increase because of imperfect joining seals and accidental damage during installation, penetration by rock fragments etc. The design objective for new facilities and the main target for impoundment remediation is the elimination of all factors that could lead to continued seepage. This is the design target for all of the UMTRA sites. For example, Tuba City, Arizona, modelling calculations support the conclusion that continued seepage will occur at a very low rate. This is confirmed by monitoring over a period of ten years. This site is located in an arid environment. In more humid climates and under other site specific conditions, some seepage is likely to occur. Good practice therefore requires the installation of seepage collectors and treatment systems. Long term treatment implies a commitment to costs in the future. Therefore any such system is best planned in such a way that passive treatment will eventually be adequate However, passive treatment systems cannot be regarded as walk-away solutions. Acidity is the master variable controlling the behaviour of most radionuclides and other toxic metals. A variety of ways of treating acid rock drainage (ARD) and acid mine drainage (AMD) have been developed (see also Section 7.6), such as slag leach beds and oxic or anoxic limestone drains [105]. A recent IAEA report provides more details on ARD/AMD treatment systems, including biological systems [106].

— At Ranger mine a seepage interception system was installed around the perimeter of the embankment impoundment. The seepage is monitored, collected in a sump, and pumped back into the tailings dam.

— At Key Lake, seepage collection is engineered into the tailings facility structure, through placement of a filter blanket under the tailings pile. Seepage pore water is expelled into the filter as a result of increasing overburden pressures and hydrostatic head as the tailings pile gets thicker. The seepage is pumped to the waste treatment circuit for treatment [23].

Permeable reactive liners and barriers are an important new development in the management of tailings seepage *in situ* [107]. These make use of chemically reactive materials such as zero-valent iron, peat or compost, carbonates, phosphates, zeolites etc. to sequester mobile contaminants from the seepage water [106] [108].

5.5.9. Covers

5.5.9.1. Design objectives

The objectives of cover installation on uranium mill tailings are to minimise radon and dust emission, shield the environment from gamma radiation, minimise water and oxygen infiltration, control erosion, and to form an aesthetically acceptable landscape that fulfills these technical objectives.

5.5.9.2. Historical review of covers in the USA

The many years of research into cover design for effective long term isolation led to a gradual evolution of cap design, based on an improved understanding of the probable behaviour of early designs in response to natural erosive forces over the long term, and an appreciation of the benefits of harmonising designs with natural forces by incorporating vegetation and landscape evolutionary parameters [109]. Early designs focussed on radon attenuation and longevity of the structure. They comprised three layers: a compacted soil layer that formed the radon barrier, a rock rip-rap layer to provide protection of the radon barrier from erosion, and a layer of coarse sand between the two that was designed to act as a bedding layer for the rock and as a drainage layer for rainfall to percolate off the slopes of the containment structure. The design of the first rock cover constructed by UMTRA is shown in Fig. 10. The cover at Shiprock, New Mexico, comprises a relatively thick (over 2 m) compacted silty sand radon barrier layer and a 300 mm rip-rap layer. The greater thickness than usual of the radon barrier that was needed for radon attenuation reflected the relatively coarse texture of the material used to construct it, and high ^{226}Ra concentration in the tailings [110]. This cover was installed in 1985. In 2001, the disposal cell was investigated with a piezocone and although the study did not meet the full objectives to completely characterise the moisture distribution, it appears that the cover is performing as designed [92].

After the US EPA published draft groundwater quality standards in 1987, greater design emphasis was based on low permeability for the compacted soil layer. In 1989 UMTRA adopted a standard for low-permeability caps that required a saturated hydraulic conductivity of less than $1 \cdot 10^{-9}$ m/s, and a permeability not less than the underlying tailings so as to prevent water ponding in the tailings mass. The approaches taken to achieve the standard were either highly compacted native soil, or bentonite-amended soil.

Fig. 11 shows a cap design incorporating a low-permeability cover provided by about 0.5 m of highly compacted silty clay soil, the upper two thirds of which were amended with bentonite. In contrast to the Shiprock cover, this one provided a considerably more efficient radon barrier. The design also included covers for protecting the radon barrier against erosion, frost, and infiltration.

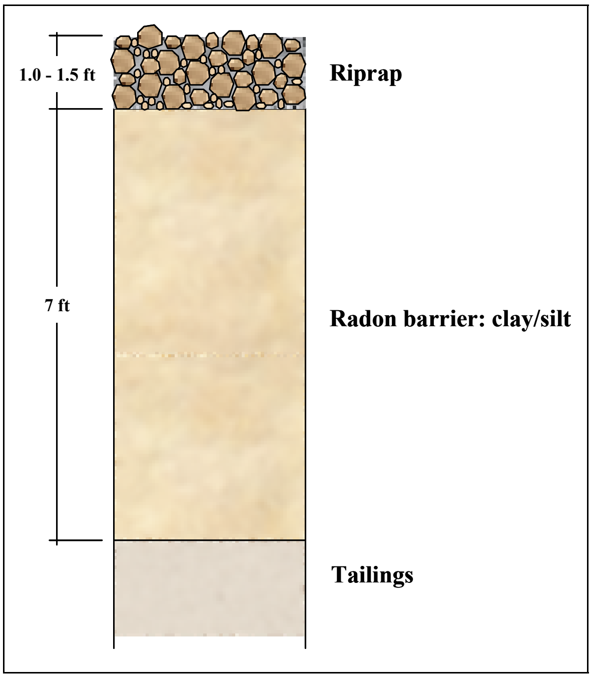

Fig. 10. Cover design for the UMTRA Project Shiprock disposal structure, New Mexico, after [111].

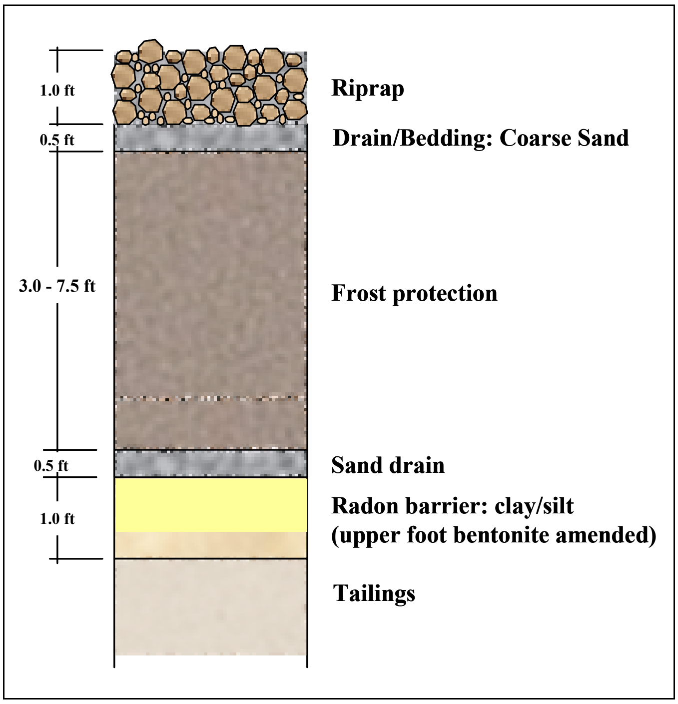

Fig. 11. Cover design for the UMTRA Estes Gulch containment structure, Colorado after [111].

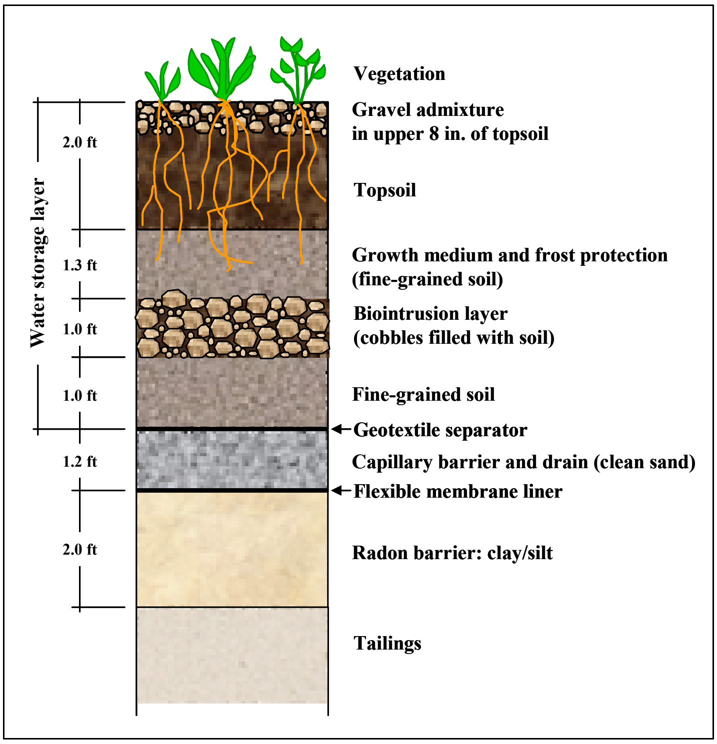

Fig. 12. Cover design for the UMTRA Monticello containment structure, Utah after [111].

A design shift in the late 1980s and 1990s resulted from the observation that ecological plant succession is inevitable and that continued intensive maintenance is impractical over extended time-scales. Fig. 12 shows a design that takes these factors into account [112]. The multi-layer cover design combines fundamental ecological principles with engineered barriers. The capillary barrier under a thick soil 'sponge' mimics the natural soil profile, in which thick loess stores precipitation that is eventually lost through evapotranspiration, thus maintaining unsaturated conditions in the subsoil. The cover is also designed to control radon flux, bio-intrusion and erosion, and to protect critical interfaces from frost. Studies of natural analogues suggest that the cover performance is likely to improve over the 1000-year design life [113].

A key theme is that whilst the same general principles should be applied, designs must be site specific and based on data collected at each site. This concept appears to widely held (see Annex XIII). Experience in the Western USA has shown that vegetative covers are not always viable in arid climates. This could be overcome with a multi-layer cover that addresses the objectives defined above, or by building a more traditional design using a compacted clay layer covered by rock.

Some caps have been in place long enough for useful data to be available on their performance (e.g. Beaverlodge [114]). Thus the development over time of a capillary barrier was assessed after a twelve year operational period [115]. Some ironoxyhydroxide precipates originating from acid drainage were found, but these did not reduce permeability significantly. but the issue of long term stability is still a major concern.

The design life of UMTRA structures is 1000 years. However, satisfactory containment may be required for time-spans between one and two orders of magnitude greater than this in order to guarantee effective isolation until levels of radioactivity have declined sufficiently.

Attempts are being made to quantify longevity of structures through estimations based on short term testing [116]. Estimation of proposed landform stability in the very long term is based on modelling, such as the modelling conducted for the Ranger mine constructed landform [117]. The results can help to identify areas of poor performance, which insight then is fed back into design adjustments.

Cover performance is not only of concern in the context of uranium mill tailings, but in any near surface repository and landfill design. In the US, a programme to compare different cover design under different climatic conditions was initiated [118].

5.5.9.3. Design Criteria

The design criteria for covers are related to the remediation objectives, and address geotechnical, radiological, hydrological, geochemical, ecological and aesthetic requirements. In general, covers comprise multiple layers, each with a specific function. For example, clay layers are typically used to control radon emanation and water infiltration. Vegetative covers control wind and water induced erosion and moisture infiltration by encouraging evapo-transpiration. Coarser material is used for moisture storage, as drainage layer and capillary break, and to discourage animal and human intrusion. A large body of information exists relating to the design of municipal landfills in various countries, e.g. [119], that may be indirectly applicable to uranium mill tailings closure and remediation.

— **Longevity** – Longevity is an important consideration in that covers may need to be engineered for life spans of the order of 200–1000 or more years. Erosion resistance is the key feature here. The other funcitionalities listed below must also be retained over the design life.
— **Sealing and shielding function**s – Gamma ray shielding can generally be achieved with a 0.5 m soil layer. To control dust, a vegetative cover based on an adequate soil layer as a plant growth substrate can be applied. A rock cover can also serve the purpose of eliminating the fugitive dust problem. Radon emanation typically is controlled by the application of a compacted clay layer and a reletively thin layer of compacted soil appears to be sufficient. Radon diffusion modeling assists in deriving the relevant design paramenters and material properties. The resistance to damage in freeze-thaw cycles may need to be investigated carefully.
— **Water infiltration** – Infiltration of atmospheric precipatation can be managed by the proper design of the sealing layer. The nature of the sealing material, moisture and density of the material at placement, and placement techniques are of upmost importance for achieving design criteria. Typical saturated hydraulic conductivity specified for sealant layers are of the order of 10^{-9} m/s or less. The performance can be assessed by evaluating the hydrologic water balance (Fig. 13). Laboratory tests are generally not representative of true field conditions and can produce results that differ from field conditions by as much as two orders of magnitude. In general, field (lysimeter) measurements are a more appropriate method for parameter, as hetero-geneity in permeability distribution is a function of scale. To achieve the necessary low permeabilities, sealant layers have to be compacted to obtain maximum dry density at optimum moisture content, to typically meet or exceed the 95% Standard Proctor density, e.g. [120]. These criteria are best demonstrated by the use of field test sites.

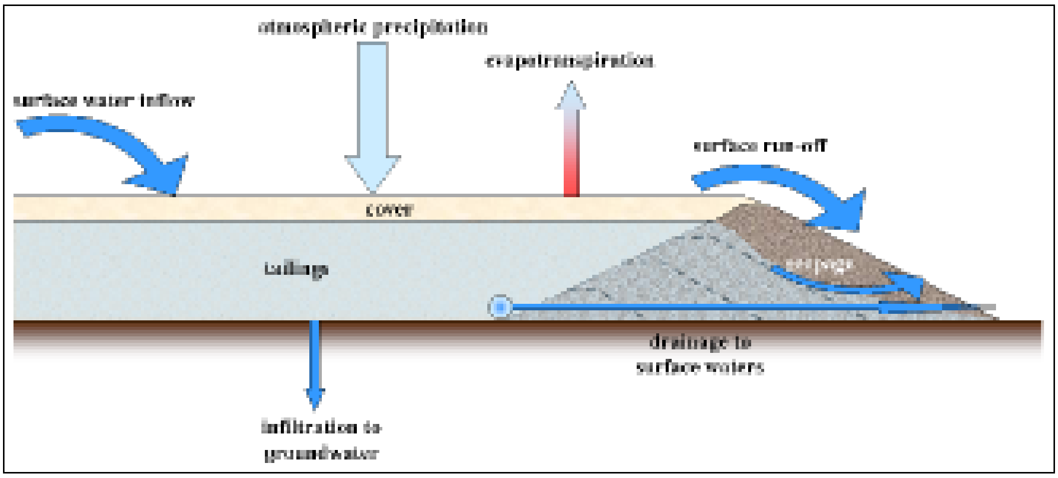

Fig. 13. Water balance in a covered tailings impoundment.

- **Gas Infiltration** – Design criteria aiming at limiting water infiltration and radon emission will generally also effectively control the inflow of gasses. It is important to prevent the infiltration of oxygen into tailings, as this will result in oxidation reactions that could lead to acid generation and, hence, mobilization of contaminants. The introduction of oxygen dissolved in water is generally a more important mechanism than gas infiltration.
- **Erosion prevention** – Erosion can be prevented by surface countouring that reduces and breaks slopes, the use of rip-rap or cohesive clay layers, and the development of vegetation covers. Vegetation covers provide good protection against erosion, but require site specific research to establish, which plant ecosystem would likely to be sustainable under the given conditions. Some supportive measures, such as application of polymer coatings, maybe needed to help establish the vegetation cover. Natural analogue studies provide a valuable tool for predicting long term erosion impacts.
- **Water storage layer** – A water storage layer, consists of multiple layers above the sealing layer, that act as a ‘sponge’, adapting to weather cycles. *Inter alia*, this layer prevents the drying-out of any sealing clay-layers. Drying cracks in the sealing layers would compromise their retaining capabilities for gases and infiltrating waters. In order to obtain the desired properties, this layer should be at least 1.5 m thick [120].
- **Bio-Intrusion** – In many areas, burrowing animals or deep roots can penetrate the sealing layer resulting in a loss of functional integrity. Provision can be made for the protection of the integrity of the sealing layers. For instance covering layers can be made thick enough and layers of rip-rap introduced that tend to discourage burrowing animals. Such layer, if thick enough, would also help to discourage humans from digging-up the tailings material.

5.5.9.4. Potential failure mechanisms

Inadequate design and poor implementation can result in the failure of tailings covers. Longevity objectives depend on adequate care and maintenance. Possible causes of cover failure include:

— Differential settlement
— Dessication cracks
— Bioturbation
— Root penetration
— Human and animal intrusion
— Extreme weather events
— Changes in the design base (e.g. climate changes)

Cover design may consider these factors in a probabilistic approach. Where possible, engineering design intends to prevent such events, but a certain amount of care and maintenance is likely to be inevitable [121]. With recent extreme events of flooding in Europe and elsewhere, long-established design parameters, such as maximum rain intensities and rainfall- function for catchment areas have come under scrutiny.

6. OUTSTANDING ISSUES

6.1. Overview

The main issues of public concern regarding mill tailings containment are the risks of catastrophic failure and continuing discharges that may lead to loss of life, property, infrastructure, to detriment to the environment and public health. The overall trend in the frequency of catastrophic failures has been downward over the past decades, although the larger sizes of newer impoundments mean that catastrophic failure may have potentially greater impacts than previously. No catastrophic failures have been reported for purely uranium tailings impoundments for more than 20 years. On the other hand, chronic releases of radon, dust and contaminated water and the associated potential health impacts and environmental contamination are a concern at many sites around the world.

The underlying causes for chronic release have been the subject of much research, as have been applicable methods for remediation. There is a large body of information available, focussed on major uranium production centres in western countries. Table 6 at the end of the chapter sets out the alternatives for uranium mill tailings containment as discussed previously, and attempts to evaluate the advantages and disadvantages of each method. Similarly, Table 7 sets out the alternatives for uranium mill tailings isolation and stabilization discussed in Chapter 5.8, and makes observations of the advantages and disadvantages of the different approaches and techniques.

It should be noted that one major driving force behind technological developments are tighter legal requirements and regulations.

6.2. Issues relating to the physical properties of tailings

The major root cause of catastrophic failure and ensuing dispersal of tailings material is the poor settling behaviour of tailings. Consequently, much research and technological development focuses on effective and cost efficient methods to dewater tailings either before or after impounding.

The physical properties are closely related to the chemical properties of tailings, as these typically constitute a finely dispersed, gel-like system.

(1) Research into the (site specific) dewatering behaviour of tailings with a view to improve dewatering is needed.

Table 6. A comparison of the advantages and disadvantages of the different approaches to uranium mill tailings containment [compiled by S. Needham].

Disposal option	Advantages	Disadvantages
Above ground	> Can operate simultaneously with mining > May be cheap to establish if tailings used in construction > Valley fill sites may have low construction costs > Whole tailings can be contained > Tailings pond can also function as evaporative pan to assist in mine water management > Most widely used > Tailings easily accessed for reworking if required	> Authorities may regard this type as only temporary storage & tailings may need to be relocated e.g. below ground level at end of mine life > May require construction of associated structures to minimise risk of environmental impact in the case of failure, or to collect/treat seepage etc > Seepage control essential > Expensive if built as water containment structure > Post close-out settlement may take a long time and lengthen period before operator can be released of responsibility > May need long term maintenance > Long term risk of tailings spill, increasing as structure weathers and erodes > Increases land area impacted by mining > Airborne and waterborne dispersal of contaminants possible following erosion etc
Below ground: in pit	> Very long term containment possible > Unlikely to ever require maintenance > Whole tailings can be contained > Pit preparation costs unlikely to be as high as above ground options > Airborne dispersal of contaminants effectively impossible > Structural failure of containment virtually impossible	> May need pervious-surround work to minimise ground water contamination risk > Construction cost of impermeable containment could be high if suitable pit not available > Not normally possible to operate simultaneously with mining at the same location > Requires a suitable pit to be available pre-mining, or for all ore to extracted prior to milling (e.g. Nabarlek) > May involve double-handing of tailings if no pit available at commencement > Re-claiming of tailings if required for further treatment will be difficult owing to depth
Below ground: underground mine workings	> Very long term containment possible > Unlikely to ever require maintenance > Can possibly incorporate whole tailings > Can be operated simultaneously with mining > Airborne dispersal of contaminants effectively impossible > Structural failure of containment virtually impossible	> Slimes may need to be contained separately > Need suitable groundwater conditions > Mine waste water management system needs to be able to cope with evaporation requirements > Tailings not available for reprocessing
Below ground: purpose-built containment (underground void or surface pit)	> Very long term containment possible > Unlikely to ever require maintenance > Whole tailings can be contained > Can be operated simultaneously with mining > Airborne dispersal of contaminants effectively impossible > Structural failure of containment virtually impossible > Site can be selected in low-permeability country rock > Benign rock available for unrestricted use in construction	> Construction required before milling commences > Mine waste water management system needs to be able to cope with evaporation requirements > Suitable site may be remote from mill and increase slurry/paste transport & infrastructure costs > Paste stabilization normally necessary for underground and optional/preferable for pit.
Deep lake	> Can operate simultaneously with mining > Cheap to establish > Whole tailings can be contained > Very long term containment possible > Unlikely to ever require maintenance > Whole tailings can be contained > Airborne dispersal of contaminants effectively impossible > Structural failure of containment virtually impossible	> Authorities may not allow this approach to tailings disposal > Requires nearby water body not otherwise used for social or economic benefit (i.e. fishery, water supply, recreation) > Risk of water contamination and tailings redistribution from disturbance by major flood of changed climatic conditions

Table 7. A comparison of the advantages and disadvantages of methods for stabilizing and isolating uranium mill tailings [compiled by S. Needham].

Methods	Advantages	Disadvantages
Containment preparation:		
Low-permeability membrane	High short-medium term security against seepage; easily applied to floor and walls of surface containments.	High cost; prone to accidental damage; difficult to repair after tailings discharge commenced; unknown long term performance; application limited to above ground containments; cannot be retrofitted.
Clay seal	Permeability decreases with overloading; low cost if local material available.	High cost if local materials unavailable; application limited to above ground and in-pit containments; cannot be retrofitted.
Grout	Targets known weak zones; ease of application.	Misses unknown zones of weakness; grouting compound may degrade on interaction with tailings pore water, cannot be retrofitted; possible high drilling costs involving specialist equipment
Permeable surround	Potential long term high security around entire containment against groundwater contamination; low maintenance.	Suitable high-permeability material needed; may clog; application limited to below ground containments; cannot be retrofitted. High cost (but distributed though operational phase)
Under-drain/basal filter bed	Aids settlement.	May clog; cannot be retrofitted; water disposal must be accounted for
Tailings preparation:		
Neutralization	Reduces acid producing potential and mobility of U and other heavy metals.	Availability and cost may affect viability
Thickening	Reduces water content of tailings; useful in low evaporation regions to achieve water loss.	Increased pumping costs; or alternative placement techniques.
Paste	Allows addition of compounds to significantly improve chemical and physical stability of tailings pile and containment.	Present use limited to underground containments; longevity unknown; tailings probably not recoverable. *[note: area of considerable development potential, research results discussed in Chapter 7].*
Cycloning	High stability product; provides sand-grade material for construction; removes main contaminants.	Slimes disposal
Tailings discharge and deposition		
Slurry beaching	Increase stability of embankment walls; easier ponding and collection of decant water.	Not applicable underground
Thickened tails placement	Improved immediate surface stability and access; improved control of placement (e.g. layering)	Higher transport costs (pumping or trucking)
Dry cake placement	High immediate surface stability.	Higher transport costs (ie trucking).
Barrier/reactive layers	High control over design and placement; permits tailoring to address variations within tailings pile etc; potential wide range of treatments available.	Suits only paste or dry cake deposition of tailings; high placement costs (pumping or trucking), largely untested technology *[note: area of considerable development potential, research results discussed in Chapter 7].*
Tailings dewatering at discharge:		
Thickened tailings	Reduce water content problems, especially in low evaporation areas. Reduced use of clean water.	Higher transport costs (pumping or trucking) compared to slurry
Paste tailings	Reduce water content problems, especially in low evaporation areas; allows addition of chemical and physical stabilisers.	Long term performance and effects on diagenesis etc unknown
Dry cake	Removes water removal and stability problems, significant reduction in leaching and groundwater contamination risks; potential for seismically active, cold, arid regions. Reduced use of clean water.	Untested in uranium mill tailings
Tailings dewatering in-situ:		
Under-drains	Improves settlement density, reduces instability and chemical contamination.	Cannot be retrofitted; may clog.
Horizontal drains	High improvement to settlement density, reduces instability and chemical contamination; design may be modified during operation.	Cannot be retrofitted; may clog.
Wells/jet pumps/electro-osmosis	Improves settlement density, reduces instability and chemical contamination; can be targeted to specific sites. Suitable for *ex-post* treatment.	May require closely spaced grid of wells with attendant higher costs (see also Table 4).

Methods	Advantages	Disadvantages
Evapo-transpiration	Some improvement to settlement density, reduces instability, inexpensive.	Restricts access to tailings surface; of limited benefit once supernatant liquid evaporated unless used in conjunction with wells or wicks; no removal of contaminant load.
Sand drains/wicks	Some improvement to settlement density, reduces instability, inexpensive and low maintenance. Suitable for *ex-post* treatment.	May take many years to reach dewatering capacity. Surface loading speeds it up.
Tailings surface treatment		
Water cover	Limits radon flux, dust	Negates dewatering as an option to improve settlement density. Must be removed and followed by dewatering/settlement works prior to remediation.
Wetting	Reduces dust and radon flux; assists in evaporative water loss.	Components need regular inspection, maintenance, and switching/moving.
Sealants	Reduces dust, infiltration, improves access to tailings surface; good temporary protection between depositional or remedial stages.	Suitable only for short term protection.
Decant water treatment		
Recirculation to mill	Reduces clean water uptake, low cost, suits no-release criteria.	Possible build-up of agents may decrease mill circuit efficiency.
Treat and release	Contains contaminants within the mine system.	Public perception of uncontrolled environmental harm; probable environmental impacts in event of system failure.
Seepage control		
Seepage detectors	Provides early warning and allows early intervention in event of significant seepage	Costly; in themselves do not treat the problem; difficult and expensive to retrofit.
Collector wells	Targets known confined seepage zones.	Limited sphere of influence; unlikely to collect seepage from unknown/new seepage points; requires monitoring wells to validate performance.
Interception drains	Minimises transfer of contaminants to groundwater or surface water systems; can extend around perimeter to provide high levels of assurance; essential backup for all operating tailings facilities.	Requires monitoring wells to validate performance.
Return to containment	As for decant water treatment	Essential
Treat and release	As for decant water treatment	Essential
Recirculate to mill	As for decant water treatment	Essential
Groundwater monitoring	Provides essential information for effective water management during operational phase	Not practical for long term management/stewardship post closure
Capping		
Engineered design	High security short term (<1000 years); well-known technology; extensive experience, guidelines, geotechnical expertise.	Unknown long term (1000–50000 years) performance; probable high future maintenance costs; incompatible with natural processes, forces and systems; poor aesthetics.
Ecological design	Caters for/conforms with natural processes; enhances prospects for long term security against environmental harm; probable low future maintenance costs; good aesthetics	Unknown technology and costs; possible long lead time for research to deliver sufficient site specific knowledge to factor into design.

6.3. Issues relating to containment

A recent review of risk assessment and contingency planning in the management of mill tailings concluded that there is no such thing as 'fail-safe' facilities for tailings management [82]. Neither regulations, design specifications, nor management systems can be relied upon in isolation to provide assurance against containment failure: all three must be applied, in a framework of quality assurance and post-closure care and maintenance, to deliver a high probability of tailings containment security. Examples exist of failure related to containments not being built as designed; regulators not checking that all requirements were provided for in construction and operation, and worst-case scenarios not being taken into consideration in deriving design specifications. It is therefore critical to ensure that:

(2) *containment design is based on comprehensive, site -specific risk analysis;*

(3) *containment construction follows design specifications rigorously;*

(4) *appropriate operating procedures, coupled with quality assurance are adhered to, including a regulatory system that checks that all design and operational requirements are applied during construction, throughout operational life, remediation, close-out and during the stewardship stage.*

There is much information in the literature on the high cost of remedial works undertaken at areas contaminated by past practices in uranium tailings management, in particular from the UMTRA programme in the USA, remediation activities in Germany, other European countries and Australia. It is clear that in cost terms, prevention of contamination is much more effective than cleaning it up afterwards. This is corroborated by experience from other mining industries. In relation to the general performance of uranium mill tailings containments, the main concerns relate to longevity of containment, and seepage to groundwater. On the other hand, current knowledge and practices relating to radiation control, dust, and security can be considered generally satisfactory. Areas for consideration in relation to longevity of containment and environmental impact via seepage are:

(5) *a comprehensive site specific risk analysis that includes an assessment of likely environmental impacts for both the operational and post-operational stages;*

(6) *a containment design that incorporates components designed specifically to prevent environmental impact to the highest level achievable during the operational life of the containment within reasonable cost;*

(7) *a choice of the type of, and site for, the containment that considers the long term risks and costs. For example, higher tailings slurry pumping costs to take advantage of an old pit in the district may be preferable to construction of a dam adjacent to the mill where seismicity, available construction materials, surface water flow etc may represent significantly higher risk of failure during the life of the containment.*

(8) *design approaches that develop concepts that offer confidence beyond a 1000 year design life provided through conventional engineering design. Such concepts of containment performance and containment life may include features that enable natural processes to interact with the containment and the tailings within it in a way that improves long term stability rather than diminishing it (ecological design).*

6.4. Issues relating to tailings chemistry

Whilst some research into tailings chemistry, and considerable research into fixation of uranium, is being undertaken, the knowledge of tailings chemistry appears to be incomplete, and therefore, the potential environmental impacts of tailings water and its mixture of heavy metals and other toxic compounds are not fully understood. There is little acknowledgement in the research literature published in the uranium sector that non-radioactive constituents, particularly heavy metals, arsenic and organics may have a comparable or greater impact than uranium and its progeny. The non-uranium mining industry has identified acid mine drainage as the major environmental issue it faces, and major world-wide initiatives have been developed to organise research into mitigating the problem. Clearly, removal of the acidity problem reduces the mobility of metals, including uranium, and significant environmental problems can be avoided. There is valuable information available in non-uranium mining research that could be beneficially transferred to the uranium mining and uranium mill tailings area. More knowledge is required on the combined chemical behaviour and toxicity of the total chemistry of uranium mill tailings effluents.

(9) *There is significant knowledge coming to hand about methods to reduce the chemical mobility of uranium that focuses on application after deposition, for example as barriers or as agents to assist in effective capping. Whilst some attention has been given to the possible application of this technology at or before tailings deposition, the opportunities should be explored more fully in order that seepage is less likely to be generated in the first place. A key factor in the generation and mobility of tailings water is obviously the water content of the tailings. Significant research progress is being made into thickened tailings and paste technology to generate low-water and even dry tailings. These methods are now widely applied in non-uranium metal mining, but cannot easily be applied in retrospect due to high costs. Attention may need to be given to assessing the suitability of several technologies for tailings stabilization (water contents reduction, cementation, various chemical fixation techniques), either before, or during placement into the containment.*

Lack of knowledge on the evolution of the tailings body is a significant limitation on modelling of long term stability and performance of the containment. Little is known of the controls on and effects of diagenetic reactions, the stability of new mineral parageneses, and their long term evolution and performance. For instance the porewater pH and Eh may change over time resulting from successive mineral parageneses. The solubility of metals, including uranium is governed by the Eh and pH conditions within the tailings pile. Diagenetic processes will also be governed by reaction kinetics. The role of biological processes, for example the reduction of sulfate by bacteria, may also need to be considered.

(10) *a better understanding of the long term mineralogical changes within uranium mill tailings would allow the design of chemical treatment likely to promote desirable processes that could, if cost-effective, be used to enhance the stabilization of tailings impoundments.*

6.5. Passive systems

During remediation and after final closure, low levels of environmental contamination may still continue to exist. Passive technologies may be employed to deal with continuing seepage and residual groundwater contamination. These systems are being increasingly used at many uranium mines and tailings facilities to treat effluent by using the assimilative capacities of various naturally occurring processes [106]. They include adsorption onto soil particles, absorption by certain minerals and organic compounds, uptake by vegetation, uptake and fixation by microorganisms, and (co-)precipitation as various mineral species- often as iron compounds. Research into the processes at work is extensive, as the potential for these 'bio-reactors' is being widely pursued by all areas of the mining industry, as well as other industry sectors such as mineral processing, chemicals fabrication and abattoirs.

Passive systems range from engineered systems, such as reactive barriers, through enhanced natural wetland systems, to a reliance on natural attenuation processes [106]. In most cases, these are not walk-away solutions, but require a detailed understanding of the physical and chemical systems involved, adequate care and maintenance and long term monitoring to verify performance objectives [122]. It is worth noting that contaminants attenuated in passive systems may be re-released by changing environmental conditions and that the removal of contaminants from seepage results in the creation of new contaminated solids. These may require later remediation themselves, if not disposed of safely. These systems are generally applied as combinations with other passive or active components.

The use of the term ‘passive systems’ reflects the hope that the level of intervention required at closed sites in order to achieve adequate levels of environmental protection would be minimal or none. However, there is little information available on the sustainability and time-dependent effectiveness of these natural processes. It is likely that the capacity for such systems to continually take up contaminants has a finite limit. ‘Natural analogue’ studies as have been undertaken in the context of geological disposal of radioactive wastes [113] [123] may also hold a clue for the long term behaviour of tailings impoundments.

Current thinking is moving to the concept of ‘minimal intervention’ that recognises the likelihood of a finite assimilative capacity for natural systems. A wide range of these questions was discussed for instance at recent conference in a dedicated session of the ustilization and limitations of natural attenuation as a long term environmental protection approach [124] [125] [126] [127] [128] [129] [130] [131].

Concepts for the acceptable isolation of uranium mill tailings must, in view of the very long time frame involved, accept the inevitability of interactions between the tailings pile and its containment with the natural environment. Indeed, the concept of the ideal tailings isolation arrangement is one, in which the physical and chemical processes in the tailings eventually make use of natural attenuation processes so that nature is harnessed to improve stability with time. The concerns for long term stability and likelihood of failure, based on conventional engineering methods, are thereby reduced. ‘Natural forces’ become an assistant to longevity instead of its enemy; the overall design concept becomes one based on harmony with nature.

(11) Studies into natural processes to assimilate effluent from tailings piles may be promoted to determine their capacity to improve stability over long time frames, and thus provide feedback into improved physical and chemical systems designed for present-day approaches.

(12) It may be worthwhile to keep an open mind for the possibility of future impoundment design and placement techniques that may harness natural processes to reduce long term risk of failure and environmental harm by effectively assimilating the tailings pile and containment into the environment.

7. NEW APPROACHES AND RESEARCH

7.1. Overview

A large proportion of research relevant to management of uranium mill tailings has been produced since the early 1980s mainly in the USA, Spain, France, Canada, and Germany. In all of these countries, the government recognised the potential health risks posed by improperly closed and unremediated uranium tailings and stimulated research towards solutions, passed legislation, and allocated funds to enable clean-up to begin. A comprehensive and detailed review of the level of knowledge of uranium tailings management developed in the Canadian National Uranium Tailings Programme is given by [8]. During the same period, new major uranium mining and milling operations were opened. Through their licensing and operational requirements, new methods of tailings management had to be developed, based on extensive research in the uranium and non-uranium mining industry.

There are, therefore, two important sources of information on new approaches to uranium tailings management. These stem from the remediation of old tailings impoundments and from the development of new mines and mills. The general trend of these approaches is towards site specific solutions, based on risk assessment and with the objective of long term isolation of tailings from the environment, utilizing natural processes as far as possible.

7.2. Design and siting of containments

Research in this area is commonly undertaken through a risk assessment framework that considers all reasonable options with the objective of selecting the preferred alternative to minimise the probability of failure and environmental harm. Examples include general principles in risk assessment and design [132], developing site specific risk assessment methodologies (e.g. [133]), and selection of the lowest-risk alternative for a specific region, such as concluding that subaqueous deposition is best for decommissioning at facilities at Elliot Lake in Canada [94].

Another line of research concerning design and siting is the development of methods and models for prediction of long term stability. One area of investigation is the development of short term testing techniques from which reliable long term predictions of stability can be made ([116], using UMTRA repositories as the study sites). Long term column tests also provide data on which to base long term predictions [134]. Another is the development of computer modelling based on ground-truthed parameters, such as regional erosion rates, present and likely future climate conditions, and ground surface conditions measured on waste rock dumps and natural analogue sites. This approach has been applied to estimate longevity and likely failure points for the Ranger mine landform contained in the mine closure plan [117] [135]. The concept of ecological design of tailings repositories is advanced for the uranium mill tailings deposits in Germany for reasons of reduced risk of failure, lower maintenance requirements, and harmonizing with natural processes [136]; this work concludes that inherently stable landforms are the way of the future in tailings repository design.

Research is also continuing into the effects of vegetation development and root penetration on the stability and longevity of covers. Lessons learned from UMTRA research and observation show that encroachment by vegetation onto covers is inevitable [137]. Issues such as vegetative encroachment on rock covers may have to be managed under long term care and maintenance [121] until a better understanding of these issues has been achieved.

In humid climates, vegetation will develop in any uncontrolled area. Vegetation is used for erosion control and the management of the water balance within covers. Root penetration may however pose a problem. Since root shape and penetration can have a major influence on water balance, a large amount of experimental work has been undertaken in this field world-wide. Extensive research has been carried out to investigate the natural encroachment of vegetation onto barren sites. This will give an idea of the possible plant succession on tailings impoundments. Planning of vegetation covers aims to achieve a sustainable vegetation cover, relying on predicted natural succession, rather than on active management. Vegetative cover designs normally build on indigenous plants as far as possible, and try to avoid becoming a source of invasive alien vegetation, following the principles of ecosystem sustainability.

7.3. Physical stabilization

Research into physical stabilization can be broken down into issues that deal with consolidation of existing tailings piles, enhancement of settlement rates and densities, application of paste technology to uranium mill tailings, and developments in paste technology and grouting systems.

7.3.1. Consolidation

Consolidation occurs naturally in existing tailings piles, leading to a decrease in volume and an increase in geomechanical strength. The consolidation rate generally decreases exponentially, ultimately becoming very slow. Consolidation rates are dependent on pile thickness, grain size distribution, (initial) water content, and pore pressure or overburden. It may be enhanced by dewatering and overburden material.

Tailings settlement/consolidation studies include site specific studies at Ranger mine, Australia, in Thuringia, Germany and Saskatchewan, Canada. At Ranger, high-energy peripheral discharge of tailings (*i.e.* they cascade down over benches from a pipe at the top of the pit wall) results in rapid segregation of the coarse fraction near the pit wall on one side, such that the centre of the pit is filling with fine tailings and slimes [89]. Similar situations, where deep piles of very fine, low density tailings have been allowed to accumulate in pits have caused serious problems for decommissioning through very long settlement periods and excessive differential settlement of the surface [138]. A different tailings discharge system has been recommended for the Ranger pit, similar to the Deilmann pit system [139], that aims to limit segregation, enhance consolidation, while achieving a low hydraulic conductivity.

In Thuringia, laboratory tests have been undertaken to determine hydraulic conditions within Wismut tailings [104] so that consolidation modelling (applying non-linear finite strain consolidation theory) could be refined. Thus estimates of settlement rates and the time taken to achieve desired settlement densities can be improved. This information can then be used to parameterise techniques such as grid spacing of deep wicks, or surface loading rates, to achieve the required settlement performance within the five-year target time. Comparison of modelling results and observed tailings profiles in the Culmitzsch impoundment suggests that the degree of consolidation is sensitive to the thickness of the slimes zone, and not to tailings pile thickness [140]. The settlement of fine tailings cannot be predicted using the consolidation theory of Terzaghi, which is applicable to homogeneous, soil-like tailings only. The behaviour of fine material depends on water content under a time dependent load, and finite strain models are necessary [27] [141]. The 1-D model of Schiffman has been further developed by Wismut to a 2-D model that can be applied to real-world 3-D problems [140].

At the Key Lake and Rabbit Lake facilities, Saskatchewan, Canada, adequate consolidation was not achieved due to formation of ice within the tailings during winter. Various techniques have been used with partial success to address this problem (see Annex II).

The material type and the deposition method produce different patterns of heterogeneity within tailings impoundments. Accurate mapping in three dimensions is therefore necessary to predict the settlement behaviour within a tailings impoundment.

Total settlement of a tailings pile can range from a few percent up to 30%, over varying periods of time, but generally less than five years (see Fig. 14 for an example). Most settlement occurs in the initial years. Typically at least 90% of the settlement should have taken place before a cover can be installed. The progress of settlement can be monitored by surveying settlement plates. Settlement can be accelerated by loading with a surcharge and various dewatering and compaction techniques.

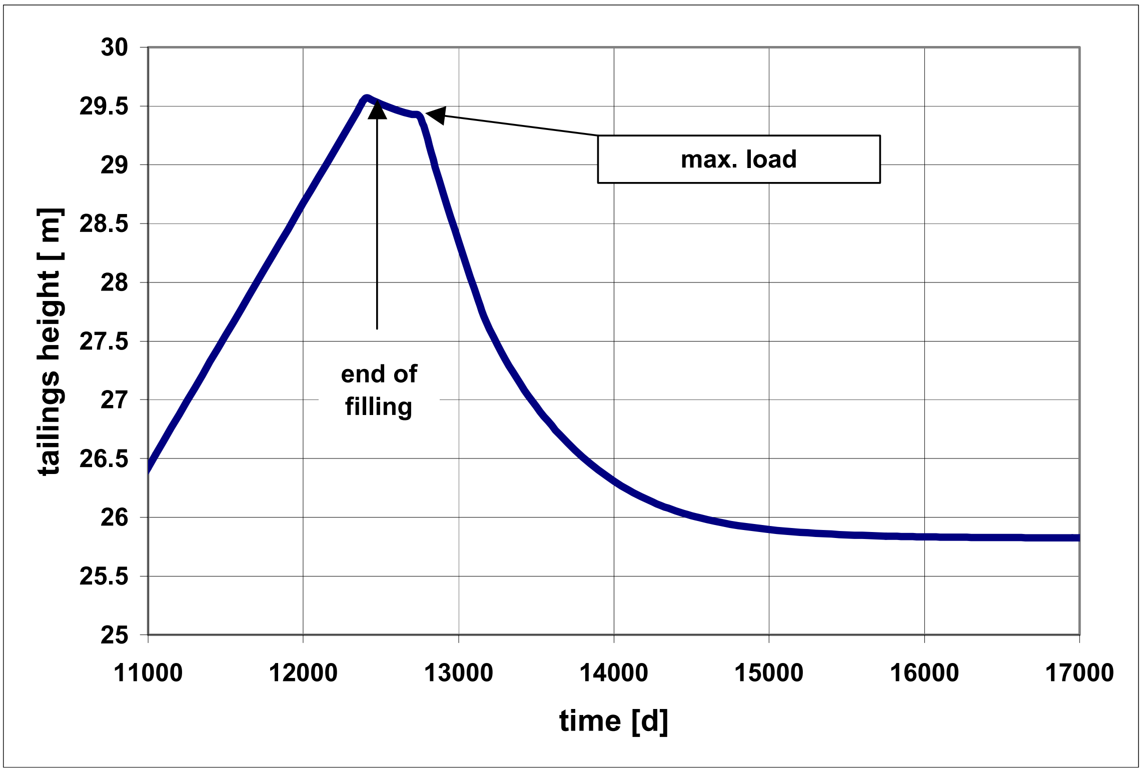

Fig. 14. Evolution of settlement of fine slime tailings under a surcharge of 300 kPa (ca. 15 m cover). It is assumed that the surcharge increases lineraly from 0 to the maximum over 1 year. Central part of Rožná K1 tailings pond (see also Annex IV).

Other research includes the addition of material to increase consolidation rates [142]. For instance, shredded plastic is used to improve permeability and speed up settlement.

7.3.2. Paste technology

Research into paste technology is substantial and created considerable interest in the industry [143]. In very simple terms the production of tailings paste includes the thickening of a tailings slurry to a low moisture content to produce a pastelike material that is still pumpable. The moisture content must be low enough that esentially all pore fluids are held as colloidal moisture or by capillary forces. Synthetic flocculants are added to achieve rapid settling of dense fine solids aggregates, a deep bed thickener to promote self weight consolidation in the settled solids and dewatering channels to relieve the excess pore pressures.

There is, however, little research that relates specifically to paste technology in uranium mill tailings. Potential application of the technique was reviewed in relation to the recommencement of uranium mining and milling at Canon City, Colorado. It was found to satisfy the requirement of new state regulations that specify minimal free drainage under gravity from any tailings generated [144]. A study is underway in the Ukraine to evaluate the suitability of cycloned uranium mill tailings for incorporation into mine back fill paste (see Annex XII). The study aims to determine optimum composition of the backfill using Portland cement as the binder. Special considerations are the potential leachability leading to groundwater contamination, radon emanation rates and their implications for worker safety in operational parts of the mine, and impact of seismically-induced fracturing of the cemented backfill, which commonly occurs within 5–10 years of emplacement.

Research utilizing non-uranium tailings for studies into paste technology includes:

- Macroscopic and microscopic studies of cementiferous metal sludge during consolidation [145];
- Consolidation and flow characteristics of thickened tailings paste and implications for surface placement [99].

Useful reviews of developments in and use of tailings paste include:

- An assessment of the sustainability of high-density thickened tailings technology [146];
- Evolution of application of thickened tailings pastes in Australia – advantages of thickened tailings for surface central thickened discharge for remediation, including a description of stability, slope, run-off and infiltration characteristics [147];
- General discussion of advantages of paste technology for surface disposal of tailings, in terms of strength, permeability, slope characteristics, water balance, acid generation, stability and cost [148];
- The development and application of full tailings paste, incorporating slimes as well as coarse fractions- describes underground use, advantages in respect of seepage and evaporation, and the potential for use in surface tailings disposal [149].

Paste handling may pose radiological hazards. Research in this regard may be necessary before this technology can be adopted more widely in the uranium mining industry.

7.3.3. Grouting

Grout addition to tailings may have a dual purpose, to increase the physical strength as well as chemical binding, as discussed subsequently. A variety of grouts, both for injection into the ground or mixing on site have been studied in the past for improving the geotechnical properties of materials. In the context of waste disposal and environmental remediation this technology has found recent interest:

- Field trials into the ground injection of colloidal silica grout to create an impermeable barrier [150];
- Development of a proprietary dry spreadable or injectable compound (Envirobond™) for stabilizing metal-contaminated sludges via meatal complexing involving oxygen, sulfur, nitrogen, and phosphorus [151]; and
- Grout-like compounds to improve erosion resistance (Annex XI).

7.4. Chemical stabilization

7.4.1. Overview

Research into chemical stabilization of tailings or similar contaminated materials is very active and pursuing several fronts. This summary deals mainly with investigations into chemical fixation of uranium and other metals, and mitigating acid mine drainage. A few examples are also examined of research into interactions between uranium tailings and groundwater, as well as some recent work reviewing the geochemical characteristics of tailings, metal mobility, and implications for environmental impact.

7.4.2. *Geochemical impacts*

A general review of issues relating to chemical impacts from surface tailings containments looks at modelling trends, treatment technology and future directions in relation to the introduction in Australia of new national water quality guidelines that expect mine and other operations not perturb the chemistry of natural waterways outside the natural local range [152].

The different geochemical characteristics of tailings, related to variations in ore type and gangue mineralogy, salinity, acid producing potential, and degree of weathering are discussed in terms of their impacts on operational planning and long term management of tailings [153]. Enhanced mobility results from both acid and alkaline leach processing. The leachability and mobility of metals and other compounds from alkaline leach processing is discussed in relation to the Schneckenstein area in Germany [38] [154].

Modelling of aquifer systems is undertaken to reduce uncertainties in predicting future impacts and designing groundwater remedial programmes. The models are applicable to seepage from surface mines and tailings piles [155], as well as from underground mines, where the data can be applied to predicting effects from planned underground placement of tailings [156].

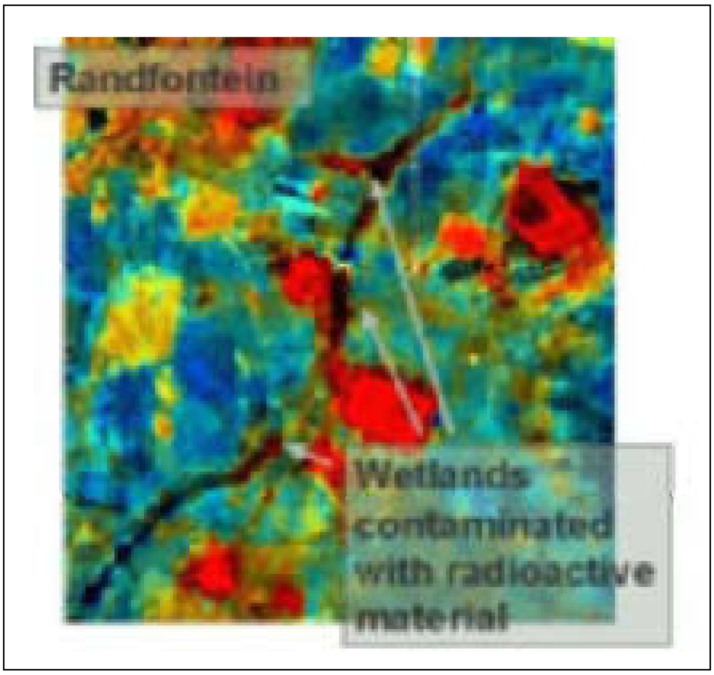

Fig. 15. Total count airborne radiometric image of a portion of the West Rand (South Africa) radionuclides are adsorbed onto sediments in wetlands downstream of tailings dams (courtesy H. Cotzee, 2002).

A study of geophysical conductivity and resistivity techniques to monitor acidic plumes from uranium waste rock piles has application in the tracking of contaminant plumes from tailings impoundments [157]. Airborne radiometric surveys (see Fig. 15) have been successfully applied to the mapping of physical and chemical transport of radionuclides from tailings piles in the near-surface and surface environment [158].

7.4.3. *Fixation technologies*

Research in this area aims to make contaminants resistant to leaching and mobilization, by converting them to stable mineral or organic compounds, or by binding (or 'micro-encapsulating') them in natural or man-made compounds that coat and protect them from leaching solutions, or by encapsulating them within an inert and impermeable barrier. These techniques variously have potential application as tailings thickener or paste additives upon tailings discharge and placement, as grouting compounds, as interlayers within the tailings pile, in *ex post* applications as injectable fluids, barrier surrounds, grouts, and as components of or additives to cappings.

Natural chemical fixation: in some cases there may be naturally occurring factors that promote chemical stability in the tailings. These should be investigated as part of the planning and design process for tailings management. Such effects have been understood for a long time, for example carbonates in a sandy aquifer at Elliot Lake, Canada, which retarded the development of an acid plume and caused uranium to precipitate from seepage water [159]. The study of natural (secondary) fixation processes is at the core of many natural analogue studies (see Chapter 4.3 in [106] and references therein).

There are many different approaches to developing applications that harness the fixation properties of natural materials, and can be broadly divided into development of binders, and development of barriers.

Inorganic binders involve the mixing of a compound with the bulk material to be treated, for example at the discharge stage for tailings slurry or pastes. Binders promote both physical and chemical stability, and research in this area is closely linked to work described in the section on physical stabilization. The most widely used binder is Portland cement, because of its ease of availability and handling, low technology requirements, relatively low cost and well-understood cementing and neutralizing capacity [160]. Another material with substantial promise is fly-ash, used to induce cementation and reduce permeability through the precipitation of secondary minerals [161]. The application of silica to tailings to coat particles – 'micro-encapsulation' has been proposed as another potential form of binder [162].

Mixing ratios of cement to waste, and water content, can significantly affect strength, leaching resistance, and cost of application. Optimal ratios for use of cement as a binder in low-level mixed waste is found to be 0.5–2.0 kg/kg cement and 0.3–0.33 kg/kg water [163], but these ratios are prohibitive for treatment of the large volumes of tailings. The physical properties of a range of cemented tailings backfill mixtures studied in the Ukraine showed an approximate increase in compressive strength (from 3.0 to 7.0 Mpa) with cement content (from 200 to 400 kg/m^3), but performance varied between different tailings types (see Annex XII). In studies and review of the literature for the Jabiluka Environmental Impact Study, it was noted that there is no quantitative data available on the leaching characteristics of uranium mill tailings under buried saturated conditions, and that flow rates and adsorption behaviour of specific contaminants in contact with moving groundwater is unknown [95]; the mine proposal provided for the addition of 1–4% cement to the tailings destined for underground backfill. This was expected to reduce permeability from 10^{-7} to 10^{-10} m/s and significantly limit leaching of contaminants.

Other studies have shown the potential for inducing precipitation processes in order to reduce permeability and immobilise contaminants through the injection of supersaturated solutions. These solutions promote precipitation of gypsum, barium sulfate, calcium hydroxide, calcium carbonate and aluminium or iron hydroxides, which reduce permeability through crystallization in pore spaces, provided precipitation can be delayed through pH adjustment and/or addition of degradable polymers (see Annex XI).

Polymeric binders. Another area of relevant research is the development of inorganic geopolymers for immobilization of metals. The geopolymers are variously based on Al. K or Ca silicates, and are designed for mixing with wastes or encapsulation of the waste mass. Information is coming to hand on their specific application to uranium mining wastes, e.g. [164]. Tests show their superior performance in terms of high mechanical strength (attrition and compressive strength), resistance to leaching, fire and bacterial resistance, and durability relative to various Portland cements [165] [166]. Many mining/energy industry by-products containing aluminium and silicon could potentially be used in polymers for construction, ad primary aluminosilicate products such as kaolin, feldspar and mica could be used as reactants in geopolymerization.

Chemical barriers for fixation involve the placement of interlayers, surround ‘filters’, or selective positioning of prepared materials that will react with percolating water to prevent or limit the passage of contaminants beyond the barrier materials, e.g. [108]. The barriers exploit natural redox reactions to promote absorption and precipitation of mineral and organic compounds and complexes.

Natural redox reactions studied in Polish tailings demonstrated that iron hydroxide (goethite), haematite and gypsum are precipitated as solids and could be used to stabilise uranium and other contaminants if these reactions can be induced and controlled. A layered tailings/barrier system is proposed where the barrier contains CaO or $Ca(OH)_2$ and anhydrite to control vertical migration of sulfates and induce redox reactions and iron oxidation, leading to uranium precipitation and radium fixing in Ca, Ba and Sr sulfates, [167] and Annex VI.

Permeable reactive barrier systems containing zero valent iron have been installed for treatment of U and other metals [168] [169] [170] [171]. These barriers demonstrate excellent removal of U. Examination of the reaction products has been conducted at a series of the permeable reactive barrier sites [168] [172] [173]. Although the results of these characterization studies are inconsistent, all of the reports indicate that at a portion of the U entering the barrier system is reduced to U(IV), whereas some portion may remain in the U(VI) oxidation state. Other metals that are commonly associated with uranium mine waste, including As, Mo, Se, V, and Zn are also removed from the groundwater, possibly as reduced phases (e.g., V_2O_3) or as sulfide minerals (As_2S_3, ZnS) [169] [172]. However, iron corrosion products, and certain common elements in groundwater such a sulfate, chlorine and carbonate, have a negative effect on the immobilizing process. Humic substances have been shown to enhance the immobilization rate [174]. Brown coal admixed with iron has been shown in laboratory and operational trials to cause acid buffering, sorption and degradation of organic matter in association with sulfate reduction, increasing the effectiveness of immobilizing uranium as well as cobalt, zinc, nickel and other metals [131].

A proprietary metals stabilizing agent (ENTHRALL®) has been developed in USA in which the primary agent is calcium sulfide that is claimed to bond strongly to a range of metals to form insoluble metal sulfides [175]. The process requires less bulk material for treatment than conventional cement or lime-based systems, and has passed the US EPA’s Multiple Extraction Procedure that simulates 1000 years of activity. It is available in liquid, powder and granular solid form for a range of treatment alternatives. The granular product is to be tested at a proposed US DOE demonstration programme at the Mound Site in Ohio, where it will be added to an oil/mixed metals waste [176]. Gel enhancements to improve effectiveness of permeable reactive barriers have also been proposed [177].

Microbiological techniques have been used for more than a decade for the remediation of metal and oil-contaminated soils [106], and the potential for application in reducing the contaminating potential of mill tailings is evident. However, a significant question in the application of such techniques in permeable barrier technology for inorganic pollutants is the long term stability and fate of the contaminants immobilised in the barrier. Research to date has focussed on achieving immobilization, assuming that water quality standards would be met as a result of low solubility of the reduced phases. However, potential remobilization (for example through complexation or oxidation resulting from long term changes in groundwater regime) is a significant issue when planning for remediation or uranium mill tailings because of the very long term stability and environmental protection required. Some research has commenced in this area, which indicates that As Cr and Se form different phase types and that some types are available to leaching, although this is a small proportion of the total metals fixed by the permeable barrier [178]. The PEREBAR programme was initiated in the European Union to further investigate the long term behaviour of permeable reactive barriers, including possible changes to porosity and reactivity that may impair their long term performance [179]. Accelerated testing methods, increasing efficiency via chemical ligands, and potential for combining with electrokinetic effects are to be conducted and investigations are to include on site work at an old uranium mine in southern Hungary.

Redox-reactive barriers can also be constructed using dissimilatory metal-reducing bacteria that fix metals through enzyme-induced reduction, or enzymatically generated ferrous iron from naturally occurring ferric iron minerals [180]: microbially produced ferrous iron can reduce and immobilise metals and radionuclides as well as beneficiate material contaminated with chlorinated organics or nitro-aromatic compounds. The application of bacterial reactions was also investigated in the Mine Environmental Neutral Drainage Programme (MEND) to lower reactivity in sulfidic uranium mill tailings and reduce permeability, therefore impeding leachate flow paths and metal removal [181].

A variety of other in situ fixation techniques for groundwater contamination are available and these have been reviewed, for instance in [106].

The range of different approaches to chemical stabilization being pursued in current research world-wide offers many potential benefits for uranium mill tailings management, at the placement stage and *ex post* remediation in the tailings pile and of environmental contamination caused by the pile.

7.5. Encapsulation and covers

Much research information on the design and placement of covers dates back to the 1980s, and consists mostly of research work done under the UMTRA programme in the USA, or similar work in other western countries. Recent descriptions and review papers of the lessons learned, such as the evolution of thinking and practice in cap design during the UMTRA programme [109] are still relevant to work being performed in recent times in other countries on the same subject, such as selection of capping materials in China (see Annex III). In the absence of major seminal reports distilling all the major results of the UMTRA programme, review papers are very useful in ensuring the valuable information collected by the UMTRA programme are not forgotten. Collaborative projects between the USA and Canada, where most of the 1980s research was done, and other countries now developing remediation programmes for uranium mill tailings, are effective pathways for knowledge exchange: IAEA working groups bringing together representatives from many different countries to focus on the subject [7]; the Uranium Mining Exchange Group (UMREG) meetings held in conjunction with major conferences such as UMH III [204] [205] or ICEM'03; Canada/ Germany interaction through bilateral workshops [182]; European collaboration to assist former soviet block states under the PHARE programme [12].

New research into capping design etc. includes investigations into the feasibility of incorporating barrier technology for improved physical and chemical isolation and surface stability, and the reader is referred to Section 5.5.5 for discussion on these topics as they relate to capping design and practice. Information from other areas of environmental research into site remediation is generally applicable to the restoration of uranium mill tailings facilities, such as bioremediation of soils, and use of sewage products to promote revegetation [183].

The UMTRA work focussed on tailings piles that were mostly dry and mostly in arid environments. Therefore, there was less attention to issues relating to dewatering and stabilization of wet tailings in preparation for remediation. However, this is a major issue in many countries where uranium mill tailings are deposited as slurry in temperate, mountainous, taiga or tropical climates (Table 2). The reader is referred to the discussion in Section 7.3.1 relating to settlement and density of tailings. The difficulties being experienced at Ranger mine provide an excellent example of the problems and costs that can arise from low-density tailings in a tailings dam: capping trials [184] indicated that *in situ* capping would be cost-prohibitive and resulted in a decision to transfer tailings to a mined-out pit on the site. Fines and slimes are now pooling in the centre of the pit and indicate major dewatering and settlement complications in preparation for and during remediation unless tailings deposition techniques are changed [89]. Early analysis such as this is critical to allow adequate adjustment to operational procedures relevant to preparation for capping, in order to avoid major problems during remediation and possibly major capping failure.

7.6. Effluent containment and treatment

Effluents and particular acid mine drainage has received considerable attention from the base metal and precious metal mining sectors in recent years, not the least via the Canadian MEND programme. This programme did include some research directed at uranium tailings – laboratory lysimeter studies of oxidation, leaching and limestone neutralization characteristics of uranium mill tailings and waste rock [185], and investigations into the application of 'wet barriers' (i.e. lake disposal) for pyritic uranium tailings [186] [187]. A broad range of techniques for characterizing ore and waste products for their acid-producing potential, and methods of treatment to reduce acid production through selective mining and waste placement, neutralization and isolation have been developed and widely promulgated by industry and government (e.g. Environment Australia's Best Practice Environmental Management in Mining booklet series). Other material on AMD includes investigation into leaching of Ra U Pb and Th by mine acidic solution [188], the problem of arsenic mobilization at some Saskatchewan uranium mines in acidic solutions and the conclusion that dry disposal is preferable to subaqueous disposal in such circumstances [189], a proposal to use fly-ash to stabilise and solidify pyritic tailings through its neutralizing capacity and promotion of precipitation of secondary minerals [161], and a case study for acid treatment at an uranium mine in Brazil ([190], Annex I and references therein).

The application of wetlands as bioreactors and geochemical barriers is discussed, and research needs are outlined [152]. A more comprehensive discussion of their application to acid drainage treatment is provided in [106].

Surface contouring is of major importance for the overall optimization of cover systems. The final landform of a tailings impoundment will determine its run-off characteristics and erosion resistance. This has been tested extensively at various Wismut sites [191]. In this context the maxim of designing with nature, rather than against it, holds in particular. Erosion losses can be minimised by keeping potential gradients low.

7.7. Management systems

"Mining companies face the challenge of effectively and efficiently managing tailings facilities through a life cycle, from initial site selection and design, through construction and operation to eventual decommissioning and closure". [81] (see Fig. 16).

The general trend in research relevant to management systems, and post-closure management in particular, reflects the trend towards practices consistent with the principle of sustainability, and some recent presentations stress the importance of continual improvement in operational and management practices in order to achieve that principle, e.g. [192]. Significant attention is given to the advantages of utilizing passive systems in order to achieve sustainable and manageable long term outcomes in long term surveillance and maintenance, e.g. [193].

A notable observation is the evident emphasis in recent research towards developing management systems that are compatible with natural systems, and where natural processes can be used to assist in protecting the environment from the risk of detrimental impacts. Natural attenuation is identified as a 'new approach' for solving environmental problems through harnessing natural processes to improve long term environmental protection through biodegradation, stabilization, source removal etc. [194]. However, such concepts have formed a crucial element in the performance assessment for the geological disposal of radioactive wastes, where it is the main function of the host rock.

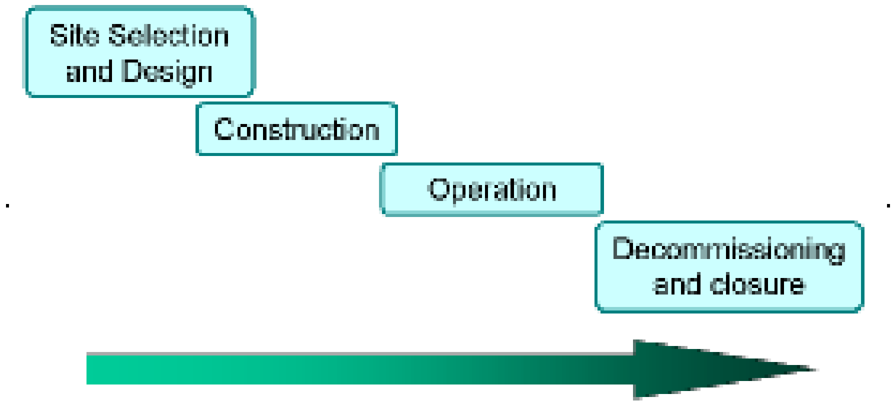

Fig. 16. The life-cycle of a mining and milling facility (courtesy H. Cotzee).

Studies into natural attenuation include:

— A systems approach to incorporating natural attenuation processes in decommissioning of Cluff Lake, northern Canada [124];
— Studies into the sorption capacity of organic lake sediments at Cluff Lake [127] and of lake-bottom algae at Rabbit Lake [128];
— Studies on different soils, peat and clays to select materials for incorporation into tailings barriers and covers (see Annex III);

- Natural attenuation processes as possible elements in flooding strategies to manage contamination from underground mines (reduction, precipitation, sorption, density stratification, zero-valent iron) [125];
- Use of wetlands and land irrigation to remove contaminants through soil, sediment and plant sorption/uptake – at Ranger mine Australia [126], Elliot Lake, Canada [195], and Saxony/Thuringia, Germany [129].
- The capacity for chemical reduction and fixation of uranium through natural diagenetic processes and enhancement of diagenesis through addition of gypsum etc., crossing into stabilization and barrier technology research (see Annexes VI and X).

However, it is difficult to assess for how long such systems will remain effective [130], and details of the processes by which contaminants are removed are not well understood. Research is under way to evaluate the mechanisms that control and limit the effectiveness of passive systems including 'bioreactors'. This includes studies into organic fixation and speciation [196], and investigations into methods to determine the feasibility and likely performance of systems based on sulfate reducing bacteria [197].

Some discussions are promoting the concept of strategic planning that incorporates a variety of passive systems in the post-closure landscape to promote assimilation of potential contaminants and develop a land-use, based for example on agro-forestry or nature conservation [198]. Such approaches lead to the conclusion that conventional approaches to acceptance criteria for close-out [199], and requirements for post-closure monitoring [200] will soon undergo revision.

7.8. Long term research priorities

Taking into account the preceding discussion there appears to be a need to focus research at

- aiming for reducing the reliance on active maintenance, and
- where systems do not perform according to design specifications, they will have to be actively maintained for the period that research is undertaken to explain the design flaw and rectify the shortcomings

8. PERFORMANCE ASSESSMENT OF IMPOUNDMENTS

8.1. Purpose of Performance Assessment

Performance assessment normally includes the verification of compliance with applicable regulations, as well as site specific performance objectives. A performance assessment also provides a tool for the optimization of remediation measures and the identification of shortcomings in design, operating procedures and management systems. Since the design and operation of many tailings facilities relies on assumptions and models, performance assessment supplies information that is useful in the verification and calibration of these assumptions and models. Future research and development priorities can be identified on the basis of deficiencies within a design or system thus assessed.

8.2. Conceptual Approaches

Two basic approaches exist, risk-based assessment and design-based assessment. A risk-based performance is based on the calculated dose to a hypothetical receptor at a given point of compliance. A design-based assessment is based on compliance with engineering design specifications. The risk-based assessment appears to be the more commonly accepted methodology, as it allows the development of optimised and cost-effective procedures. The design-based assessment is prescriptive, making it easily applied and verified. Design-based assessment was used in the UMTRA programme in the USA, while the WISMUT clean-up in Germany used a risk-based approach.

Performance parameters may be calculated using deterministic or probabilistic methods. These are complimentary methods and are defined as follows:

— The deterministic approach or 'best estimate analysis' is performed on separate scenarios and results in 'best-estimate values' for the input parameters, such as source term, recharge, tailings seepage, groundwater flow and transport, radon diffusion, and distribution coefficients. This approach is well suited for testing by parameter sensitivity analysis.
— The probabilistic approach is based on a statistical distribution of multiple parameters using a Monte Carlo-simulator to calculate an average value, such as a range of a calculated dose at a given point of conpliance.

8.3. Performance assessment approaches

The actual approach adopted for a performance assessment will depend on the conceptual starting point, site specific parameters, as well as possible requirements by the licensing authorities. The concept of performance assessment has been extensively developed in the context of radioactive waste disposal. For instance, the Sandia National Laboratory has adopted the following, six task process for the performance assessment of the Waste Isolation Pilot Plant near Carlsbad, New Mexico [201]:

(1) Disposal-system and regional characterization through collection of data on waste properties, facility design, and regional geology and hydrology.

(2) Scenario development that identifies combinations of events and processes whereby contaminants might be released outside a 'controlled' area, and subsequent determination of which scenarios to model.

(3) Probability modeling that provides estimates of the likelihood that the retained scenarios will occur.

(4) Consequence analysis, including uncertainty analysis, that predicts contaminant amounts released and associated uncertainties from the calculations.

(5) Regulatory-compliance assessment through construction of CCDFs (hypothetical CCDF illustrated below) for comparison of modeling results with long term performance criteria in [3].

(6) Sensitivity analysis that determines the parameters that most influence modeling results.

This assessment procedure was intended for use at high level waste repositories. Tailings disposal facilities present a lower level of risk, and, although similar principals may be applied, the rigor of design and level and intensity of monitoring will probably be far lower

8.4. Baseline data and regional characterization

It is necessary that the impoundment system in its regional context be understood well and relevant site investigations are needed. The data produced from these site investigations, both in a design or remediation context, will provide baseline conditions and allow the prediction of the behaviour of the facility in the short, medium and long term. Typically, these time frames would cover the operational, closure, and post-closure phases respectively.

Medium and long term predictions are based on data for the local conditions, models calibrated to local conditions, and possibly the study of engineered or natural analogues [113] [123]. An important aspect of the use of predictive modelling is a procedure to verify the predictions of the model via monitoring as part of the performance assessment procedure. Where observed conditions differ significantly from predictions, the regulatory regime should enforce corrective action if necessary.

8.5. Monitoring

Recently, the IAEA has published a comprehensive set of guidelines for the scope and performance of monitoring programmes for uranium mining and milling residues [121] [203]. The short term effectiveness of the impoundment structures is monitored during the disposal and environment surveillance stage, using the appropriate means (monitoring networks). In the longer term, effectiveness can no longer be demonstrated directly, due to the time-scale. The isolating ability of the repository, therefore, has to be forecast using models that can be updated, if required, as new data are gathered.

Regardless of the time period in question, the approach followed to demonstrate that impact is acceptable typically consists of several iterative steps:

- description and understanding of the behaviour of the various structures making up the impoundment,
- selection of realistic exposure scenarios derived from events or processes that are representative of the potential risk of degradation of the impoundment,
- analysis of the effects of these processes and events on the effectiveness and stability of the repository structures and how their environment changes, along with an assessment of the radiological consequences of the exposure scenarios selected.

The isolating ability of impoundments for uranium mill tailings depends on the effectiveness of the impoundment structures as a whole, be they man-made or natural.

The main items for assessing the behaviour of the repository are typically:

- measurements and investigations carried out on the impoundment and its environment, from the start of operation to the present time:
  - as regards transfer through the atmosphere: radon concentrations, activity of dust transported in the form of aerosols, and external exposure;
  - as regards water pathways: physical, chemical and radiological characteristics of surface and groundwater;
  - description of the steps taken or planned to restrict use in order to prevent human activities (e.g. bans on house building or digging on the impoundment site, ban on use of water from the repository site etc.);
  - means implemented to maintain the repository structures and the means used to check them and ensure their integrity;

- geotechnical stability in the short term with respect to settling, including differential settling, changes in gradients and materials properties (changes in porosity, migration of fines, changes in volume or loss of cohesion), fluctuations in piezometric level (saturation risk);
- geotechnical stability in the medium and long term;
- description of the radionuclide pathways in relation to their speciation, hydrology and the hydrogeology of the repository and its environment as well as the consequences resulting from geotechnical changes.

Since the migration of contaminants from a tailings disposal facility may occur over long periods of time and, hence, the long term performance of the facility must be assessed, it is often useful to look at analogues. These may be existing engineered structures or natural analogue sites.

8.6. Radiological impact

8.6.1. Concept of critical groups

The radiological performance of a tailings impoundment would be evaluated with respect to certain scenarios and critical groups. A realistic critical group is representative of individuals likely to receive the highest doses in the immediate environment of the tailings facility for a given scenario. Aspects to be considered in identifying a critical group include the degree of self-sufficiency, current local lifestyles and, in particular, the origin of water supply.

The assessment of the future radiological impact caused by potential releases of radioactivity to the environment typically is based on current technological levels and lifestyles. However, any extrapolations of living conditions into a distant future would be largely speculative.

8.6.2. Scenarios

8.6.2.1. Approaches for generating scenarios

Scenarios are collections of actual or predicted features and events that will lead to certain exposure patterns. The process for developing scenarios, however, typically is rather informal and relying on expert opinion. This holds in particular for 'future' scenarios. The diversity of events, and their sequences and consequences may lead to a complex system to be analysed.

In order to facilitate the development of scenarios, to formalise the reasoning, and to avoid any suspicion of 'arbitrary' selection of scenarios, a systematic approach in generating scenarios may be adopted:

1. identifying and establishing an inventory of all possible processes, their origin (natural or anthropogenic), the probability of their occurrence, and the effects that they will have on the integrity of the impoundment;
2. identifying all the different possible states of the impoundment, their combinations, and their possible changes;
3. the elimination of certain combinations of their states due to respective incompatibility or lack of relevance, considereing the events or processes which could produce them;
4. categorization of combinations of states with respect to the type of resulting exposure and critical group;
5. generation of formalised exposure scenarios that reflect these categories of combinations of states.

Such approach would be expected to result in a limited and tractable number of formalised exposure scenarios representative of different categories of events or sequences of events.

8.6.2.2. Resulting exposure scenarios

Typically two types of scenarios are developed using such approach:

- A reference scenario that corresponds to a ‘normal’ development of the impoundment, taking into account:
  - normal changes in the natural environment around the repository,
  - all natural factors that can lead, for various reasons, to gradual modifications over time of the performance, the structural integrity, and the impounded residues.
- ‘Event’ scenarios: these scenarios would reflect increased deterioration of impoundment and its environment, or human actions that would endanger the integrity of the impoundment.

Superimposed on the development of scenarios may be probability analyses, which help to further eliminate from consideration improbable scenarios.

8.6.2.3. Radiological consequences of these scenarios

The possible radiological consequences of these scenarios would be assessed using the additional individual effective dose to members of the critical group. These assessments are based, as much as possible, and in particular for evaluation of short term scenarios, on the results of measurements made at the site of the environment and its immediate environment.

Assessment of the additional effective dose includes:

- external exposure,
- internal exposure due to intake by inhalation and ingestion of these radionuclides.

These assessments typically are supported by:

- geometrical simplifications and/or simplifications of phenomena adopted, showing that they are conservative,
- sensitivity studies used to identify important phenomena and parameters and providing a justification for simplifications made,
- estimations of the uncertainty ranges.

The radiological consequences resulting from the presence of a tailings impoundment are then compared to the doses that would be received on the site without tailings. Reference would be made to baseline data, if available. Otherwise, the effective natural background dose would need to be estimated from measurements at nearby locations, but in an environment with characteristics comparable to those of the site. Variations in the natural background dose, estimated or measured, would also be taken into account.

8.6.2.4. Normal conditions- reference scenario

The radiological impact from the tailings impoundment would be assessed for reference scenarios using the additional effective dose calculated for a period at least equal to the length of time during which the disposal structures is estimated to remain effective. For this period explicit uncertainties can be taken into account. Beyond this period, the results will have an additional uncertainty attributable to the uncertainty in the impoundment development. Dose estimates, therefore, would be supplemented by qualitative assessments of the factors that cause the uncertainties.

8.6.2.5. Hypothetical conditions — 'event' scenarios

Hypothetical conditions result from certain random events, whether naturally-occurring or related to human activity, are likely to involve a risk of higher addtional effective doses.

In order to capture the random nature of such events, a probability of their occurrence would be estimated whenever possible. The acceptability of the additional effective doses would be assessed as a function of the probability of occurrence, taking into account the characteristics of the random events, the duration and types of radionuclide transfer through the biosphere, the characteristics of the exposure pathways, and the type critical group(s) considered.

8.7. Non-radiological impacts

While the above discussion focused on radiation dose as an overall measure of performance, it should be noted that there are other environmental stressors that need to be taken into account. Relevant stressors typically include *inter alia* toxic heavy metals, acid drainage, and dust.

The methods to derive scenarios and assess risk would be very similar to those discussed above. The evaluation, however, would not be carried out on the basis of a dose to a critical group, but with respect to resulting concentrations in certain environmental compartments as stipulated in relevant national legislation.

Similarly, methods exist to evaluate conventional risks from geotechnical failure [76] [80].

9. SUMMARY

9.1. Historical practices

Tailings produced from the milling of uranium ore in the 1950s and 1960s were regarded as little or no different to other non-radioactive mill tailings in terms of their potential hazard. The general poor level of knowledge as to the potential radiological effects is clearly demonstrated by use of uranium tailings as a building material in many places close to uranium mines and mills. The tailings were placed as a matter of convenience and cost in the nearest receptacle, which would commonly be a natural depression or lake if available. If no suitable natural depression was available, one would be constructed by building an embankment or low dam across a valley or across the lower slopes of gently falling ground. Where no depressions or slopes were available the tailings were sometimes placed on flat ground in dry climates, or a ring dyke would be built in wetter areas. However, there was generally little or no attempt to prevent flow of waters that had contacted the tailings into streams and rivers. Scant regard was paid to dispersal as dust, and generally no action was taken to remediate areas affected by a containment failure or to collect and return the spilled tailings back to the containment.

Containment practices began to improve in the mid-1960s, and more substantial impoundments were commonly constructed, especially at new mine and mill sites where improved tailings dam designs were incorporated into tailings management plans – such as the upstream method of construction where cycloned tailings would be used to build raises on the dam embankment. However, unsuitable practices for tailings disposal continued at some sites until the 1970s and later, especially where uranium production was subject to tight government control and secrecy.

Some of the options for placement of tailings that we would consider suitable today were used historically, but more by coincidence than design. An example is the backfilling of old open pits; however, no amendments were made to limit the potential for contamination of surface or ground waters. In general, no consideration was given to the different climatic situations in which the tailings were stored and the different levels of risk of containment failure or the consequences to human health or to the environment. This lack of concern was based on ignorance of the risks and hazards. Development of an understanding of these risks and hazards grew out of the growing body of knowledge about the effects of radiation on the human body. Legislation aimed primarily and protecting the health of workers was developed from the 1940s, but it was not until the mid 1960s that concern for impacts on the wider community and on the environment led to a major change in the way that the risks and hazards of uranium mill tailings were perceived, including the long term nature of those risks and hazards. From this time a major change took place in the approach to design of tailings containments. A new approach gradually developed that considered issues such as climate, possible agents for containment failure, long term containment, and the values and sensitivity of the surrounding environment in order to reduce the risk of containment failure and potential hazards to the environment.

9.2. Environmental impacts

Poor practices in the placement and management of mill tailings in the past have contributed significantly to the negative legacy of uranium mining. The dangers specific to uranium mill tailings relative to tailings from other types of mines were unknown and they were treated in the same manner. That manner was to dispose of the tailings cheaply and conveniently – in topographic depressions to contain tailings and liquor in wet environments (the depressions may have been valleys, lakes etc), or to pile them on flat areas in drier climates. The main vector for contamination in wetter regions is water erosion, leading to contamination of water courses and lakes, while in dry regions the main vector is dust. The contaminants comprised radioactive material as well as toxic metals and other chemical compounds, in many instances accompanied by acidity and salinity.

As was the general case in the mining industry in the first 60 years of last century, there was little or no maintenance of tailings piles; for example, if fences had been provided they were not mended or replaced if breached or stolen, and there was no rigorous restriction on access to and re-use of the tailings material.

Health and environmental impacts of a wide variety occurred, relating variously to:

— Catastrophic failure/collapse of containments, in some cases causing deaths;
— Uncontrolled public access to and inappropriate re-use, leading to increased radiation doses and cancer risks;
— Overflow and/or seepage of tailings liquor into surface water systems, causing contamination and ecosystem impacts along rivers and in lakes;
— Seepage into surface and groundwater systems, leading to contamination of water resources important for drinking, irrigation, public amenity and tourism.

The recognition of radiation exposure due to the re-use of tailings as a building material in the USA, mainly as aggregate for concrete production and for fill material, led to an understanding of the level of risk that tailings posed to human health. Subsequently, the first regulations applying specifically to uranium mill tailings were developed. An understanding of the risks posed to the environment has been slower to develop, starting from a concern that uptake and bio-accumulation in the food chain could increase the total dose to humans.

Assumptions using man as the indicator species to determine a safe radiological dose rate for all species appear to have been useful approximations upon which to base standards for protecting the natural environment from the risk of damage from ionizing radiation. However, the long term risks of chronic exposure are not understood, particularly in terms of potential long term (i.e. genetic) effects on species populations, density, ecosystem dynamics, and biodiversity. A new body of research is building in this area that focuses on risks and effects in the fabrication, power generation and disposal parts of the nuclear fuel cycle; it has yet to adequately address risks arising from lower activity levels from tailings impoundments.

Uranium mill tailings are made up of a cocktail of potentially toxic components, some of which are known to be significantly more toxic that the radioactive minerals. Studies of impacts in the past, as well as planning better tailings management in the future, needs to take into consideration the full suite of toxicants present in uranium mill tailings.

9.3. Remediation programmes

Remediation of uranium mill tailings piles has been undertaken and/or is in progress in virtually every country, where uranium has been or is being produced. Early attempts at remediation have been variable in their success. Assessments of effectiveness are few in number and generally limited in quantitative and comparable information. Even with the most careful planning, there is room for certain critical concerns to be overlooked. As knowledge on environmental sensitivity grows and regulations and standards evolve to reflect this knowledge in response to public concern, the design standards for remediation will become more demanding.

Much is being learned from the remediation programmes as they are undertaken and as monitoring data are collected after completion. A distinctive message that comes out of the work done to date is that there is no single best approach. Each site must be studied closely to determine the optimal approach. The amount and type of work, and the associated costs, will differ markedly from site to site. Short-cuts will probably lead to parts of the work having to be re-done. Methods of construction and design that are harmonious with and emulate nature are preferable to engineered designs that are built to resist, rather than to harness natural processes for long term stability.

The challenge of remediating areas of environmental impact from old mines has proven difficult on technological and economic grounds. Even the well-funded Uranium Mill Tailings Remediation Action Programme (UMTRA) in the USA took almost twenty years to undertake (Annex XIII). Remediation works and costs are most demanding when planning and incremental preparation for closure and remediation are not integral to the operational life of the mine and tailings facilities. Where uranium mining is undertaken by the private sector, regulations may be developed and imposed to ensure adequate and timely planning and funding for remediation to reduce or remove the probability of liabilities being transferred to government. Where uranium mining is undertaken by government institutions, similar arrangements may be put in place to ensure that remediation planning and implementation are identified as the responsibility of the operators. Funding normally takes place on the basis of approved programmes and is subjected to comprehensive control.

9.4. Present day practices

In general terms the technologies used in the uranium mining industry are no different from those used for other mill tailings. Most of the environmental problems experienced with uranium tailings are similar to those found at other mines and mills. In fact, a significant source of environmental hazard related to uranium mill tailings may come from the non-radioactive components of the contaminant inventory. Hence, the principles developed for other types of mining can generally be applied to uranium mill tailings. Examples of most of the different types of containment and method of their construction can be found in the uranium mining industry. For tailings facilities developed during the last 25 years, detailed information is readily available and in the public domain.

The containment of mill tailings is closely related to the site specific setting of the mine and mill. The main considerations for defining the most appropriate practice are technical, environmental, economic and societal. While engineering design provides a wide range of options for the containment of mill tailings, the nature of the site where they are located generally determines the containment method employed. What can be considered the best or the most cost-effective containment will differ from site to site, depending on climate, ore/tailings type, impounded volume, seismicity, remoteness, available transportation and logistics, and availability of natural or constructed openings. Clearly, a careful analysis needs to be undertaken for each individual site to ensure that the most appropriate option is chosen.

Tailings dam failures have occurred in the past, but their occurrence and frequency are not readily related to certain types of dam or construction methods. There is statistical evidence to support the view that most failures relate to the saturation, liquefaction and rheological characteristics of wet tailings held in dams, and that the risk is much higher during the operational phase. Clearly, water management is a critical issue for risk reduction. Dams can fail after closure, mainly as a result of earthquakes, geotechnical factors, and overland flooding. Often, poor or non-existent care and maintenance are at the root of these failures. Accordingly, close-out designs should pay particular attention to mitigating these hazards. A large body of information exists with respect to tailings dam failures outside the uranium industry that is directly relevant to uranium tailings management.

Containments other than dams that utilise natural depressions, such as lakes, or constructed voids, such as mine pits and underground openings, offer certain advantages because of their inherent physical stability after closure (Annex XII). Risk from earthquakes and overtopping is mostly removed. The main mechanism for environmental pollution from below ground containment is the contamination of groundwater.

Methods available to reduce the risk of pollution from containments involve reduction of possible contaminant source terms by tailings preparation and treatment (Annex II and VI)), isolation from the environment by covers and seals, as well as active and passive effluent treatment. In some cases relocation may be the most appropriate remedial solution, particularly, if the tailings are in the immediate proximity of populated areas.

A large body of work is available on capping design. Cover designs and construction needs to be site-specific, taking into account the local climate, availability of construction materials, nature of the tailings impoundments, regulatory constraints, as well as local community demands and acceptability. Although site specific information is needed for each site, it is probable that research work into designs is being repeated unnecessarily in several countries. It seems to be time for this large body of work to be assimilated so that it is readily available and comprehensible to all. Nevertheless, the local availability or non-availability of generally preferred construction materials, such as clay for cappings, and the limited financial resources to obtain these materials on the world market, may make it necessary to develop local designs in some Member States (see Annex III-China for instance).

Research specific to uranium mill tailings containment goes beyond research into containment of non-uranium mill tailings, because of the focus on longevity and the problems specific to radioactivity (particularly radon emanation). Specific areas of research into longevity include, climate change and variability, erosion, long term geotechnical stability, monitoring, and long term model development and calibration.

However, non-uranium tailings also pose long term hazards, particularly due to acid mine drainage generation [202]. There is potential for the benefits of research into uranium tailings management to feed into other, non-uranium tailings research areas and *vice versa*, and for co-operative research on shared issues of concern, such as the longevity of physical and chemical performance of containments.

In summary, considerable progress has been made in the safe containment of uranium tailings over the past 20–30 years.

9.5. Outstanding issues

The outstanding issues relating to stabilization and isolation of uranium mill tailings concern the confidence in long term secure containment, prevention of seepage from the containments, and chemical mobility of contaminants in the short and long term. A new concept is developing that seeks to embrace natural processes as much as possible in improving physical and chemical stability by understanding and taking advantage of natural assimilation and attenuation (ecological design). This could lead to a fundamental change in the way that impoundments are designed, away from structures engineered to keep their contents out of contact with the environment, to structures that somehow utilise natural processes to assimilate the entire structure and its contents with the surrounding environment over the long term.

9.6. New approaches

A hiatus in the level of research occurred in the 1990s following the conclusion of much of the research undertaken in north America under the US Uranium Mill Tailings Remediation Action Programme and the Canadian National Uranium Tailings Programme. The results and lessons learned from those programmes are still relevant today, and form the basis of basic research into design and planning of remedial works in other countries today. However, the results of the earlier work are not always readily accessible and a comprehensive critical evaluation is not available, so that valuable funds and research resources are not spent on un-necessary repetition and duplication of research.

The main foci of current research are methods to stabilise tailings (Annexes II,VI, IX, and X). Whilst the above discussion attempts to divide the description of this work into physical stabilization and chemical stabilization of tailings piles, and stabilization of covers, much of the work being done cuts across all three areas. This is because the majority of the work is on methods to ‘cement’ together the tailings with agents that are able to absorb or co-precipitate the contaminants (with emphasis on the metals) as a result of redox reactions and modifications to pH. Therefore both chemical and physical stability result from the one process. The agents vary from tried and tested Portland cements, to other inorganic and organic reagents, and high-tech polymers (Annex XI). Some harness and enhance natural diagenetic processes, and others take no account of them.

The result is a large body of work offering tantalizing potential opportunities for application in the isolation and stabilization if uranium mill tailings, at the placement stage, during non-operational pre-closure stages, during remediation preparation, remediation works, and *ex post facto*. However, much of the work is not sufficiently developed and demonstrated to allow the potential benefits, costs, and operational requirements to be evaluated. Also, techniques that have been developed and implemented successfully for non-uranium tailings have not been adequately tested on uranium tailings for their suitability to be fully apparent (e.g. acid drainage mitigation techniques that would also reduce uranium mobility in the case of acidic/sulfidic tailings, paste and dry cake technology for dewatering and pacing tailings).

The requirement for high security tailings isolation for periods of time in the order of thousands of years is peculiar to the uranium mining industry, even though sulfidic tailings dumps are now known to pose environmental risks for up to a thousand years. There is a growing level of interest in a new approach to designing mine closure in a way which is cognisant of the effectively infinite time frame, rather than approaches based on finite design lives and, therefore, probable future failure. Similarly, nature provides various mechanisms that assist in stabilization of contaminants, and there is now high interest and research activity into understanding these processes so that their potential can be maximised in order to develop better 'passive' systems and reduce the probability of failure inherent in interventionist systems.

A new generation of stabilization techniques based on high-tech polymers and sophisticated organic chemistry is developing (Annex IX), and promises to provide more effective and durable techniques for operational and *ex post* stabilization than those based on Portland cement paste technology. This field of research is developing rapidly. Much of the information is based on laboratory scale testing with only limited field trails, and information on its feasibility for application in the field is generally lacking.

9.7. Performance assessment

Performance assessment normally includes the verification of compliance with applicable regulations, as well as site specific performance objectives. A performance assessment also provides a tool for the optimization of remediation measures and the identification of shortcomings in design, operating procedures and management systems. These assessment cover radiological and non-radiological, as well as conventional geotechnical aspects.

As the design and operation of many tailings facilities relies on assumptions and models, performance assessment supplies information that is useful in the verification and calibration of these assumptions and models (Annexes I and V). Future research and development priorities can be identified on the basis of deficiencies within a design or system thus assessed.

Two basic approaches exist, risk-based assessment and design-based assessment. The actual approach adopted for a performance assessment will depend on the conceptual starting point, site specific parameters, as well as possible requirements by the licensing authorities.

A performance assessment would comprise a variety of elements, including the establishing of site baseline conditions, monitoring for the short term performance, and predictive modelling for the medium to long term perfomance. The radilogical evaluation of the assessments would be carried out against different exposure scenarios developed and on the basis of doses received by respective critical groups. The non-radiological evaluation is usually carried out against prescriptive target values, such as permissible contaminant concentrations in certain environmental compartments. A well-developed body of standards exists for conventional engineering hazards.

10. CONCLUSIONS

It can be observed that the technologies for the disposal of tailings from uranium milling have reached a certain maturity. There remains, however, some uncertainty over the long term performance of impoundments. Such uncertainty is not unique to tailings impoundments, but inherent in all engineered structures on the surface of the Earth. The long term performance of below ground disposal facilities appears to be more predictable.

The uncertainties stem largely from uncertainties in the design base, such as the long term development of climatological parameters, or from random events, such as human intrusion scenarios. This document discusses various strategies to counteract these uncertainties, but focuses on technical means.

Tailings themselves pose to major challenges to ensure their long term stability: controlled consolidation and minimization of the ensuent release of contaminants. Dewatering of tailings is a major technological effort, but for newly generated tailings standard technologies exist, e.g. paste technology. The situation is different for legacy sites. Consolidation and solidificaction technologies are still in the development stage and this document discusses a few examples that are promising. Such solidification technologies will also help to keep the potential source term for contaminants low, if the physical barriers are breached.

The successful stabilization of legacy sites faces a variety of transitory challenges, for instance wind or water induced erosion problems before any cover providing for long term stability is fully operational. This report describes methods to reduce the effects of erosion.

However, as technical solutions alone are likely not sufficient to ensure long term stability, some form of institutional control may be needed. This document demonstrates the usefulness of an integrated approach, involving risk assessment, technical and management measures, as well as stakeholder participation.

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GLOSSARY

AMD	Acid mine drainage
ARD	Acid rock drainage
CEC	Cation exchange capacity
Decant Solution	Solution that is removed from the surface of a waste retention system after the solids have settled out. Some solids are always removed with the decant solution.
Institutional Control	Institutional control consists of those actions, mechanisms and arrangements implemented so as to maintain control or knowledge of a waste management site after closure, as required by the regulatory body. This control may be active (for example, by means of monitoring, surveillance, remedial work, fences) or passive (for example, by means of land use control, markers, records) [122]
ISL	*in situ* leaching, technique to remove metal value from host rock by acid or base.
Natural analogue	"...an occurrence of materials or processes which resemble those expected in a proposed geological waste repository" [124].
Paste Technology	Technology to rapidly dewaters tailings to produce a pastelike material that is still pumpable. Synthetic flocculants are added to achieve rapid settling of dense fine solids aggregates, a deep bed thickener to promote self weight consolidation in the settled solids and dewatering channels to relieve the excess pore pressures.
Riprap	Loose stones produced by crushing hard rock. Used as packing, foundations, in covers etc.
Stope	Underground cavity resulting from the extraction of ore
Tailings	The remaining portion of the metal-bearing ore consisting of finely ground rock and process liquid after some or all of the metal such as uranium has been extracted [7].
Working levels	A unit of potential alpha energy concentration (i.e. the potential alpha energy per unit volume of air) resulting from the presence of radon progeny or thoron progeny, equal to 1.3×10^5 MeV per litre. In SI units, a working level is 2.1×10^{-5} J/m^3.

ANNEX I. BRAZIL

A CASE STUDY ON THE URANIUM TAILINGS DAM OF POÇOS DE CALDAS URANIUM MINING AND MILLING SITE

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I-1. ABSTRACT

This Annex describes the geochemical processes controlling the mobilisation of heavy metals and radionuclides in the tailings dam of the Poços de Caldas Uranium Mining and Milling Facility. It was shown that residual pyrite oxidation causes the production of acid drainage that leaches metals and radionuclides from the solid phase. The remediation scheme, application of dry cover on the tailings, was focused on the reduction of oxygen diffusion into the tailings and radon exhalation from the material Three principal final covers designs were taken from the scientific literature aiming to obtain the most adequate performance for the studied situation The designs studied included: (i) a compacted clay liner (CCL); (ii) a composite liner (CL); and (iii) a capillary barrier (CB). Relevant processes investigated were: (i) saturated hydraulic flow; (ii) unsaturated hydraulic flow (only for the capillary barrier); and (iii) radon exhalation to atmosphere. The computer models utilized for the analyses were: (i) the program Hydrologic Evaluation of Landfill Performance (HELP); (ii) the program SEEP/W; and (iii) the program RADON.

I-2. Introduction

In planning the closeout of a tailing impoundment or pile, the planners must have a good and comprehensive understanding of the hazards involved and the means of controlling such hazards.

The specific activity of such materials is typically low in comparison to that of some wastes from other parts of the nuclear fuel cycle, although tailings that result from the milling of high grade ores can represent a radiological hazard comparable to that of low level radioactive waste. While tailings and residues from the mining and milling of radioactive ores can generally be categorized as very low level waste, the quantity of material requiring disposal is much larger than the quantity of waste from other parts of the nuclear fuel cycle.

As part of the planning for the decommissioning/closeout of mill tailings impoundments, an assessment will have to be made to determine, if the present impoundment and site will safely retain the tailings and the contained radioactive and chemically toxic pollutants for the length of time required by regulatory criteria. If the safety assessment shows that the present impoundment would not be safe for the required length of time, remedial actions would have to be undertaken to bring it up to standard.

If the site is unsatisfactory and the impoundment cannot be improved to compensate for site inadequacies, it may be necessary to relocate the tailings on-site or to a new site.

In planning for the closeout for the closeout of tailings impoundments, the following guidelines should be considered [I-1].

(1) Radioactive and non-radioactive contaminants released to the environment during and after closeout should not exceed authorized limits and should be as low as reasonably achievable, taking into account economic and social factors;
(2) Reliance on long-term active institutional controls as a means of adhering to regulatory criteria after closeout is complete should be minimized;
(3) Passive barriers, either natural or engineered, should be favoured
(4) Containment systems should be designed so that degradation leads to gradual rather than sudden release of contaminants
(5) Requirements for the maintenance of containment systems should be minimized and options requiring frequent maintenance should not be used.

In addition to the above items, planning for the closeout of tailings impoundments may include, as appropriate, the following information:

(1) Plans for the stabilization of tailings impoundments in situ or relocation to another site;
(2) Plans for the control of spillage and dust;
(3) Methods for the long term control of contaminant releases;
(4) Methods for the control of intrusion and unauthorized removal of tailings;
(5) Methods for the control of groundwater contamination;
(6) Plans for the cleanup and release of contaminated areas.

The major technical features associated with tailings impoundments include covers, vegetation, perimeter dams, dykes or embankments, basal features, water diversion and control features and foundations. For the closeout of an impoundment, these features must meet regulatory standards, or be brought up to these standards by remedial work.

In most cases, tailings in an impoundment may have to be covered to limit radon exhalation, moisture infiltration, gamma radiation, oxidation, intrusion by human, plants and animals and erosion.

Cover material can be classified broadly as natural (soil, rock, clay, vegetation, etc.), artificial (plastics, asphalt, soil/concrete mixtures, etc.) and water. The materials may be used in different combinations, configurations and thickness. A combination of two or more materials may improve cover performance and provide added resistance against detrimental process such as erosion and biointrusion. However, in some cases, for example, where long term settlements are to be expected, a complex cover design may have disadvantages because of its vulnerability.

The long term performance of synthetic material is questionable, since they may degrade and fail to function as required. In addition, they may be too expensive. A water cover will prevent radon exhalation, erosion and intrusion. Where tailings are susceptible to acid production, water may be a desirable approach for excluding oxygen. Issues such as groundwater contamination and retaining dam stability should be included in the assessment of water covers. If the water level is controlled by an artificial dam, long term institutional control may be required to ensure that the integrity of this structure is maintained. Should the dam fail, the water cover, and possibly a portion of the tailings, would be lost. In addition to the significant hazards associated with the dam failure itself, problems could arise with radon exhalation, dust, erosion and migration of tailings.

One disadvantage of a water cover is that it creates a hydrostatic head on the porewater system, possibly causing contaminants to move into groundwater or surface water systems. An alternative is to place the tailings into a natural water body, both during production or as a relocation option, if required. Although this is a promising technology, there may be considerable social pressures against sacrificing a lake for such waste disposal. Table I-1 shows a checklist for cover component functions.

Table I-1. Checklist for cover component functions [I-1].

Cover Component	**Purpose and function**
Erosion barrier vegetation *(top slopes only)*	Evapotranspiration of moisture that enters the soil Reduce infiltration Stabilize soil and reduce erosion Minimize impact of rain splash
Erosion barrier small diameter rock layer above topsoil on pea-grave/soil mulch *(top slopes only)*	Provide additional protection against soil erosion, used in conjunction with vegetation Reduce evaporation rates within the underlying soil layer in drier environments – prevent drying of the radon barrier.
Rooting medium *(top slopes only)*	Provide rooting medium for vegetation Store water for plant growth Protect the underlying biointrusion layer from surface exposure Provide frost protection
Frost protection (random fill) *(top and side slopes)*	Protect the underlying layers from the effects frost heave and frost penetration Preserve the physical properties of the underlying layers
Choked rock filter (layer of pea-gravel overlying a layer of coarse aggregate) *(top and side slopes)*	Prevent piping of soil into erosion/biointrusion barrier Drain infiltration as rapidly as possible to retard root growth
Erosion/biointrusion layer (50-100 cm of cobbles with a low coefficient of uniformity to prevent biointrusion) *(top and side slopes)*	Drain infiltration as rapidly as possible to retard root growth Impede burrowing animals Act as a capillary break at the bottom of the layer to prevent upward movement of water and downward unsaturated flow (enhances the moisture storage capacity) Control top slope erosion if vegetation and top-soil are eroded away
High permeability drain (15 – 30 cm layer of pea gravel overlying clean sand)	Drain water laterally off the pile to limit infiltration Protect the underlying liner system from displacement and rock penetration.
Infiltration barrier liner system (top slopes only) or high percentage bentonite mix (with silt or sand)	Intercept moisture Control infiltration Inhibit infiltration while mature vegetation community is establishing or after severe disturbance of the vegetation
Radon barrier (clay/silt) (*top and side slopes)*	Inhibit radon emanation Limit infiltration

The objectives of the present work were:

(1) to assess the mechanisms that determine the mobilization of radionuclides from the tailings pond of Poços de Caldas Uranium Mining and Milling Facility into the environment;
(2) to propose remedial actions for the tailings dam.

I-3. Description of the tailing pond

The Poços de Caldas tailings pond was designed to receive the solid wastes from the milling of uranium ore. In addition to this material, the products resulting from the acid mine/rock drainage neutralization processes, generated in the open pit mine and waste rock dumps, were also deposited in the tailings pond.

The uranium ore was formerly mined in an open cast mine. It has been estimated that $94.5 \cdot 10^6$ tons of rock have been removed during mining operations. Only 2% of this amount was subjected to physical and chemical processing. The milling process applied to the uranium ore after the physical processing stage consisted of the addition of an oxidant (pyrolusite) to the ore, and subsequent sulphuric acid leaching. The uranium was then extracted from the solution using an organic solvent, and precipitated with NH_4OH. The chemical processing stage produced large quantities of liquid and solid wastes that were discharged into the tailings pond for the deposition of solids.

The liquid effluent from the tailings pond is treated with $BaCl_2$ for the purpose of Ra precipitation, and the precipitate is stored in two settling ponds. The remaining liquid effluent is then released in to a creek. The amount of materials deposited in the tailings dam is shown in Table I-2.

Table I-2. Amount of materials deposited in the tailings dam [I-2].

Type of Material	**Amount [tons]**
Milled Ore	1,764,976
Sulfate	135,168
Pyrolusite	35,049
Phosphatic rock	6,770
Calcareous rock	74,150
Lime	35,800
Total	**2,052,913**

Table I-3. Average composition of the wastes in the tailings dam [I-2].

Element	**Concentration [%]**	**Element**	**Concentration [%]**	**Element**	**Concentration [Bq/g]**
ZrO_2	0.15	MoO_3	0.02	^{226}Ra	2.5
Al_2O_3	23.4	K	11.2	^{210}Pb	3.4
SiO_2	54.0	CaO	0.25	^{228}Ra	1.4
SO_4^{2-}	2.3	S^{2-}	0.5		
Fe_2O_3	3.9	Mn	0.02		
U	0.018	P_2O_5	0.09		
Th	0.004				

The initial volume of the tailings dam was planned to be 2,170,000 m^3. Due to the fact that the slurry resulting from the neutralization of the acid rock drainage began to be deposited at the tailings dam, an increase of 220,000 m^3 in the total volume of the system had to be achieved. The deposited slurry amounts to 66,000 tons, corresponding to the treatment of $1.34 \cdot 10^4$ m^3 of acid water. The total flooded area of the dam corresponds to 0.23 km^2, i.e., 27% of the entire system (0.86 km^2). The average composition of the solids deposited in the tailings dam is reported at Table I-3.

I-4. Geochemical Processes

The main factors affecting the extent and kinetics of metal release from tailings include the nature, composition, pretreatment, and dispersion of solids, and the composition, volume and flow rate of the leaching solution [I-3].

However, metals and radionuclides are not uniformly distributed in the solid material from the tailings. It has been suggested [I-4] that the occurrence of various mechanisms that may contribute to radionuclide and metal stratification in tailings could include:

(1) the accumulation of fines, including precipitates of gypsum and metal hydroxides, in the lower zone because of settling processes during initial deposition of the tailings;
(2) the precipitation, co-precipitation and adsorption of major metals and trace radionuclides from neutralized process water; and
(3) pyrite oxidation, leaching and desorption of metals and radionuclides in the top upper zone, producing highly acidic, high total dissolved solids (TDS) pore water which predominantly migrates downwards (solute transport) with precipitation, co-precipitation, and absorption, occurring in the neutralized buffer zone (redeposition).

The detailed discussion of the geochemical behavior of heavy metals and radionuclides associated with the Pocos de Caldas tailings can be found elsewhere [I-2]. A concentration gradient can be observed downward from the top surface of the tailings (Fig. I-1).

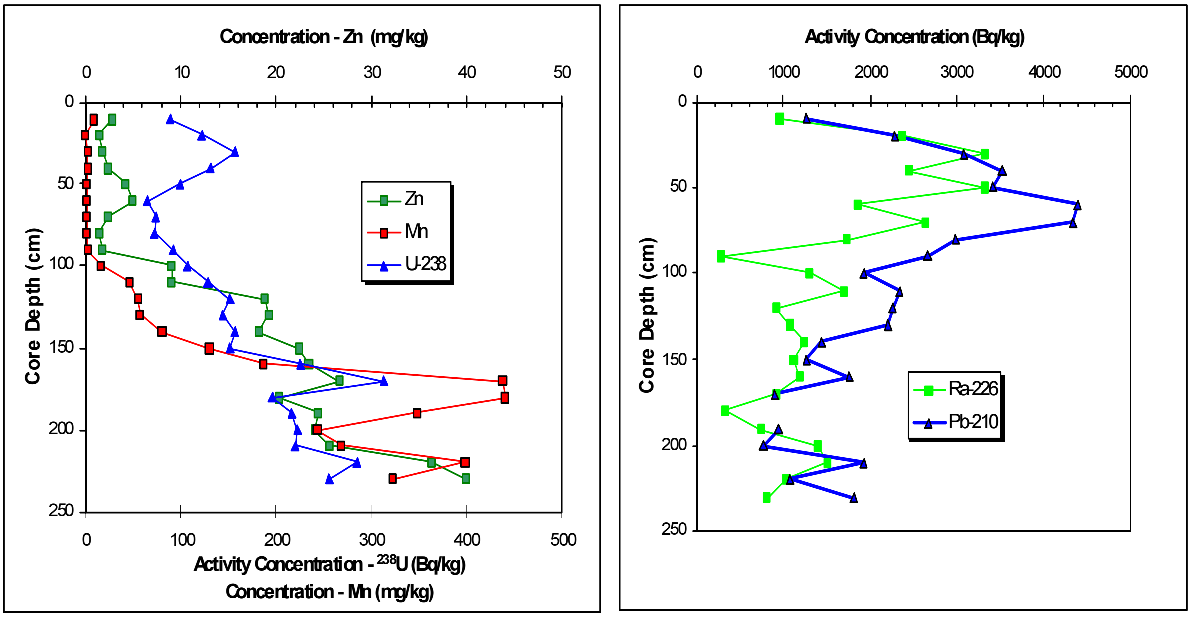

Fig. I-1. Metals and radionuclides distribution in the Pocos de Caldas tailings [I-2].

It can be supposed that the mechanisms accounting for the stratification of metal and radionuclides in the tailings involve the oxidation of residual pyrite (0,2%), caused by means of oxygen diffusion into the tailings. Bacterial activity may play a relevant role here. The acid produced in this zone is able to remove significant amounts of Zn, Mn and ^{238}U from the tailings. These elements are then transported downward with the flux of seepage water. As the acid water reaches a saturated zone with high pH values, the precipitation of these elements occur. The elevated pH are caused by the fact that the wastes were discharged in the tailings dam as a high pH solution. Part of it reacted with the acid produced in the upper zones and the rest migrated to the deeper zones. Since effluents are no longer discharged to the tailings dam, rain water is the only water supply to it. As a result, elements from the superficial layers are washed out and transports them to the bottom layers. The elements in solution are then precipitated in the deeper regions of the tailings dam. This scheme, however, does not hold for the Ra and Pb isotopes. These radionuclides are co-precipitated with gypsum and thus immobilized within the tailings. They might be eventually migrate as gypsum dissolves, this reaction being kinetically controlled. Table I-4 shows the concentrations of radionuclides and pH values of the seepage water in two different depths of the tailings.

Table I-4. Physical-chemical characteristics of pore water in the tailings dam, values in brackets indicate ranges [I-2].

Chemical Species	0.50cm	3.0 m	Unit
^{238}U	2.89 (0,15-11)	0.53 (0,17 – 0,90)	Bq/L
^{226}Ra	1.27 (0,33 – 5,0)	0.36 (0,23 – 0,45)	Bq/L
^{210}Pb	1.56 (0,07 – 4,27)	0.11 (0,09 – 0,15)	Bq/L
^{232}Th	0.10 (0,01-0,7)	< 0.010	Bq/L
^{228}Ra	< 1.30	< 0.23	Bq/L
Mn	6.0 (0,34-23)	449 (222 – 880)	mg/L
Fe-tot	63 (7,0 – 340)	162	mg/L
Zn	0.36 (0,10 – 0,80)		mg/L
Ca	236 (2,60 – 410)		mg/L
Dissolved oxygen		<0.008	mg/L
S^{2-}		< 0.02	mg/L
SO_4^{2-}		4.35 (3,0 – 6,65)	mg/L
pH		5.8	

The results closely agree with those reported by Davé et al. (1982) [4]. The oxidation zone would correspond to the zone of metal and radionuclide transport (except for Ca, ^{226}Ra, and ^{210}Pb) and would be about 1 m downward from the tailings surface. In the saturated zone (in which a pH around 6.0 prevails) metals would be precipitated. This region would correspond to an accumulation zone. Figure I-2 shows these processes in a schematic way.

These findings have an important bearing on the long term impacts of the tailings dam in respect to underground waters contamination. The accumulation zone acts as a barrier for metal and radionuclides migration. The acidification of these waters will be a slow process and very much dependent on: i) the kinetics of the residual pyrite oxidation taking place in the upper layers of the tailings, and ii) the amount of acidic waters that may be generated by this process. Some other issues may be considered, for instance the residence time of waters in the tailings dam and the movement of the oxidation front (which will be related to the consumption of the oxidizing material).

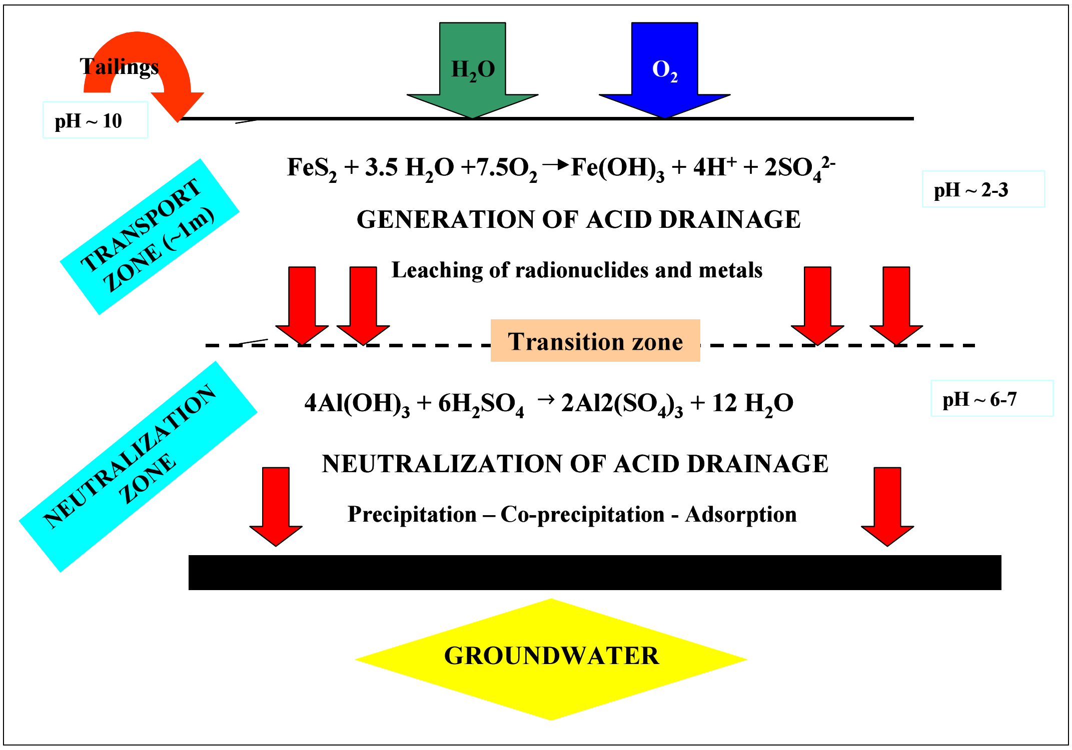

Fig. I-2. Conceptual model of the geochemical processes taking place at the tailings dam.

I-5. Remediation Strategy

Notwithstanding the above considerations a pragmatic approach to the problem can be proposed. If it is accepted that metal and radionuclide mobility will be dependent on pyrite oxidation, controlling this process would be the key issue in the remediation strategy to be proposed.

This can be achieved by means of preventing oxygen diffusion into the tailings. The most practical way to do so is by covering the tailings with a material that shows a low oxygen diffusion coefficient. In this way, pyrite oxidation would be prevented and the seepage water would remainat a pH that would favour the precipitation of the contaminants. The cover would also have an essential role in reducing water infiltration, although this would no be the major objective.

Covering would also be effective in reducing radon exhalation from the tailings and shield gamma rays. If radon exhalation would not be a major issue in terms of environmental concentration increases of the gas, it may be of concern in case of intrusion and building developments on the tailings surface. A most probable scenario would include house construction by poor people that may find the area an attractive place for housing.

The long-term efficiency of final covers is of major concern when designing final covers for uranium mill tailings impoundments.

In the establishment of a cover system for the uranium mill tailings for the Poços de Caldas facility, the approach used in the present project consisted of the assessment of the efficiency of different final covers systems by means of various computer models used in an integrated manner. Therefore, output data calculated from certain models are used as input data for other programs.

In general terms, a complete evaluation of final covers for the closure of uranium mining and milling tailings impoundments should take into account both geotechnical and geochemical aspects (which have been described above).

In turn, geotechnical investigations are concerned with the capacity of the systems to minimize water and oxygen percolation into the bulk of tailings. In addition, the effectiveness of the gas barrier layer of the covers to reduce radon exhalation towards the atmosphere has to be considered. In this regard, the geotechnical investigations comprise (i) water balance analyses; (ii) assessment of the saturated and unsaturated flux of water through final covers; and (iii) evaluation of radon flux attenuation across the gas barrier layer.

I-6. Methodology adopted for the modeling

The particular aspects involved in the closure of uranium mill tailings impoundments were investigated through computer modeling. Specific computer models were applied.

The HELP (Hydrologic Evaluation of Landfill Performance) computer model, version 3.07 [I-5] was employed for this simulation. The radon attenuation capacity of the gas barrier of the covers was investigated by utilizing RADON program, version 1.0 [I-6]. The limit value of exhalation ratio was adopted in terms of flux and followed the default value established by USEPA for uranium mill tailings impoundments closure, equal to 20 $pCi/m^2{\cdot}s$ (= 0.74 $Bq/m^2{\cdot}s$) [I-7].

The HELP model has a simplistic approach for estimating unsaturated flow. In particular, HELP cannot reasonably model the behavior of capillary barriers [I-5]. In this regard, the model herein employed to analyze unsaturated flow was the SEEP/W program, version 4.20 [I-8].

The sequential modelling approach adopted in this work consists in utilizing the infiltration results predicted in the near-surface HELP modeling as the top boundary condition for the unsaturated flow modeling.

Computational modeling ends when saturated and unsaturated fluxes as well as radon flux are lower than a defined threshold and when water infiltration is adequate to help preserving the initial saturation degree of the compacted barrier layer. Otherwise, new thickness of the layer for the cover systems should be selected, or alternative cover designs should be modelled in order that hydraulic and gas fluxes as well as cover properties are properly controlled.

I-7. Hypothetical final covers studied

Figure I-3 shows a schematic representation of the different hypothetical final covers simulated in this work.

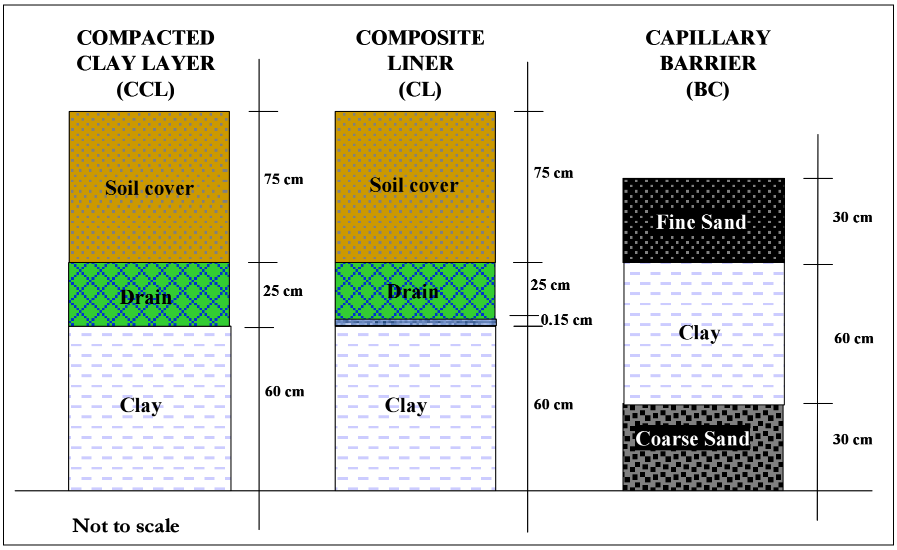

Fig. I-3. Hypothetical final covers evaluated

Table I-5. Input data used in the HELP model.

Parameter	**Soil Cover**	**Drainage Layer**	**Geomembrane**	**Compacted Clay Layer**	**Coarse Sand**
Layer type	Vertical percolation	Lateral drainage	Geomembrane liner	Barrier liner	Vertical percolation
Layer thickness [cm]	CCL & CL = 75	CCL & CL = 25 CB = 30	1.5 mm	60	CB = 30
Porosity	0.417	0.33	0	0.445	0.39
Field capacity	0.045	0.08	0	0.43	0.05
Wilting point	0.018	0.03	0	0.35	0.025
Initial water content	0.045	0.08	0	0.445	0.05
Saturated hydraulic conductivity [cm/s]	1.0×10^{-2}	2.6×10^{-3}	2.0×10^{-13}	1.0×10^{-7}	8.4×10^{-3}
Slope [%]	0				
Maximum length of drainage [m]	10				
Cover area [m^2]	400				
SCS runoff curve number	CCL / CL = 75.8 CB = 81.0				
Depth of the evaporative zone [cm]	30				
Maximum leaf area index	0 (bare ground)				
Initial snow water content [mm]	0				
Growing season	whole year				
Input data for the geomembrane of the CL final cover design	Pinhole density in geomembrane liner = 1 per hectare Geomembrane liner installation defects = 6 per hectare Geomembrane liner placement quality = good				

The Compacted Clay Layer (CCL) and Composite Liner (CL) cover designs were taken from [I-9]. The compacted clay and lateral drainage layer material properties of these covers were taken from [I-10]. The soil cover layer material properties and the characteristics of the 1.5 mm thick HDPE geomembrane were taken from the HELP default database. The Capillary Barrier (CB cover) design as well as its materials characteristics were taken from [I-10].

As the soil parameters for the three proposed cover designs were assumed to be same for the same type of layers, the behavior of the layers comprised of identical materials can be compared for the different cover designs. Each proposed cover was separately analyzed and comparisons between the different covers were made. Table I-5 lists parameters adopted.

I-8. HELP Modeling – Saturated Flow

I-8.1. Input Data

The data used for the HELP modeling are presented in Table I-5 These same values will be also used as input data for the different models when required.

Geotechnical parameters of the soils for the suggested CB (i.e., lateral drainage, compacted clay liner and coarse sand layers) were taken from [I-10].

Both soil cover and geomembrane parameters were taken from the HELP default soil properties database. Precipitation and average temperature values used in the modeling were user-input for a 20 years period and were measured in the INB meteorological station located within the Poços de Caldas UMMF.

I-8.2. Results and discussion

The summary of the annual average results of the hydrologic evaluation with HELP model is shown on Table I-6. Figure I-4 presents the graphic results obtained with the HELP model for the three suggested designs.

Table I-6. Average annual results obtained from HELP modeling

Parameter	**CCL**		**CL**		**CB**	
	%	**mm/y**	**%**	**mm/y**	**%**	**mm/y**
Evaporation	37.2	593	37.2	593	38.6	615
Runoff	8.34	133	8.36	133	30	479
Lateral Drainage	51.8	825	54.2	864	29	463
Percolation	2.45	39	0.005	0.08	2.22	35

Approximately 2.45% of the total annual average precipitation at the site percolates through the compacted clay layer of the CCL cover. It corresponds to about 39 mm of percolated water per year. The same analyses were carried out to the CL and CB final cover designs.

The average annual percolation value obtained for the CL clearly shows the importance of the geomembrane to avoid water seepage through the final cover. Only 0.005% of the total annual precipitation percolates the compacted clay layer annually, which represents 0.08 mm of the total average annual precipitation of the site.

For the CB design, the estimated average annual amount of water that percolates the cover was of 2.22%, equivalent to 35 mm/y. In this situation a minimal quantity of approximately 3.5 mm/y of accumulated water can be expected to overlay the compacted clay layer.

Analyses were carried out taking into account the average monthly results of the HELP modeling due to the dry period of the site. The dry months occur from April to September and during these months the water percolation through the covers will tend to be lower.

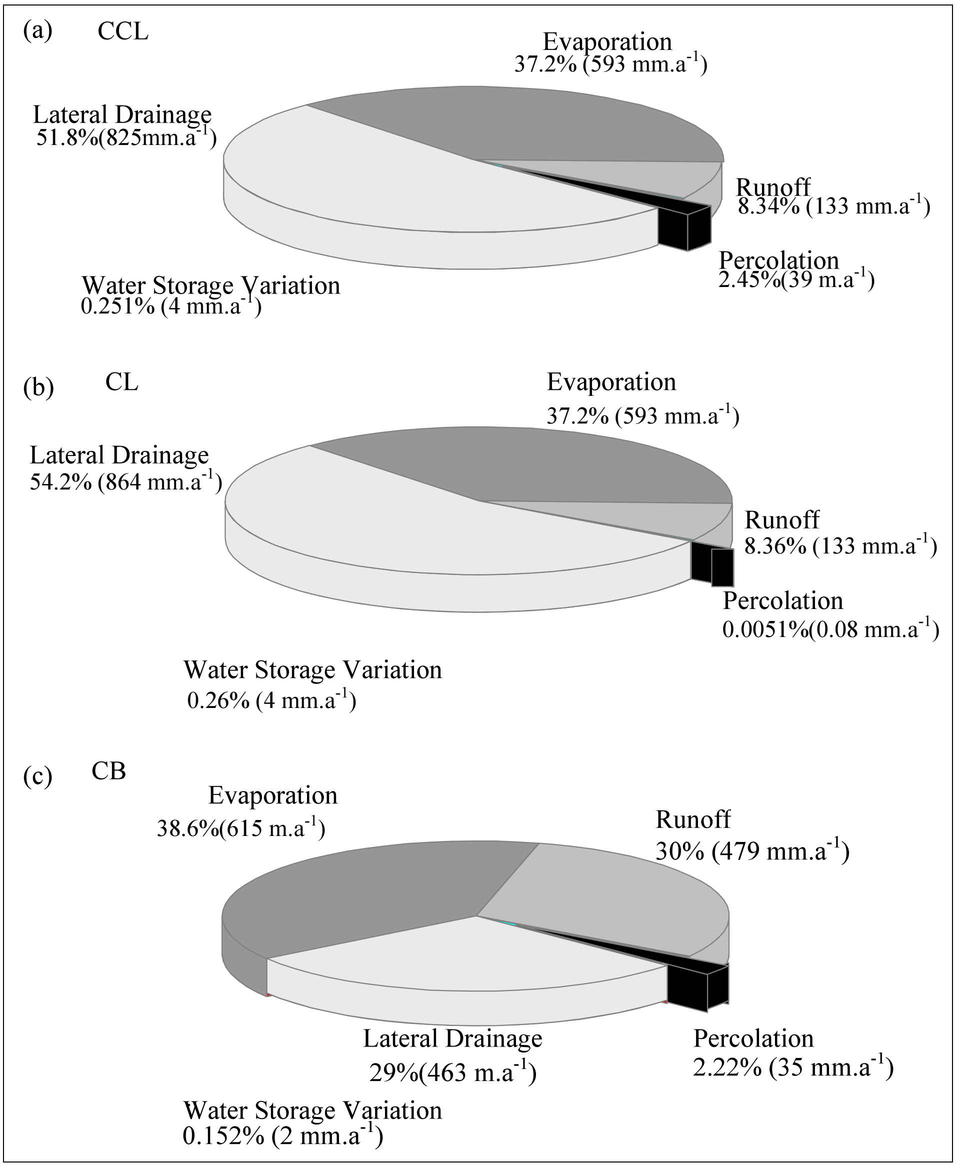

Fig. I-4. Annual average results of the hydraulic balance obtained from HELP modelling for (a) compacted clay liner (CCL); (b) composite liner (CL); and (c). capillary barrier (CB).

The results of the CCL analyses (Figure I-5a) predicted a range of water percolation values across the compacted clay layer from 2.8 to 3.7 mm per month. This analysis demonstrated that even during the dry months the flow of water migrating into the clay layer, equivalent to 2.6 mm per month, is lower than expected from the hydraulic conductivity of the material which is equivalent to $1 \cdot 10^{-7}$ cm/s.

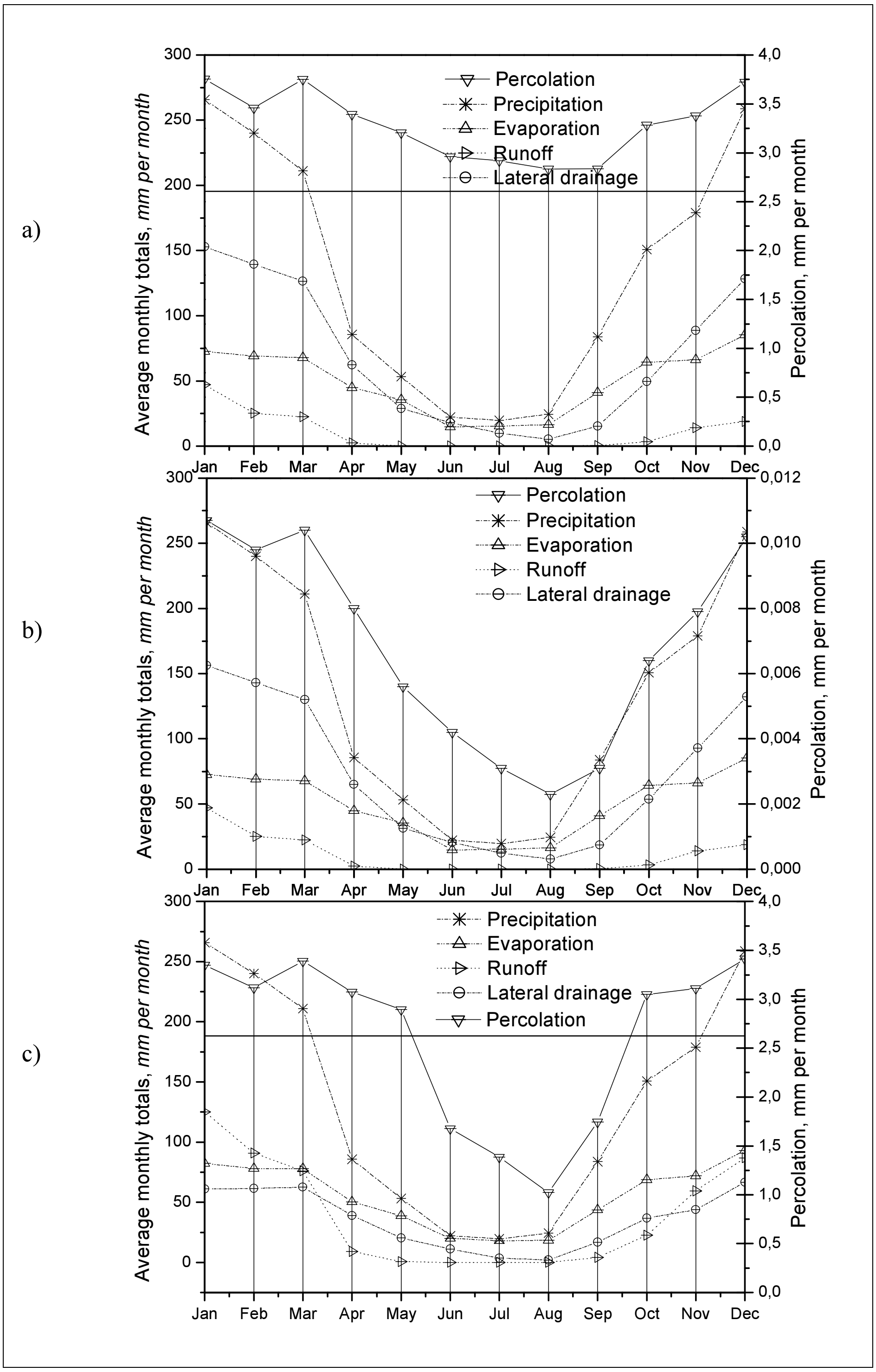

Fig. I-5. Monthly average results of the hydraulic balance obtained from HELP modelling for (a) compacted clay liner (CCL); (b) composite liner (CL); and (c) capillary barrier (CB).

Therefore, the average monthly results show the availability of accumulated water at the surface of the clay occurring for all months, including the months of the dry period. These conclusions, therefore, corroborate the conclusions obtained from the average annual results.

Figure I-5b presents the average monthly results obtained from the HELP model of the hypothetical CL cover design. The percolation values ranged from 0.002 mm to 0.01 mm per month with an average percolation value of 0.007 mm per month. The lower rates occur during the dry period.

Monthly results (Fig. I-5c) show that water percolation varied from 2.1 mm to 3.4 mm per month, and from June to September this parameter value was lower than 2.6 mm per month. This implies that for these months the predicted quantity of water migrating into the compacted clay layer is lower than possible according to the saturated hydraulic conductivity of the material. Consequently, it clearly shows the potential to shrinkage and desiccation of the compacted clay layer in the dry months.

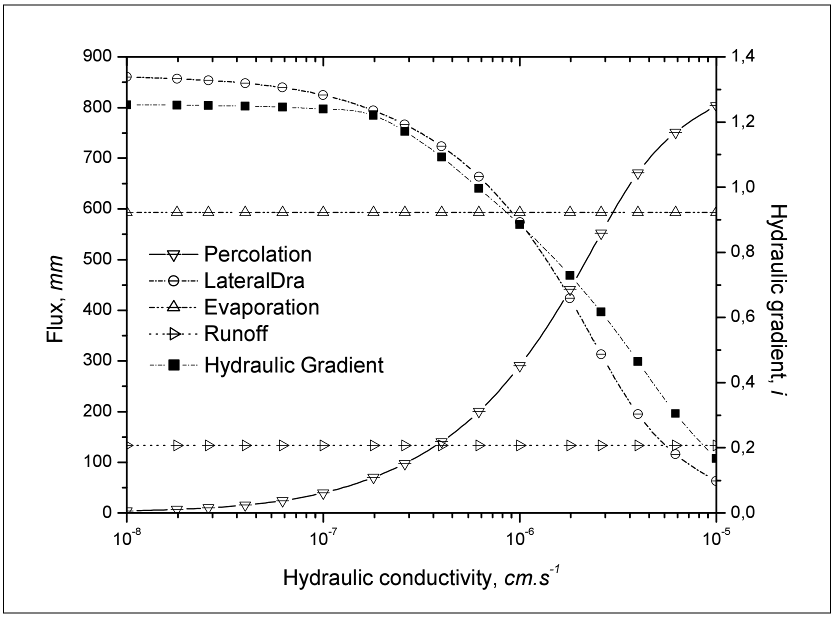

Fig. I-6. Sensitivity analysis results obtained from the variation of the hydraulic conductivity of the compacted clay layer of the CCL.

Sensitivity analyses of the compacted clay layers were performed considering that mainly the saturated hydraulic conductivity controls the percolation. The analyses were carried out by varying the hydraulic conductivity of the clay layers. The hydraulic conductivity values for the three final covers and their corresponding hydrologic parameters were taken from the annual average results obtained from the HELP modelling. The hydraulic gradient *i* was calculated from the relationship between hydraulic flow through the compacted clay layer and its corresponding hydraulic conductivity values, k_S.

The results obtained for the sensitivity analysis of the CCL clay layer are shown in Figure I-6. The percolation varies slightly until the hydraulic conductivity of the layer is equal to $1 \cdot 10^{-7}$ cm/s. From this point on, percolation starts to increase at higher rates. Since runoff and evaporation values remain unaltered as percolation increases, the lateral drainage decreases at inversely proportional rates. The hydraulic gradient is higher or equal to 1.0 until a hydraulic conductivity of approximately $7 \cdot 10^{-7}$ cm/s is reached. In other words, hydraulic conductivities equal or lower than $7 \cdot 10^{-7}$ cm/s indicate the presence of accumulated water within the layer right above the compacted clay. In a simplistic way this means that the accumulated water maintains the saturation of the compacted clay. On the other hand, hydraulic conductivity values higher than $7 \cdot 10^{-7}$ cm/s (hydraulic gradients lower than 1.0) imply a deficit of water flow throughout the compacted clay layer that suggests the increase of the potential for desiccation and, consequently, the reduction of service life of the cover.

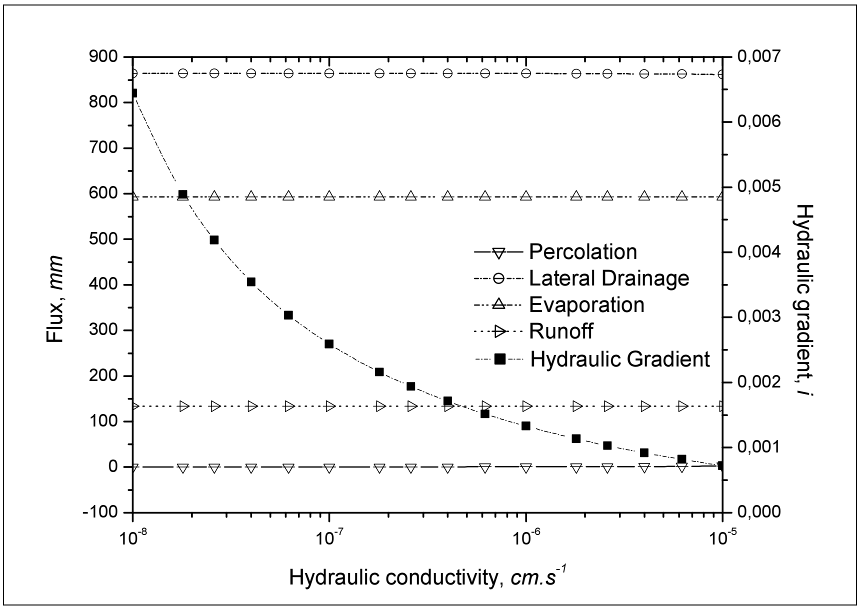

Fig. I-7. Sensitivity analysis results obtained from the variation of the hydraulic conductivity of the compacted clay layer of the CL.

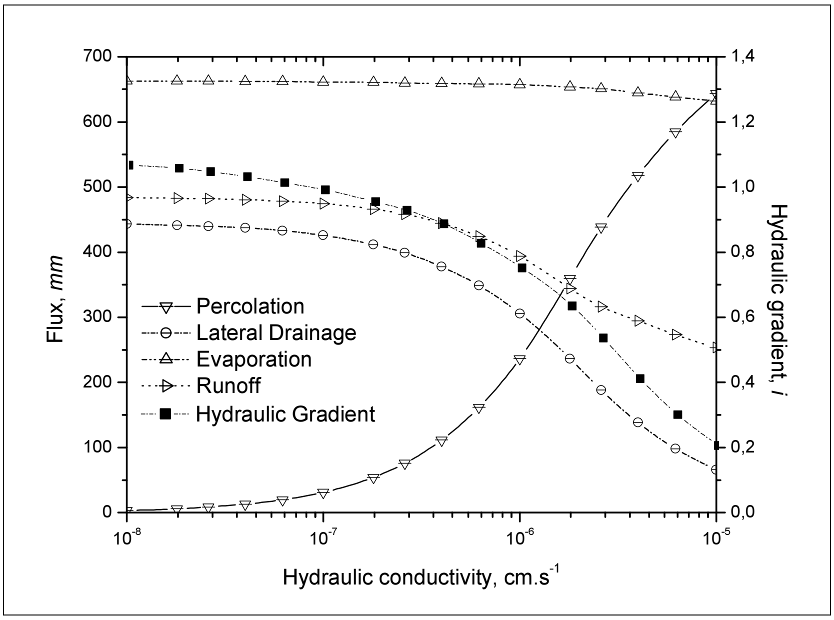

Fig. I-8. Sensitivity analysis results obtained from the variation of the hydraulic conductivity of the compacted clay layer of the CB.

Figure I-7 shows the results of the sensitivity analysis carried out by variying the hydraulic conductivity of the compacted clay layer of the CL cover. The results clearly indicate that the hydraulic flow throughout the cover is governed by the very low values of hydraulic conductivity of the geomembrane. Runoff and evaporation are kept constant, whereas lateral drainage slightly decreases as soon as the hydraulic conductivity increases. The hydraulic gradient is constantly lower than 1.0 and the percolation values are diminishing. This indicates that there is no water flow coming from the top layer migrating downwards and the total volume of inflow water migrates by lateral drainage of the upper layer.

The results of the sensitivity analysis carried by varying the hydraulic conductivity of the compacted clay layer of the CB cover are shown on Figure I-8. Hydraulic gradients lower than 1.0 result when hydraulic conductivity values are higher than $1 \cdot 10^{-7}$ cm/s. The risk of desiccation of the compacted clay layer therefore increases as the hydraulic conductivity becomes higher than this value.

I-9. SEEP/W modeling – unsaturated flow

I-9.1. Input data

The geotechnical parameters of the soil materials that compose the CB final cover, including the soil-water characteristic curves (SWCC) were taken from [I-10]. For the uranium mill tailings, the SWCC was taken from the default material properties database of the SEEP/W program [I-8] and typically represents this material.

Figure I-9 presents the hydraulic conductivity functions used in the unsaturated flow modeling of the CB that were generated by a sub-routine of the SEEP program. The hypothetical CB final cover was modelled as a two-dimensional system and under steady-state conditions. The top boundary condition was defined as the amount of water infiltration obtained from the annual average results from the HELP modeling and it was set equal to 502.4 mm/y. The sides of the finite-elements mesh were considered as a constant head boundary. The bottom of the water flux domain was supposed impermeable.

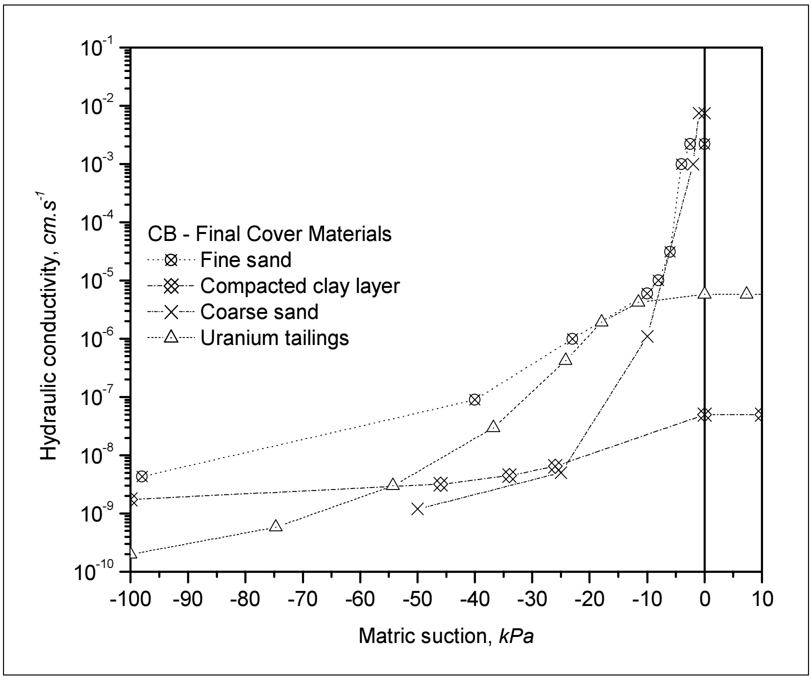

Fig. I-9. Hydraulic conductivity functions utilised in the SEEP program for modelling the unsaturated flow through the CB.

I-9.2. Results and Discussion

The SEEP/W modeling results for the proposed CB design are graphically presented in terms of hydraulic head in Figure I-10.

The head contours indicated within the clay layer suggest a low vertical saturated hydraulic conductivity. However, Figure I-10 shows that this condition is not observed in the sand base. This suggests that the sand layer shows no contribution for dissipating hydraulic head. The results from SEEP/W modeling indicated that no capillary barrier is formed under the climatic conditions found at the site considering the proposed design.

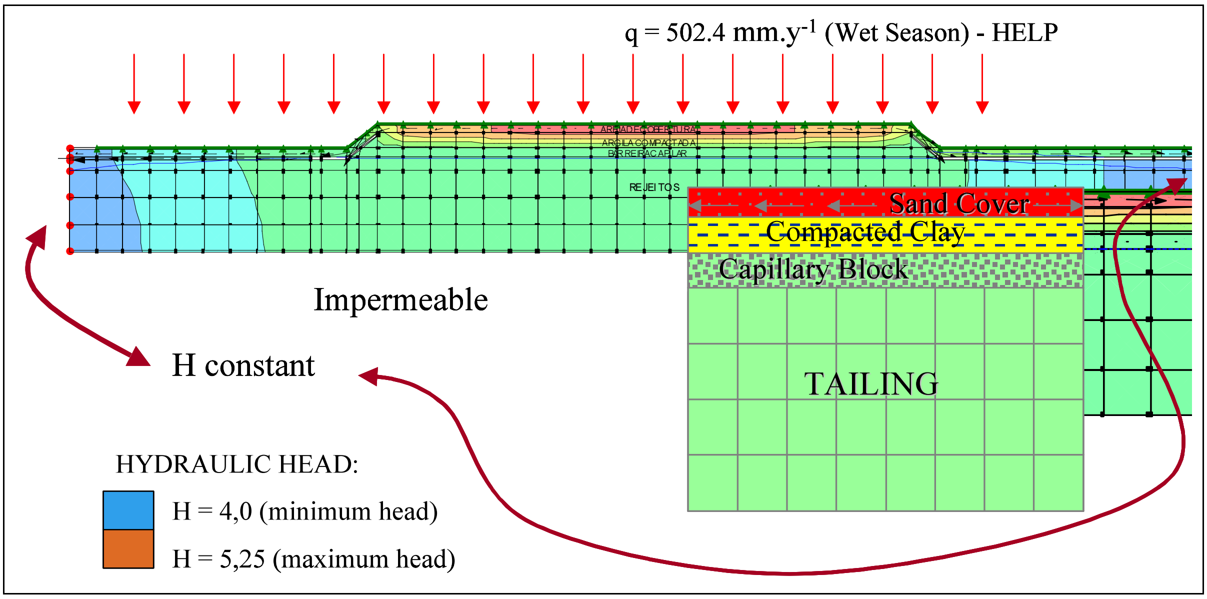

Fig. I-10. Graphical representation of the hydraulic head contours within the compacted clay layer of the CB.

The adopted hypothetical CB final cover design, therefore, acts as a CCL cover instead of a proper capillary barrier. In addition, the results indicated an average percolation flux through the sand base equivalent to 38.7 mm/y, which is consistent with the annual average flux obtained from the HELP modeling, equal to 35 mm/y (Tab. I-6). It should be noticed that the flux of water across the CB was low as a consequence of the presence of the compacted clay layer and its hydraulic conductivity, equivalent to 31.5 mm/y. Nevertheless, as it was observed in the results obtained from the HELP modelling for the CB design that during the dry months (i.e., from June to September) both, hydraulic head, and hydraulic gradients decrease. Consequently, it suggests a favourable condition for the proper working of the capillary barrier, and this new scenario was investigated in a subsequent model run. In this analysis the same finite elements mesh was used. The top boundary condition was set according to the annual average infiltration flux recorded from the HELP modeling for a dry year, equal to 100.4 mm/y. The remaining boundary conditions were maintained unchanged.

The results obtained from this analysis showed that even during the dry season the chosen CB design would not behave effectively as a capillary barrier. Flow modeling results indicated a flow of 29.4 mm/y percolating the cover, but the sand base showed no contribution to dissipating hydraulic head. The graphical results were similar to the results previously presented in Figure I-10.

Accordingly, the chosen CB design showed no capillary barrier behaviour in both scenarios evaluated under the particular climate conditions of the studied site.

Many authors [I-11] [I-12] agree that capillary barriers are generally considered functional only in regions of arid and semi-arid climates. Nevertheless, this does not imply that capillary barrier designs should be totally discarded for the site of Poços de Caldas in future analyses.

A possible suggestion is to increase the slope of the final cover in order to increase runoff and decrease water infiltration into the cover.

I-10. Radon exhalation modeling

I-10.1. Input Data

Table I-7 shows the input data required by the RADON program for modelling the diffusive exhalation rates of ^{222}Rn across the compacted clay barrier of the hypothetical final covers.

Table I-7. Input data for the RADON model

Parameter	Unit	Uranium Mill Tailings	Compacted Clay Layer
Layer thickness	cm	-	60
Porosity		0.30	0.445
Dry specific mass	g/cm^3	1.89	1.47
Specific ^{226}Ra activity	pCi/g	100	0
Gravimetric water content	%	10	0 to 30
^{222}Rn Diffusion coefficient	cm^2/s	$4.67 \cdot 10^{-3}$	function of gravimetric water content

In order to perform a conservative analysis, only the compacted clay layer was considered in the modelling. In addition, since thicknesses and material properties of the compacted clay layer are exactly the same for the three proposed final covers, the respective results are assumed to be the same for all designs evaluated in this work.

The porosity of the clay was the same as adopted in the HELP modeling. Gravimetric water content of the compacted clay was varied from 0% to 30% in increments of 5%. Since no field measurement of the radon emanation coefficient was carried out, a conservative value of 0.35 was assumed [I-13]. The relaxation length was also taken from the literature and is supposed to be equal to 200 cm, typical of uranium tailings impoundments [I-10].

Radon diffusion coefficients were calculated by the RADON program and vary as a function of both, degree of saturation, and porosity. The specific activities of ^{226}Ra in the tailings were set equal to 100 pCi/g (=3.7 Bq/g) and this value was obtained from previous field and laboratory measurements [I-2]. Although small concentrations of radioactivity in soils are frequently present, the clay material of the cover was supposed to be free of any.

I-10.2. Results and Discussion

Figure I-11 shows the relationship between ^{222}Rn exhalation rates, gravimetric water content and radon diffusion coefficient through the final cover.

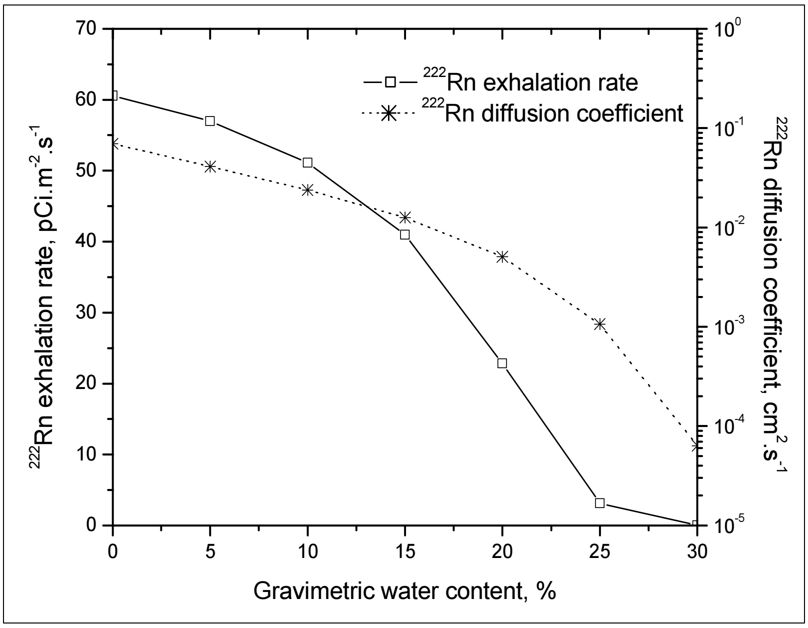

Fig. I-11. Radon-222 exhalation rates as a result of the variation of the compacted clay layer (60 cm thickness) gravimetric moisture content.

Both exhalation rates and radon diffusion coefficient decrease when the water content of the compacted clay barrier increases. It was observed, that the ^{222}Rn exhalation flux limit of 20 pCi/m^2·s (= 0.74 Bq/m^2·s) is only achieved when gravimetric water content of the compacted clay is equal to or higher than 21%. Below this value the radon exhalation rates can reach values twice or three times the flux limit of 20 pCi/m^2·s (=0.74 Bq/m^2·s). The ^{222}Rn diffusion coefficient through the compacted clay layer for a gravimetric water content of 21% is approximately $5 \cdot 10^{-4}$ cm^2/s.

I-11. Conclusions

The results indicated that oxidation of residual pyrite and the consequent acid leaching of radionuclides from the wastes are the driving force for the mobilization of radionuclide and metals in the tailings dam of Poços de Caldas. The processes are mostly restricted to the upper layers of the tailings (oxic zone); the deeper ones (anoxic zone) being an environment where radionuclides show a lower potential for migration. The remedial actions focused on the prevention of oxygen diffusion into the tailings as well as on the reduction of Rn exhalation.

Results obtained from the HELP model runs suggest that, in the long-term, different covers showed distinct behaviours concerning potential for desiccation and cracking of the clay layers due to wet and dry cycles. The study of the CB clearly illustrated this fact since annual average results indicated that the cover was properly designed to tackle long-term wet-dry cycles, whereas monthly results showed that the clay layer potential to desiccate and fail is considerably increased. It was also shown that the high efficiency of the CL is predominantly caused by the geomembrane liner that separates hydrological environments in the drainage layer above from the compacted clay liner below. For limiting the ^{222}Rn exhalation rate into the atmosphere to 20 pCi/m^2·s (=0.74 Bq/m^2·s), RADON modelling results indicated that gravimetric water content of the 60 cm clay barrier should be either equal to or greater than 21%. This value corresponds to a degree of saturation of approximately 70% for the chosen clay material.

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ANNEX II. CANADA

CAMECO RABBIT LAKE IN-PIT TAILINGS MANAGEMENT FACILITY — TAILINGS INJECTION TRIAL PROGRAMME

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II-1. ABSTRACT

The Rabbit Lake uranium mine is located approximately 800 km north of Saskatoon, Saskatchewan, Canada. The climate is characterized as continental sub-arctic with short and cool summers followed by long and cold winters. The Rabbit Lake in-pit tailings management facility (TMF) was developed from the mined out Rabbit Lake open-pit mine. Borehole investigation programs conducted since 1987 have shown the presence of frozen tailings layers within the facility. These frozen layers must be thawed prior to decommissioning of the TMF.

Cameco devised a program for thawing of frozen layers by deep injection of warm tailings. Since the spring of 2000, the primary means of tailings deposition has been deep injection. The tailings injection system, including injection well design, piping layout, control system and operational experience are described.

The results of an extensive borehole sampling program to investigate the geotechnical and geochemical impact of tailings injection are described. The investigation showed a positive impact on geotechnical properties with increased bulk dry density of the tailings due to thawing of frozen layers with subsequent consolidation. Geochemical concerns centred on the porewater concentration of several parameters (As, U, and ^{226}Ra) and whether or not concentrations would rise due to some chemical interaction of injected tailings with previously deposited tailings. The initial results appeared to indicate an effect on U concentration with no effect on As or ^{226}Ra concentrations. Subsequent investigations, reported here, showed that the apparent rise in U concentration was due to geochemical processes occurring in the sample containers prior to porewater extraction.

II-2. INTRODUCTION

The Rabbit Lake uranium mine is located approximately 800 km north of Saskatoon, Saskatchewan, Canada. The project is situated on the western shore of Wollaston Lake (58° 15^{2}N: 103° 40^{2}E). The climate is characterized as continental sub-arctic with short and cool summers followed by long and cold winters. The Rabbit Lake in-pit tailings management facility (RLITMF) was developed from the mined out Rabbit Lake open-pit mine. The design of the RLITMF included a pervious surround consisting of coarse-grained rock and sand filter material that was to be placed between the pit wall and the tailings. Mine tailings deposition began in 1985 and continues to this day. The tailings are composed of residues from the ore leaching process and precipitations from acid neutralization. To the end of December 2002 more than 5.2 million tonnes of tailings solids were placed in the RLITMF. The tailings body was approximately 425 m long and 300 m wide at the surface with a depth of about 100 m at the centre.

Drilling and sampling programs have been conducted in the Rabbit Lake In-pit TMF (RLITMF) in 1987, 1990, 1992, 1993, 1994, 1997, 1999, and 2001. The purpose of these programs has been to gather information on the geotechnical and geochemical condition of the tailings.

II-2.1. Geotechnical Conditions

The aforementioned investigation programs have shown that layers of frozen tailings have developed in the TMF. These layers correspond to tailings deposited during each winter season. The frozen layers impede the consolidation of tailings due to immobilization of frozen pore water and by reduction of the overall vertical hydraulic conductivity throughout the tailings body.

The design of the RLITMF is based on the concept of dominantly diffusive transport of contaminants from the facility to the groundwater flow system after decommissioning. In order to achieve this, the tailings matrix must be relatively impermeable (tailings 'plug') and fully consolidated, at the time of decommissioning. The frozen layers would prevent full consolidation of the tailings; therefore, some method of thawing them is required. Likewise, the long-term environmental loading from this facility is dependent on the source concentrations of various parameters; therefore, the geochemical condition of the tailings must be controlled and monitored.

After examining several options, Cameco devised a program for thawing of frozen layers by deep injection of warm tailings. Initial injection tests first performed in the mid 1990s. The current full-scale injection program was first operated on a trial basis starting in 1999, with follow-up investigations of tailings properties. Since the spring of 2000, the primary means of tailings deposition has been deep injection. Section II-2 describes the tailings injection program and its impact on the geothermal/geotechnical condition of the tailings.

II-2.2. Geochemical Issues

The geochemical data from borehole investigations have shown some vertical stratification in the concentration of various chemical parameters of concern (arsenic, nickel, ^{226}Ra) due to the milling of several ore bodies with different characteristics. The data also indicates that within any of these zones the concentrations have been quite stable over time.

Arsenic contained within the tailings is one of the primary contaminants of concern in terms of potential to affect downstream receptors. A detailed description of an investigation program into the long-term evolution of arsenic in tailings porewater may be found in [II-1] and [II-2]. Additional papers in this series will be released in 2003 and 2004.

A second geochemical concern was whether or not the injection of warm tailings at depth would cause increased porewater concentrations of any contaminants. This issue was addressed in the 1999 and 2001 borehole investigation programs.

II-3. TAILINGS INJECTION PROGRAMME

II-3.1. Tailings Injection System Design

II-3.1.1 General description

The 1999 trial injection system was composed of 4 major components: injection wells; injection pads; a piping system; and an operation and monitoring system. Injection pressure for the system was supplied by a static head difference of 85 m between the mill and the tailings surface.

Three pairs of angled injection wells were installed at 3 injection pads located along the southwest to southeast perimeter of the pit, as shown in plan view on Figure II-1. The injection wells were installed with the discharge point located beneath the lowermost frozen layer at about 60 m below the tailings surface. A pipeline system with appropriate valves and controls was installed to deliver tailings to the wellheads. The tailings flow was divided between two injection wells at any time.

II-3.1.2 Injection pad design and construction

The positions of the injection pads were chosen to maximize borehole separation from pit walls while minimizing borehole length. Three injection pads were used in this trial program. Pad size was governed by the space required for a drill rig to install the wells. Injection pads were constructed by pushing sand onto the tailings surface until a stable base was achieved. The pads were then protected with geotextile and covered with additional sand.

II-3.1.3 Injection well design

A set of design criteria for the injection wells was established, based on the objectives of prevention of damage to the pervious surround system, achieving the required flow capacity, and injection below the lowest frozen layer. The primary criteria was that the endpoint of the injection wells must be located a minimum of 10 m from the nearest point of the pervious surround to ensure that injected tailings would not impact the pervious surround. Secondly, during drilling of the injection borehole, the drill stem was not allowed to pass through the filter sand portion of the pervious surround. Therefore, in the middle depth portions of the borehole a minimum separation distance of 5 m between the casing and the pervious surround was specified. Finally, the elevation of the injection points was set based on the elevation of the lowermost frozen layer found in the 1997 borehole investigation of the facility.

The as-built injection well locations are shown in plan view in Figure II-1 and a typical cross-section view in Figure II-2. Injection points were spaced about 50 to 70 m apart laterally to avoid interference from adjacent wells. The bottom elevation of the wells was about 60 m below the tailings surface.

The injection wells consisted of 100 mm diameter (4 in. HWT) steel well casing installed using a diamond drill rig. Water, amended with drilling mud as required, was used as the drilling fluid. At critical times during drilling (just prior to making the closest pass to the pervious surround), the dip angle of the borehole was measured by acid-testing to ensure that the well maintained a safe distance from the pervious surround. After completion of each injection well, a final acid test was done to determine the horizontal and vertical position of the injection point.

II-3.1.4. Piping system design

The tailings pipeline distribution system was extended from the existing south discharge line to six injection wellheads located at 3 drill pads, as shown in Figure II-1.

A four way splitter fitting was installed at the end of the main tailings line to divide flow between: the two branches of the injection pipe manifold, a 10" (25 cm) diameter sub-aerial bypass line, and a 4" (10 cm) diameter blow-out line equipped with a 145 psi (999 kPa) rupture disc. The purpose of the rupture disc was to prevent tailings from backing-up into the mill, in the event of a plugged line and to protect the main line above this point from over-pressure conditions.

A piping control system was installed at each injection pad, to divide flow between two wells or to allow sub-aerial bypassing of tailings. Pressure gauges were installed at each wellhead and at the main feeder line (four way splitter fitting). Butterfly valves were positioned throughout the system to direct flows to the appropriate manifold sections, sub-aerial bypass lines or injection wells.

II-3.1.5. Operational procedures

A set of operational procedures were designed to: ensure that a consistent and orderly process was followed during each injection cycle, to prevent plugging of tailings pipelines, protect tailings lines and injection wells from freezing during non-operational periods, prevent spillage of tailings outside of the pit pervious surround and prevent damage to injection system components due to upset operation conditions.

II-3.1.6. Monitoring procedures

Monitoring procedures were designed to measure pressures and flows during injection for documentation of operating characteristics, determine the location of fresh tailings break-through points at the surface of the existing tailings, develop techniques for detecting upset conditions, and gather operational data for system optimization and general record keeping.

II-3.2. Operating Summary And Discussion

II-3.2.1. System start-up

Prior to the first use of the wells, the breakout pressure, or maximum pressure required to cause fluid injection, was determined. Breakout pressures ranged from 275 kPa at IW-2 and IW-4 to 830 kPa at IW-3. The minimum pressure recorded after breakout had been achieved, was 175 kPa at IW-1, IW-2, IW-4 and IW-5. The maximum sustained pressure, after initial breakout, was 275 kPa at IW-6.

The injection system was started for the first time on July 10, 1999 at IW-1 and IW-2. With the exception of IW-6, initiation of injection was relatively easy at all wells. In the first injection cycle the pressure required to initiate flow ranged from 350 kPa to 770 kPa. In all cases, full injection flow was achieved within 1 to 4 minutes of wellhead pressurization.

In subsequent start-up events, the peak pressure before injection started was typically less than 350 kPa and flow initiation occurred almost immediately.

Tailings injection at IW-6 could not be initiated in 1999 due to excessive wellhead pressure. During servicing of the well in May 2000, it was found that a large rock had become wedged in the casing shoe (during original installation), effectively plugging the pipe.

II-3.2.2. Injection pressures

Upon initiation of flow in the wells, wellhead pressures dropped to a typical range of 175 kPa to 245 kPa and remained relatively stable during any operating period. Occasional maximums of up to 420 kPa and minimums of 85 kPa occurred, due to variations in the tailings flow rate from the mill.

II-3.2.3. Upwelling tailings

Tailings injection resulted in upward flow or 'upwelling' of slurry/water to the tailings surface in various locations, typically within the central part of the injection test area. The upwelling occurred in distinct areas of about 1 to 2 m in diameter.

The first indication of upwelling tailings occurred near Pad #2 about 3 hours after injection started at IW-3 and IW-4. This took the form of small boils (2 to 5 cm in diameter) above the centerline of the injection wells. Shortly after the flow was switched back to IW-1 and IW-2 (Pad #1) upwelling began to occur in standing water about 30 m offshore from Pad #3. Upwelling material initially consisted of a dark gray foamy substance; this was followed by reddish brown slurry consistent with the injected tailings colour.

Upwelling continued throughout the remainder of the program with only short interruptions. The primary upwelling point started near Pad #3, but gradually moved to a position closer to the centre of the pit.

On a few occasions, upwelling would stop for a few hours. The tailings surface was observed to be swelling or doming on two of these occasions. When the pressure within the tailings had reached a critical level, tailings slurry initially shot into the air up to 1.5 m above the surface, then gradually declined to a steady 'boil'. In association with these events, large blocks (3 m to 5 m side lengths) of ice/frozen tailings came to the surface.

The total suspended solids (TSS) of water pumped from the pit seepage collection system remained low (normal) throughout the injection project.

The tailings surface is normally partially covered (25%) by water throughout the summer. During the injection project, the water covered area increased to about 85 to 90% of the tailings surface.

II-3.2.4. End of season

The 1999 injection system was not designed for winter operation. Difficulties with frozen lines, gauges and valves occurred during the week of October 1 to October 7. Therefore, the trial injection program was shut down on October 7,1999.

II-3.2.5. Summer 2000 programme

The injection wells required servicing prior to usage in the summer of 2000. All injection wells were cleaned using a diamond drill rig. Pressure testing revealed that two wells (IW-1 and IW-2) had broken. The remaining wells (IW-3 to IW-6) were pressure tested and found to be intact.

It was postulated that the well casing at IW-1 and IW-2 had broken due to large settlements caused by thawing of frozen layers. This settlement would have occurred progressively over the winter of 1999/2000.

Injection of warm tailings was resumed on June 10, 2000 and continued to October 15, 2000, with two short interruptions. During the summer one additional well (IW-4) also failed.

In other respects, tailings injection proceeded smoothly during the summer of 2000. The mill operators found the system easy to operate. The pit service crew found that less maintenance work was required on the pervious surround in comparison to sub-aerial deposition.

In contrast to 1999, when tailings were observed to upwell quite vigorously throughout the program, upwelling was minor in 2000. The water level on the tailings surface increased significantly, as it did in 1999, although a large portion of the change may have been due to other unrelated causes.

II-3.2.6. Winter 2000/01 injection programme

Background

The tailings injection system was modified for winter operation by reducing the system to one pair of wells and enclosing the wellheads in a heated building. Two new injection wells were installed using a sonic drill rig.

The targeted bottom elevation for the wells was approximately 348 m above sea level; however, based on acid-test data for the wells; the actual elevation of the injection points was about 360 m above sea level.

The piping system was simply a scaled-down version of the system used in 1999 [II-3], with the addition of heat tracing lines to prevent freezing during non-operational periods.

Operational Summary

The winter injection system was started for the first time on October 27, 2000. Operations were shut down on May 31, 2002 when the mill was put on extended shutdown.

The winter injection system operated smoothly until December 23, 2000, when it was observed that one of the injection wells (IW-8) was blocked. This caused excessive pressure in the tailings line. Throughout the month of January 2001, part of the tailings flow was injected and part was discharged onto the tailings surface. After the mill was put into normal weekly shutdown on January 25, the injection wellhead piping and building were removed. The sonic drill rig was used to clean the hole by drilling out the tailings sand that had blocked the hole. During removal of the drilling rods, it was necessary to pump additional mud into the well and withdraw the rods slowly to prevent backflow of tailings sand into the well. This pair of wells remained usable until the summer of 2003.

During the winter it was not possible to see the full extent of upwelling tailings due to the snowcover on the surface. In the spring a large circular cone became visible.

II-3.2.7. Injected volume of tailings

The total volume of tailings slurry injected during the 1999 season was approximately 104,500 m^3, based on mill production records and well operating periods.

The total volume of tailings slurry injected during the 1999 season was approximately 104,500 m^3, based on mill production records and well operating periods.

It has been found through various tailings investigations, that shortly after subaerial deposition, the void ratio of the tailings is typically about 4. The tailings volume, shortly after deposition, assuming a void ratio of 4 was 85,300 m^3. This volume of tailings, spread over the entire upper tailings surface, would have raised the tailings level by about 0.75 m. The increase would have been about 1.5 m if confined to the area directly affected by tailings injection.

During the course of the trial program (July 1999 to May 2001) approximately 289,000 tonnes of tailings slurry were injected. This compares to a total tailings production of about 501,000 tonnes during the period. It should be noted that while injection was being practiced, close to 100% of the flow was being injected.

II-3.2.8. Tailings surface surveys

The tailings surface in the vicinity of the injection program was surveyed prior to the start of the injection program on June 26, 1999. Contour data from the pre-injection survey are shown on Figure II-3a. The contours show a deltaic landform with smooth contours, as would be expected due to sub-aerial discharge of tailings from a spigot.

A post injection survey was performed on October 24, 1999. Contour data from the post injection survey are shown on Figure II-3b. This figure shows a remnant deltaic landform in the southwest part of the pit; however, the surface shape over the rest of the pit surface has changed significantly. Circular contours near the centre of the facility indicate the location of a steep cone shaped depression in the tailings surface. Upwelling tailings were observed in this area during the last week of injection.

In summary, the post-injection survey showed that the overall tailings surface elevation had changed very little, in comparison to the expected increase based on injected volume as discussed in Section 2.2.8. This indicates that injection was causing some thawing resulting in accelerated consolidation.

II-4. BOREHOLE INVESTIGATIONS

II-4.1. Introduction

A borehole investigation was conducted in the injection area near the end of the 1999 injection season to determine the effects of tailings injection on tailings geotechnical and geochemical properties.

A total of 5 test boreholes (RLP99-101 to RLP99-104 and RLP99-106) were drilled. The depth of investigation ranged from 22.7 m to 71.6 m below the tailings surface. The results of this program have been reported in [II-3].

A more extensive borehole investigation program was conducted in July and August of 2001, as reported in [II-4]. A total of 11 test boreholes (2001-01 to 2001-11) were drilled in the Rabbit Lake In-pit Tailings Management Facility (RLITMF) at locations shown on Figure II-1.

The objectives of the 2001 In-pit investigation programme were to:

- Investigate the impact of the tailings injection trial on the geotechnical, thermal and geochemical condition of the tailings mass; and
- Compare current conditions with previous conditions in areas not impacted by tailings injection.

The depth of investigation ranged from 5.2 m to 70.0 m below the tailings surface. Borehole locations were chosen based on proximity to previous investigation boreholes and proximity to tailings injection zones.

II-4.2. Field Investigation Methodology

II-4.2.1. Drilling

The 2001 drill program was conducted using several procedures developed in previous programs. Sonic drilling technology previously proven to be the most suitable for drilling in soft unconsolidated tailings was used.

The Cameco sonic drill rig was mounted on a steel pontoon barge. The barge was positioned at each proposed borehole location using a system of cables, winches and anchors. A separate cable system was used to tow a flat-bottomed boat back and forth from the barge to shore. The flat-bottomed boat was used to move men and supplies to and from the drill rig.

II-4.2.2. Solids Sampling And Core Recovery

The combined use of a core barrel and piston sampler provided a high percentage (60 to 80%) of core recovery over the investigation depth, (refer to [II-4]) for a detailed description of coring methods).

Tailings samples were collected at regular intervals, as well as at points of obvious changes in visual appearance. Samples for geotechnical, (MC-1 to MC-259) and geochemical (GC-1 to GC-119) analysis were sealed in 1 litre or 500 ml plastic containers and stored in the dark at 4°C, until being submitted for analysis. A separate set of small samples, labeled Ni-1 to Ni-122 were collected for the specific purpose of investigating any shift in solids composition that may have occurred due to tailings injection. Samples for this purpose were collected in 250 ml plastic containers with tight fitting lids.

II-4.2.3. Field Measurements

Field measurements of insitu temperature, pH, and Eh were made on tailings solids samples within 15 minutes of recovery from the borehole. An Orion Model 250 combination pH/Ion meter was used. An Orion combination pH/ATC electrode was used for pH measurements and temperature measurements, while a Cole-Palmer ORP platinum electrode was used for Eh measurements. Calibration of the probes was completed twice daily using certified buffer solutions (pH 4, 7, and 10) and a +300 mV ZoBell's solution with the meter corrected to the standard hydrogen potential. Electrodes were inserted directly in the core samples and held there until a stable reading was displayed.

Core temperature was measured using a thermistor probe connected to a multi-meter. This apparatus was calibrated to read within 0.01°C and the meter recorded resistance to within 0.01°C.

II-4.3. Laboratory Testing

II-4.3.1. General

All samples (259) in the MC series were subjected to moisture content determination. A total of 28 GC series samples were subjected to porewater extraction followed by chemical analysis of the porewater and solids.

II-4.3.2. Laboratory Methodology

Tailings porewater was obtained by piston squeezing of 28 core samples. The piston squeezing method for porewater extraction has previously been described in [II-1].

The storage period for these samples was longer than originally anticipated, due to a mix-up in laboratory procedures. The sample squeezing process was ultimately completed in January of 2002, meaning that samples were in storage for up to six months prior to processing and chemical analysis. Porewater samples extracted from core specimens at Rabbit Lake were sent to the Saskatchewan Research Council laboratory for chemical analysis, along with the corresponding solids material from which the porewater was extracted.

Moisture content determination was performed in the Rabbit Lake on-site laboratory using ASTM Standard D2216-92. Grain size analyses were also performed in the Rabbit Lake laboratory according to ASTM Standard D 2217-85.

II-4.4. Geotechnical results and discussion

II-4.4.1. Temperature and frost distribution

The temperature profile at borehole 2001-06 illustrated in Figure II-4(left) is quite different from the historical temperature profile (97-R2) for this area. The upper 27 m of the profile is fully thawed, while below this, only two remnant ice layers are found to a depth of about 53 m. The impact of tailings injection at borehole 2001-05, a short distance away from 2001-06, was less dramatic. In this case, the upper zone was thawed, but at greater depths the temperature was more variable.

The temperature profile at borehole 2001-10 is shown in Figure II-4(right) along with historical temperature data from 1997 (borehole 97-R3). The temperature traces for 1997 and 2001 show a strong similarity with alternating zones of positive and slightly negative temperatures. This plot shows that borehole 2001-10 has not been affected by injection of warm tailings.

The temperature profiles discussed above show good correlation with visual descriptions noted on the borehole logs. The data indicate that the impact of injection is positive in terms of removing frozen layers, but that this impact is somewhat random.

II-4.4.2. Moisture Content

Plots of moisture content (dry basis) versus depth for boreholes 2001-06 and 2001-10 are shown in Figure II-5. The 2001 borehole data have been plotted with moisture content data from the 1997 program. The 1997 data is plotted at the elevation found in 1997, which means that a given layer would actually exist about 3 to 4 m lower in 2001 due to on-going consolidation of the tailings.

The area around BH 2001-06 was subject to a major impact from the tailings injection program. This is most clearly evident (Figure II-5(left)) in the zone from about 355 masl (metre above see level) to 382 masl where it was previously noted (Section 3.4.1) that all frozen layers had thawed. The geometric mean moisture content in this zone in 2001 was 58.6%, while in 1997 (97-R2); it was 129%, (including frozen layers). It may also be noted that above 382 masl, the overall mean moisture content, (including frozen layers) was reduced from 125.9% to 67.4% between 1997 and 2001.

The moisture content profile data from BH 2001-10 (Figure II-5(right)) do not indicate a noticeable change since 1997. However; as was the case for other boreholes in 2001, the surficial material has a lower moisture content than previously observed for freshly deposited tailings. Typically, freshly deposited tailings (non-frozen) have a moisture content of 80% to 100%. The surficial tailings at 2001-10 were deposited during the injection program.

II-4.4.3. Dry Density

Plots of calculated dry density versus depth for boreholes 2001-06 and 2001-10 are shown in Figure II-6. Dry density was calculated from the moisture content using the following formula:

$$\rho_d = \frac{G_s}{1 + \frac{w}{100} \bullet G_s}$$

where ρ_d is dry density, w is the moisture content (dry basis) and Gs is the particle specific gravity, (estimated to be 2.75). Since dry density is computed from the moisture content, much of the discussion relating to trends in moisture content profiles is applicable to the dry density profiles.

Historically, the dry density of non-frozen tailings has shown a steady increase with increasing depth. The near surface density was typically in the range of 0.6 to 1.0 Mg/m^3, while the density at 50 to 70 m depth was in the range of 1.2 to 1.5 Mg/m^3. In contrast, the 2001 data for all boreholes show near surface (upper 10 m) dry densities of 1.0 to 1.5 Mg/m^3.

Dry densities in a profile most affected by tailings injection (2001-06, Figure II-6(left)) are also higher than in the 1997 investigation. In BH 2001-06, the density in the interval from 390 masl to 397 masl has shifted from the range of 0.6 Mg/m^3 – 1.0 Mg/m^3 to a range of about 1.1 Mg/m^3 to 1.3 Mg/m^3. This represents a unit volume reduction of between 10% and 50%.

The dry density of non-frozen tailings at borehole 2001-10 (Figure II-6(right)) has increased slightly at all depths when compared to 1997; this is a consequence of on-going consolidation. However, this borehole does not show the large increase of dry density that was observed at 2001-06.

The dry density of frozen tailings is highly variable over the full depth of burial. Dry densities of less than about 0.4 g/cm^3 are generally indicative of a zone that is mostly ice.

A portion of the density increase may be due in part to two other factors. First, the ore grade has declined from 1.5% U_3O_8 in 1997 to 1.2% U_3O_8 in the last 2 years of operation. A lower ore grade results in the formation of less precipitates, notably gypsum, which are of lower density than ore residues. Second, the production rate in 1999 to 2001 was about 230,000 tonnes per year, compared to an average of 350,000 tonnes per year from 1992 to 1998.

II-4.5. Geochemical Results And Discussion

II-4.5.1. Introduction

Twenty-eight wet tailings samples were selected for extraction of porewater and subsequent chemical analysis of the porewater and solids. In addition to this, 122 samples of tailings solids were collected for analysis of Ni content only.

The following discussions examine the concentration of As, Ni and U in the pore fluid and solids in more detail.

The primary objective in the 2001 geochemical investigation of the tailings was to determine if any critical parameter concentrations were being adversely effected by tailings injection. A secondary objective was to determine if any of the injected tailings were being retained at depth, or if the full flow was coming to the surface.

II-4.5.2. Arsenic

Figure II-7(left) is a plot of As concentration in the tailings solids versus elevation for all boreholes in the 1997, 1999 and 2001 investigations. Also included on the plot is a vertical line showing the average As concentration of the tailings slurry, as discharged, since July 1999. The data show that 2001 solids concentrations are in general agreement with data from 1997 and 1999. There is one zone from about 365 masl to 375 masl where the As concentration may be lower than in previous investigations. This may be due to injection of tailings with a lower As concentration. The surficial tailings show an As concentration approximately equal to that of the injected tailings.

Figure II-7(left) also shows that As concentrations may be grouped into three broad zones. Below about 380 masl the concentration varies from about 1,000 mg/g to about 20,000 mg/g. In the zone from 380 masl to about 395 masl, the concentration is much lower, in a range from 20 mg/g to about 2,000 mg/g. Finally, in the zone above 395 masl, the concentration is relatively constant between 1,000 and 2,000 mg/g, with one exception. At an elevation of about 392 masl there are 3 isolated points with a high As concentration. These zones correspond to various ore sources: the lower zone is typical of B-zone ore, the middle (low As) zone is due to Eagle Point ore alone and the upper layer is due to mixed A-zone and Eagle Point ores. The exception noted in the upper layer is due to deposition of D-zone ore tailings (with high As concentration) in June of 1996.

Figure II-7(right) shows the concentration of As in the pore fluids from all samples in the 1997, 1999 and 2001 investigation programs. Above elevation 360 masl, the As concentration in porewater generally varies from about 500 mg/l to about 22,000 mg/l, although, there is a zone at about 390 masl where the concentration is higher, this corresponds to the aforementioned zone of D-zone tailings. Below elevation 360 masl the As porewater concentration varies from 3,000 to 237,000 mg/l. This corresponds to the zone of highest As solids concentrations. This zone has been discussed extensively in previous reports [II-1] [II-3] [II-5] [II-6]. The pattern of porewater concentration versus elevation is consistent with past results, suggesting that the system is relatively stable. There is also no indication that tailings injection has caused any disturbance in the As porewater concentration profile.

II-4.5.3. Nickel

Previous tailings investigation programmes [II-3][II-5] have shown that the nickel solids concentration profile of the tailings body is quite distinct. This fact was used to investigate whether or not significant quantities of injected tailings were remaining at depth. The nickel concentration of freshly injected tailings throughout the period from the beginning of the trial injection program in 1999 to the end of the program in 2001 averaged 1,080 mg/g. This concentration is significantly lower than that of the in-situ tailings over much of the profile. Accordingly, in the 2001 investigation program, 122 samples were collected and analyzed for nickel solids content.

Figure II-8(left) shows the Ni profile for BH 2001-10, a location little affected by tailings injection, plotted with the data from 1997 and 1999. A vertical bar on this graph indicates the average nickel concentration of tailings produced since July of 1999, when the tailings injection trial first began. This figure clearly shows that the 2001 Ni concentration profile is very similar to the 1997 and 2001 profiles. In contrast, Figure II-8(right) shows the profile for BH 2001-06 in comparison with the historical data from 1997 and 1999. This suggests a shift in Ni concentration due to dilution of existing tailings by injected tailings. The geometric mean Ni concentration between elevation 360 masl and 378 masl for BH 2001-06 is 587 mg/g compared to a mean of 3,347 mg/g in the same interval for the 1997 and 1999 data. The BH log for 2001-06 specifically mentions visual evidence of mixed tailings colours in the zone from 370 to 376 masl.

Nickel porewater concentration has not been a concern relative to other porewater parameters in this TMF.

II-4.5.4. Uranium

Uranium analysis results for samples collected in the 2001 Investigation Program are plotted versus elevation in Figure II-9. This plot shows 3 clusters based on elevation in the RLITMF. There is a large group of data clustered in a range from 0 to about 1,000 mg/l below an elevation of 365 masl. The second zone from 365 to 385 masl has concentrations of up to 6,000 mg/l. In the upper zone, from 385 masl to the surface (407 masl), the concentration varies up to about 13,000 mg/l. The lower zone in this case corresponds to tailings derived exclusively from B-zone ore, while the middle and upper zones have variable fractions of Eagle Point, A-zone, B-zone and D-zone ore tailings.

The uranium concentration profile in 1997, from a smaller number of samples, is also shown on Figure II-9. This data shows only one grouping. The concentration is less than 2,000 mg/l over the full depth. This apparent contrast between the 2001 and 1997 results indicates that U porewater concentrations have been increasing since 1997. The possibility that injection was responsible could not be discounted immediately.

II-4.5.5. Summary of 2001 investigations

The primary focus of the 2001 investigation program was to assess the impacts of tailings injection on the geotechnical and geochemical conditions of the tailings in the facility.

The investigation results indicated positive impacts on geotechnical properties. These impacts include: thawing of previously frozen tailings, reduced freezing of newly deposited tailings, reduced beach lengths and increased density of freshly placed and newly thawed tailings.

With respect to geochemical properties, there was a shift in the solids concentration of some metals (principally Ni and As) within the profile due to mixing associated with the injection of tailings from an ore source with low Ni and As levels. Additionally, it was observed that Uranium concentrations in porewater were higher than in the 1997 investigation.

Cameco performed additional follow-up work to determine the cause of this apparent increase in U concentration

II-5. FOLLOW-UP TO THE 2001 INVESTIGATION

II-5.1. Potential Causes

As discussed in Section 3.5.4 above, the uranium in porewater concentration appeared to increase between 1997 and 2001. Several potential causes for the apparent increase in uranium porewater concentration were suggested; these included the method of porewater extraction, depositional history, injection effects and diversion of contaminated ditch water to the RLITMF.

II-5.1.1 Porewater extraction method

The original investigation report [II-4] suggested that a change in the method of porewater extraction from 1997 to 2001 might have caused the apparent shift in U concentration. The 1997 porewater was extracted by decantation from tailings cores and by sampling from temporary piezometers installed in the tailings body. The 2001 porewater was extracted by hydraulic squeezing of the samples to force porewater from the solids matrix. It was thought that this rather aggressive method of squeezing the samples might have contributed to the higher concentrations of U. This hypothesis was investigated by extracting porewater from several additional tailings samples collected in the 2001 program using a variety of techniques. These samples had been stored under refrigeration since being collected in 2001.

Porewater was extracted from the tailings solids by 3 methods: decantation (where possible), hydraulic squeezing and centrifugation at the University of Saskatchewan Department of Geological Sciences. The extracted porewater was sent to the SRC for analysis. The results of this experiment showed that all 3 methods of porewater extraction produced essentially the same concentration of uranium. Therefore, the porewater extraction method is not a possible cause of the apparent concentration shift for U between 1997 and 2001. A full listing of results may be found in [II-7].

II-5.1.2 Injection effects

The possible impact of tailings injection on U porewater concentrations was examined by comparing the porewater chemistry results for samples from the vicinity of injection wells with the results for samples remote from the injection location. It was observed that even boreholes located far away from the injection wells were showing elevated U concentrations. In order to exclude the possibility that porewater from the injection program had somehow migrated to the far end of the pit, a concerted effort was made during the second round of laboratory analyses to select samples from frozen zones. The results indicated that even frozen porewaters had elevated U concentrations. These frozen porewaters were derived from depths and locations where the tailings would have been deposited well before tailings injection began.

The evidence shows that tailings injection has not been the cause of this shift in U concentration.

II-5.1.3 North Drainage Ditch water

The RLITMF has always been the receptor of seepage originating from the West #5 waste rock stockpile. Up until 1997, this seepage entered the system only by groundwater seepage to the pervious surround. Groundwater entering the pervious surround is conducted directly to the bottom of the facility without mixing with the tailings; therefore, prior to 1997 the seepage had no possible effect on the tailings. In 1997 water from the North Drainage Ditch (NDD), which originates as seepage from West #5, was diverted directly to the RLITMF surface near the north end of the pit.

The uranium concentration in the NDD water has been steady within the range of 5,000 to 15,000 mg/l since 1989. It is conceivable that mixing of this water with tailings porewater could result in some elevation of porewater concentrations; however, this water would not tend to mix with porewater due to the on-going process of consolidation. Porewater is continually expelled from the tailings during consolidation, thus making it very unlikely that surface water would enter the tailings matrix. The possibility of mixing of NDD water with the tailings was also addressed from the point of view of water quality by a geochemical consultant.

II-5.1.4 Tailings neutralization change

The terminal pH of tailings being discharged to the pit was changed in 1999 after an extensive laboratory and metallurgical test program [II-8] indicated that a reduction in the terminal pH from 10 to 8.5 would result in a more stable form of arsenic being precipitated (iron arsenates versus calcium arsenates). The testwork showed small increases in some other metals would occur due to the lower pH.

According to the report on pH adjustment testwork [II-8], the U_3O_8 concentration in tailings filtrate prior to the start of the testwork was 1.6 mg/l. After full implementation of the pH profile adjustment, the U_3O_8 concentration was 2.7 mg/l. This small change would not be sufficient to account for the large change being observed in the tailings.

The Metallurgy Department at Rabbit Lake collects a daily sample known as RLPD (Rabbit Lake Pit – Dissolved) from the tailings discharge stream. The data for RLPD are quite variable over the entire period from 1995 to 2001, but become more variable and higher after the Mill pH was adjusted in 1999. The average U_3O_8 concentration in tailings discharge after July 1999 has varied between 1.45 and 10 mg/l. These higher concentrations in the discharge could be partly responsible for the generally higher U concentrations found in the pit above an elevation of 403 masl and in zones of injection. However, this would not explain any changes occurring in areas remote from the injection area or below 403 masl or frozen in place prior to 1999. Therefore, the change in tailings neutralization process can be eliminated as a possible cause of the change in the U concentration profile from 1997 to 2001.

II-5.2. Geochemical Consultant Report

Cameco retained SRK Consulting to review and assess the data from the 2001 investigation and to perform geochemical modeling of the tailings porewater chemistry. In summary, SRK concluded that:

Elevated uranium concentrations of the 2001 samples (in comparison to previous sampling events) were due to carbon dioxide interaction with the tailings samples. This interaction occurred during a lengthy period of storage after sample collection. The 2001 samples (those reported in [II-4]) were held in storage from July/August of 2001 to January of 2002 prior to porewater extraction. In contrast, the porewater from the1997 samples was decanted within a few weeks. SRK compared the field pH of samples with that recorded at the time of analysis. The results showed a general decrease in pH confirming contact with atmospheric carbon dioxide. Geochemical modeling also indicated that sample contact with atmospheric CO_2 would cause dissolution of uranium, together with a decrease in pH and an increase in dissolved inorganic carbon.

The possibility that North Drainage Ditch (NDD) water may have caused an impact on tailings porewater quality was also investigated by SRK. They constructed a series of Piper plots to compare and contrast the water quality of the tailings with that of the NDD. From this analysis, they concluded that NDD water was not responsible for a shift in tailings porewater quality. A copy of the full SRK report may be found in Cameco [II-7].

II-5.3. Follow-up to Consultant Report

Tailings porewater was extracted from the 2001 solids samples by the University of Saskatchewan Department of Geological Sciences. The first group of samples was sent to the University shortly after sample collection. The samples were squeezed over a period of a few weeks in the fall of 2001, with all work completed by November 23, 2001. (The average time from collection to extraction and preservation would have been about 3 months.) However, at this point, it was discovered that the solids portion of each sample had been discarded after squeezing. It was decided that it would be better to start with a new batch of samples so that porewater concentrations could be related to solids concentrations, if necessary. Accordingly, a second group of samples was sent to the University. These samples had been stored in a refrigerated condition at Rabbit Lake since they were collected. The elapsed time from sample collection to porewater extraction was about 6 months. The data from this group of samples was discussed in Section II-3.5.4.

In August of 2002, Cameco was informed by SRC that porewaters (preserved) from the first group of samples (solids discarded, as discussed above) were still in storage at their laboratory. This group was then analyzed for U concentration. These samples represent porewaters extracted after 3 months of storage.

The porewater samples extracted to compare extraction methods, as discussed in Section II-4.1.1, were in storage for approximately 12 months prior to extraction.

Following the report by SRK Consulting, Cameco re-examined the concentration data to see if the results of SRK's analysis could be corroborated. The uranium concentration data for all samples analyzed have been re-plotted in Figure II-10 with uranium concentration on the horizontal axis and elevation within the RLITMF on the vertical axis. Data for samples analyzed after 3, 9 and 12 months are plotted with unique markers. This plot clearly shows a stepwise increase in U concentration for samples stored from 3 months to 6 months to 1 year.

Further to this, Cameco conducted a second set of confirmatory tests. A large sample of fresh tailings was collected from the mill on March 12, 2003. The sample was split into several parts. Each of these splits was stored in a plastic container of the variety used in the 2001 investigation and stored in the same fashion. Samples of porewater were extracted by piston squeezing at intervals of 1 day, 26 days, 85 days and 198 days. The concentration of U and

HCO_3 in porewater from each of these extractions is plotted in Figure II-11. The plots clearly show a rise in both U and HCO_3 over time, this result further corroborates the analysis by SRK.

II-5.4. Summary of U investigation

The apparent rise in Uranium concentration of the RLITMF porewater was the result of a lengthy period of storage prior to extraction of the porewater from tailings core samples. SRK concluded that samples collected in 2001 were affected by atmospheric carbon dioxide, which caused a decrease in pH and an increase in dissolved uranium concentrations.

This work shows that drill core samples collected for extraction of tailings porewater should be processed as soon as possible, with a target of completing all extractions in 2 weeks or less.

II-6. SUMMARY AND CONCLUSIONS

A number of observations may be drawn from the warm tailings injection program begun in 1999.

II-6.1. Operational

The injection system was easy to start and required little attention during operation. At the start of each mill cycle it was simply a matter of diverting the tailings flow to the wellheads and then opening the valve at the wells. Injection pressures were stable over periods of hours and days.

Injection start-up pressures tended to decrease with each operating cycle. This suggests that once a well has been “conditioned” by establishing flowpaths in the tailings body, the expansion of those flows paths on each cycle becomes easier. Operational experience has shown that blocked wells can be serviced in a reasonable time using a small sonic drill rig.

The average injection pressure during continuous injection declined slightly over the duration of the program. This may indicate that once the formation has been fractured (opened), flowpaths remain open for extended periods.

Tailings injection did not have an observable effect on the pervious surround, as measured by the TSS of seepage water collected from the pit drainage system.

Survey data indicate that the tailings surface elevation declined in areas that were not subject to accumulation of upwelling tailings solids. This decline is partly attributable to normal on-going consolidation, and partly to the injection project. Settlement caused by the injection project may be partly due to release of water trapped beneath frozen layers and/or due to melting of ice within frozen layers.

The greater than usual amount of water on the tailings surface conforms with the assertion that tailings injection caused the release of trapped water or caused melting of ice in frozen layers.

II-6.2. Geotechnical

In a typical profile that has not been subject to any active thawing measures, the stratigraphy consists of alternating layers of frozen and non-frozen tailings with occasional layers of essentially clear ice. The non-frozen layers generally become progressively more consolidated with depth (time since deposition).

In areas that have been subject to the injection of warm tailings, several of the frozen layers corresponding to winter deposition periods have disappeared. The following changes in the physical condition of the tailings mass can be attributed to the tailings injection program:

- Large sections of the tailings profile from the injection horizon upward have been thawed, this is most apparent near the centre of the injection area.
- The surficial tailings deposited as a result of upwelling tailings flow, are largely non-frozen, despite being subjected to winter conditions.
- The temperature of non-frozen (thawed) layers is much higher than previously recorded. These warmer zones will slowly transfer heat to the adjacent frozen layers.
- The moisture content (void ratio) of thawed zones and that of tailings freshly deposited at the surface is much lower than recorded in 1997 and 1999. The lower moisture content and thus lower void ratio is most likely due to larger zones of non-frozen material. It may also be due, in part, to a lower production rate since the end of 1998.
- The density of surficial tailings deposited by upwelling of injected tailings is typically in the range of 1.0 Mg/m^3 to 1.5 Mg/m^3. This range is much higher than historical ranges of 0.5 Mg/m^3 to 1.0 Mg/m^3 for freshly deposited tailings.

II-6.3. Geochemical

The tailings injection program was not expected to have any positive or negative effect on tailings geochemistry. The data show that this was the case. The impacts of tailings injection on geochemical properties are as follows:

- The concentration of Ni and by inference As in the tailings solids is reduced over that portion of the profile subjected to injected tailings. This is a simple dilution effect.
- Arsenic porewater concentrations have remained within historical ranges for various zones within the tailings.
- Uranium concentrations in porewater appeared to be higher; however, further testing and analysis has shown this to be an artifact caused by lengthy sample storage.
- Samples of tailings collected for porewater extraction and analysis should be processed (porewater extracted) and preserved as soon as possible after collection to avoid geochemical changes due to intrusion of CO_2 gas.

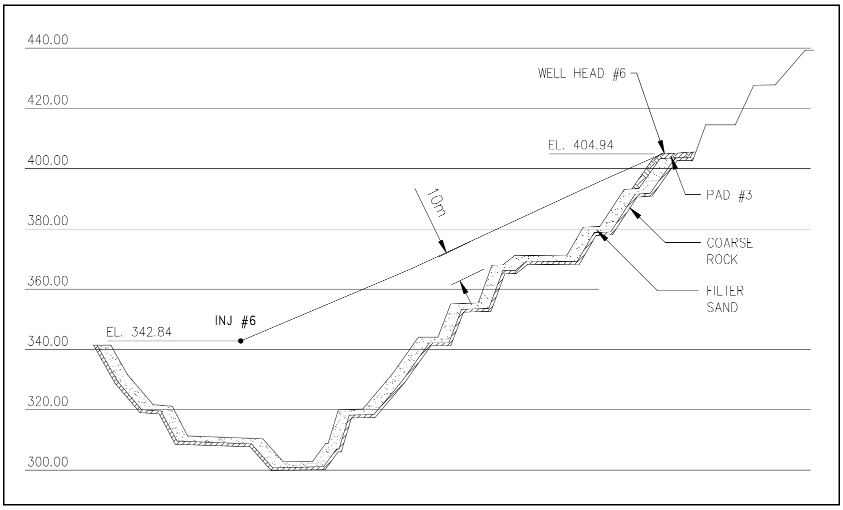

Fig. II-1. Cross-section view of a typical tailings injection well.

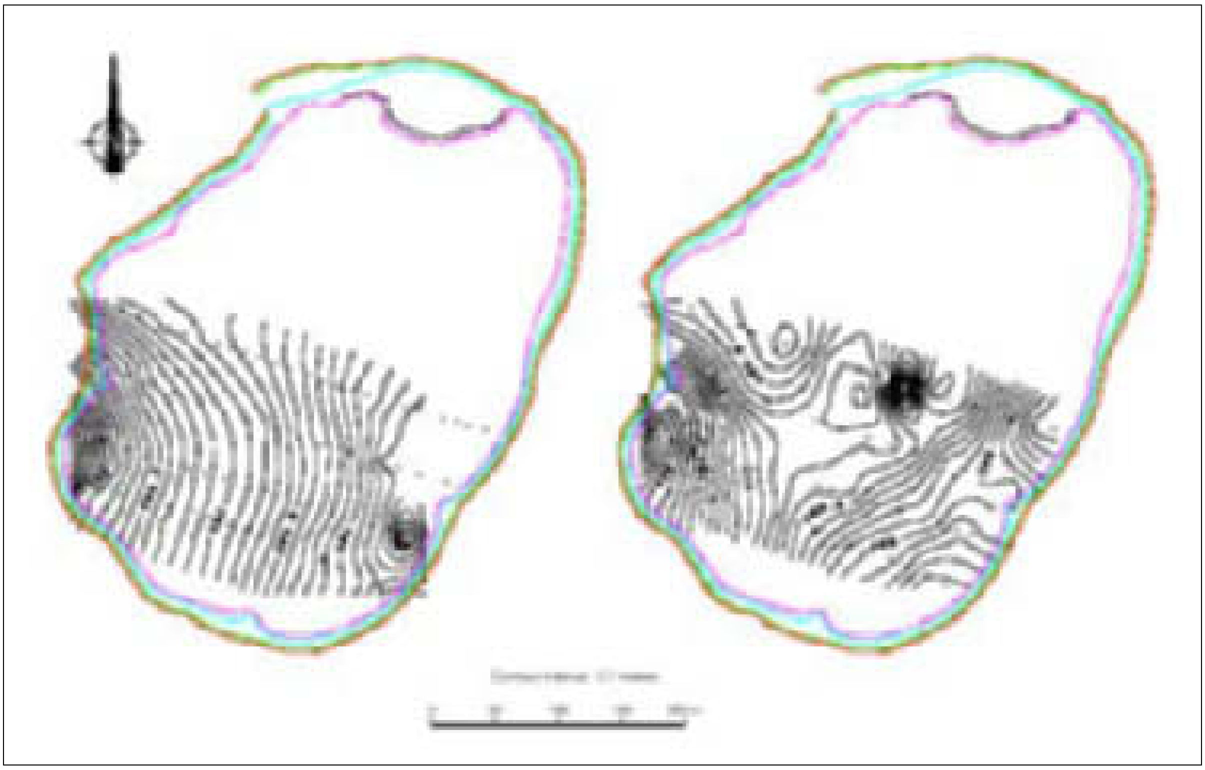

Fig. II-2. Plan views of the Rabbit Lake In-Pit Tailings Management Facility, showing tailings surface elevation contours, (left) prior to tailings injection (June 1999) and (right) after tailings injection (October, 1999).

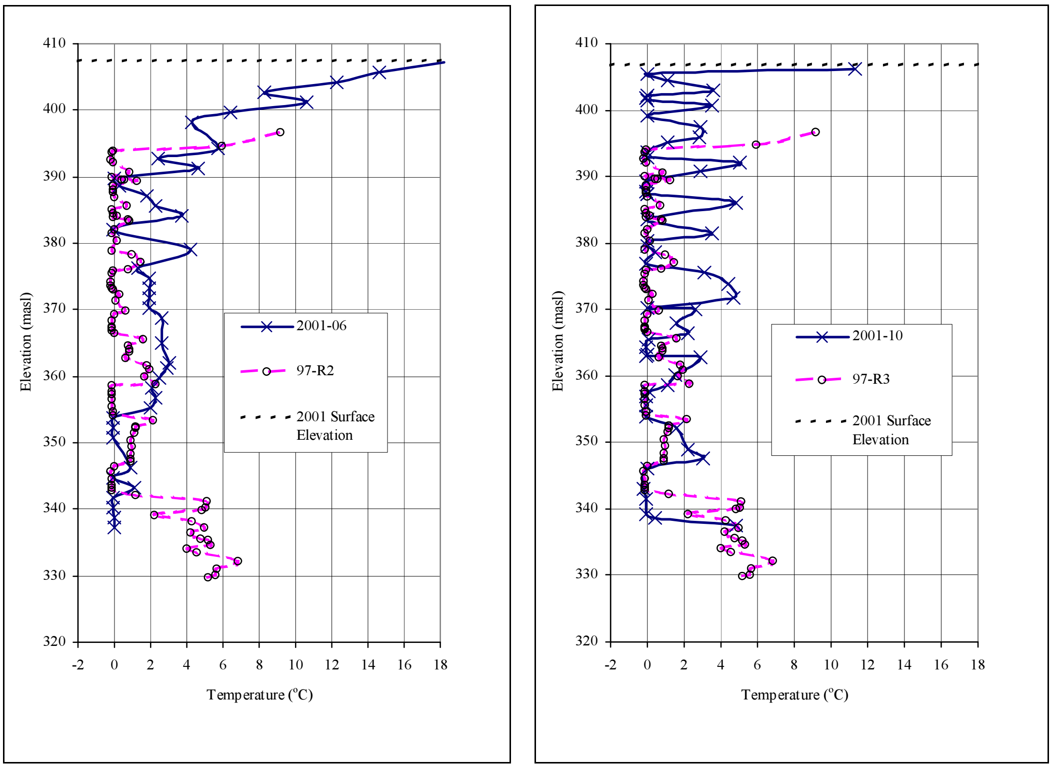

Fig. II-3. Temperature profile at BH 2001-06 and BH 97-R2 (left) and at BH 2001-10 and BH 97-R2 (right).

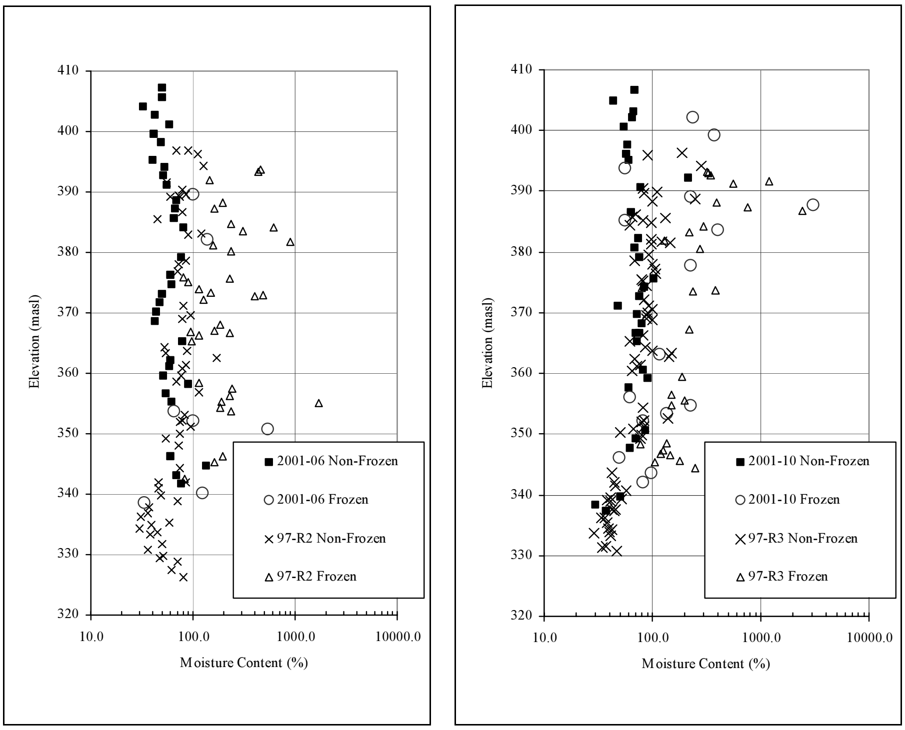

Fig. II-4. (left) Moisture content versus elevation for borehole 2001-06 and 97-R2; and (right) Moisture content versus elevation for borehole 2001-10 and 97-R3.

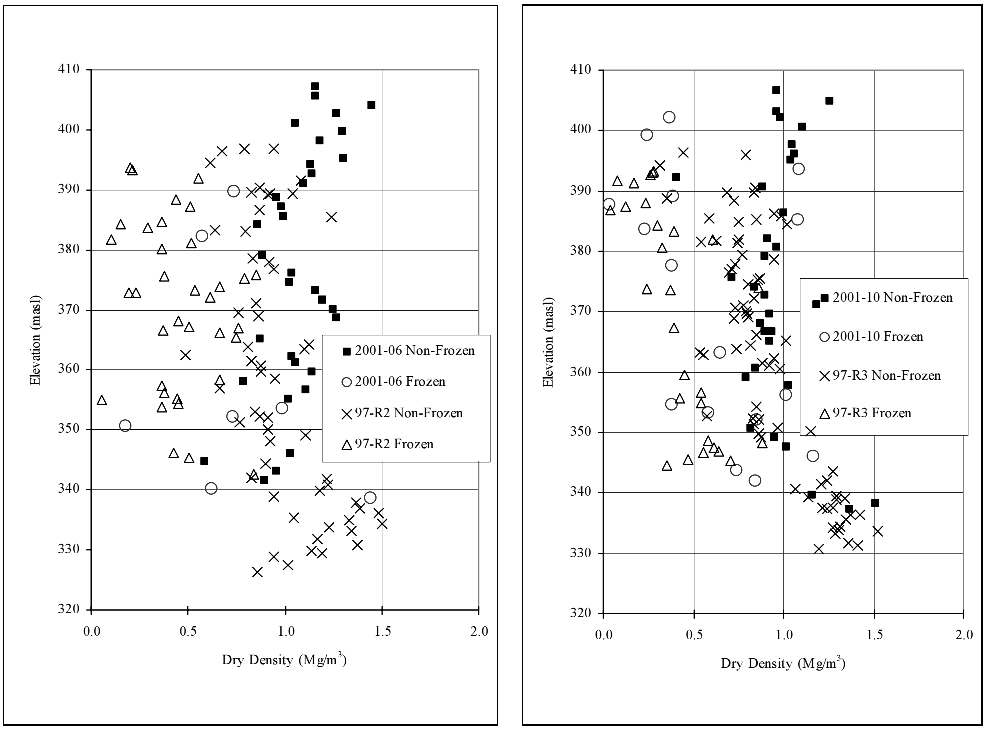

Fig. II-5. (left) Dry density versus elevation for borehole 2001-06 and 97-R2; and (right) Dry density versus elevation for borehole 2001-10 and 97-R3.

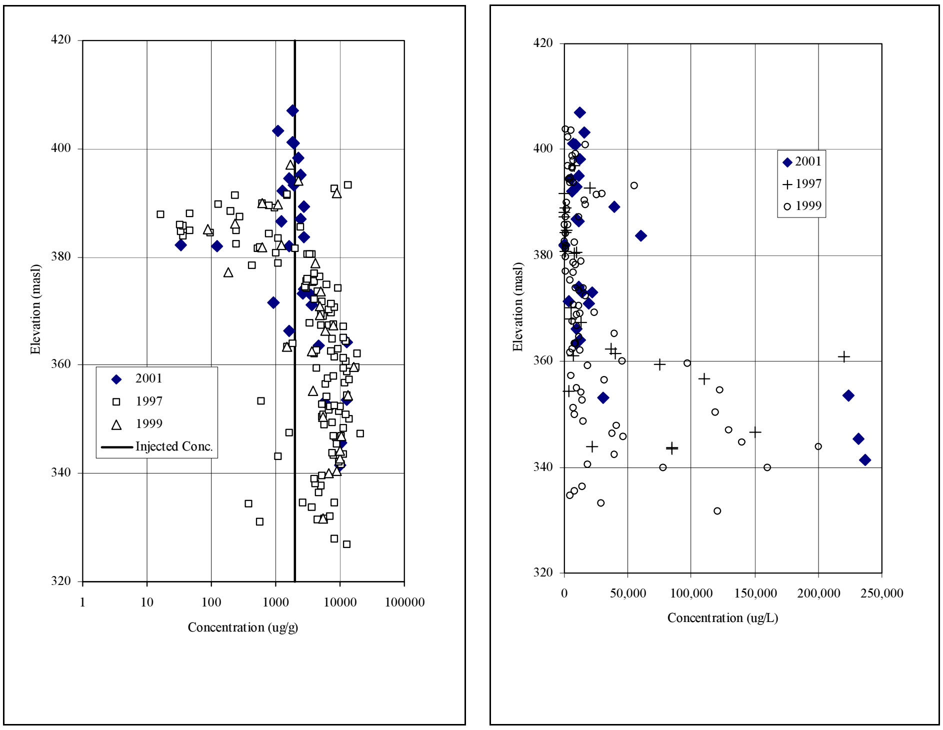

Fig. II-6. (left) Arsenic solids concentration versus elevation, and (right) arsenic porewater concentration profile, both for the 1997, 1999 and 2001 drill programmes.

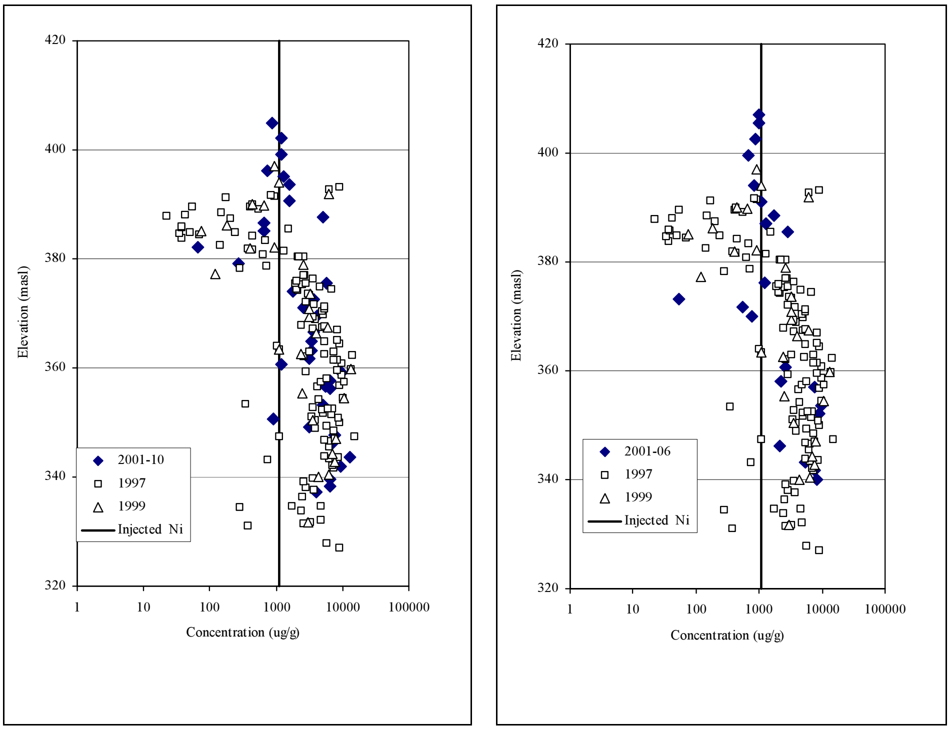

Fig. II-7. (left) Nickel solids concentration versus elevation for BH 2001-10, and (right) Nickel solids concentration versus elevation for BH 2001-06, all data from the 1997 and 1999 drill programmes.

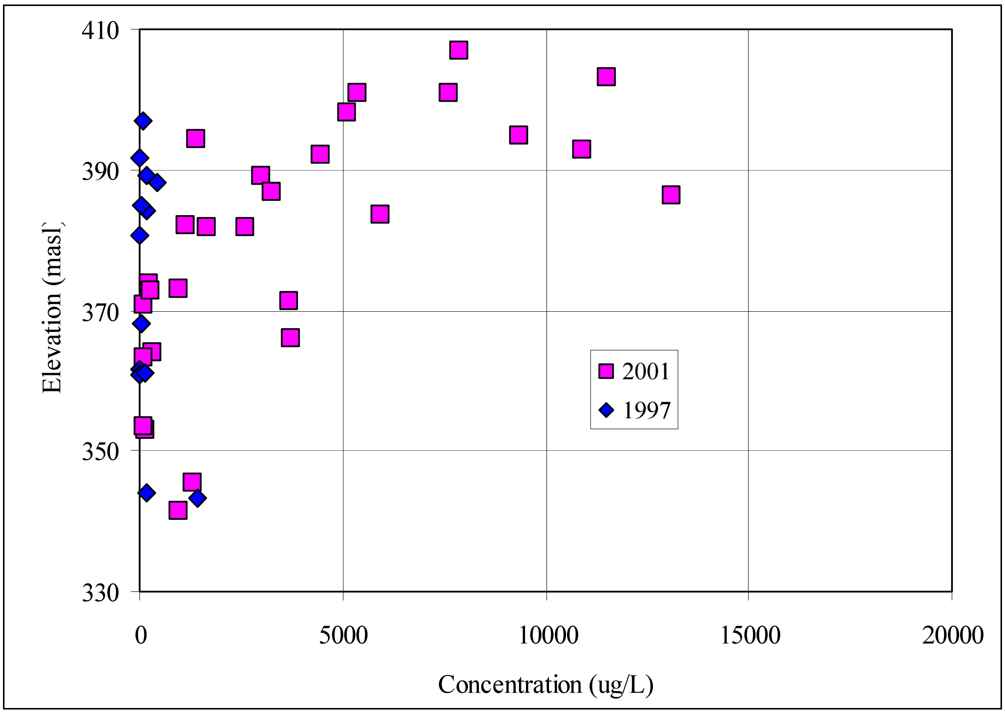

Fig. II-8. Uranium in tailings porewater for 2001 and 1997 investigation programmes.

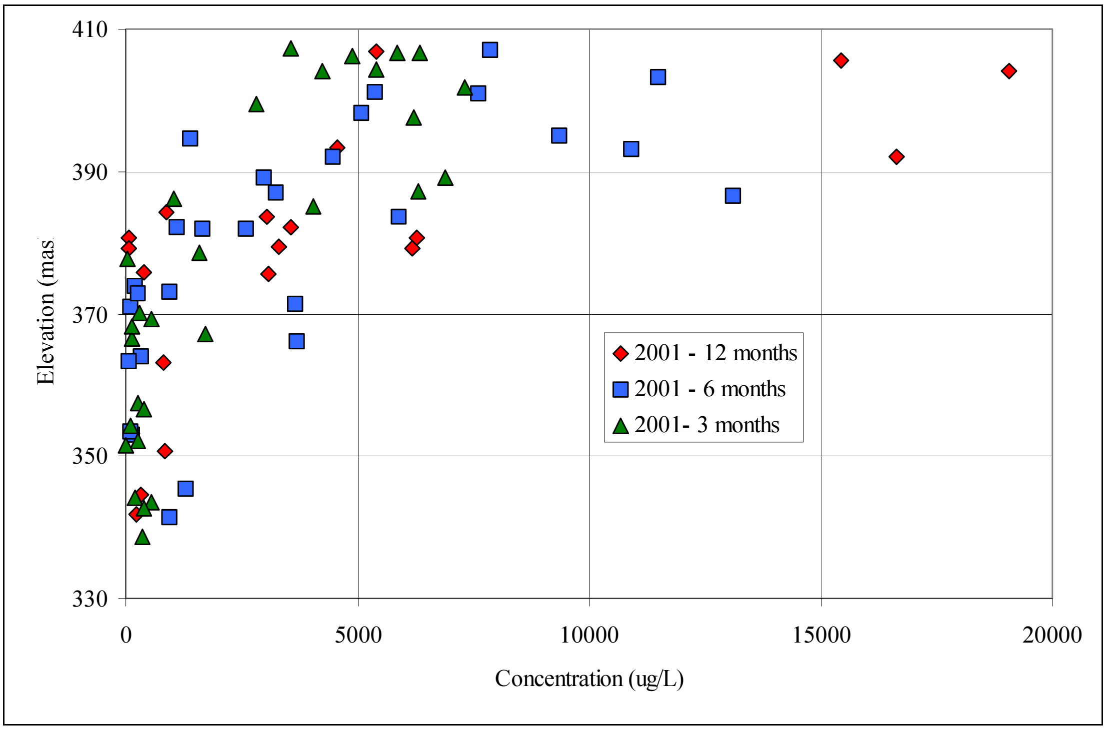

Fig. II-9. Uranium in tailings porewater. Porewater for 2001 samples extracted after 3, 6 and 12 months of storage.

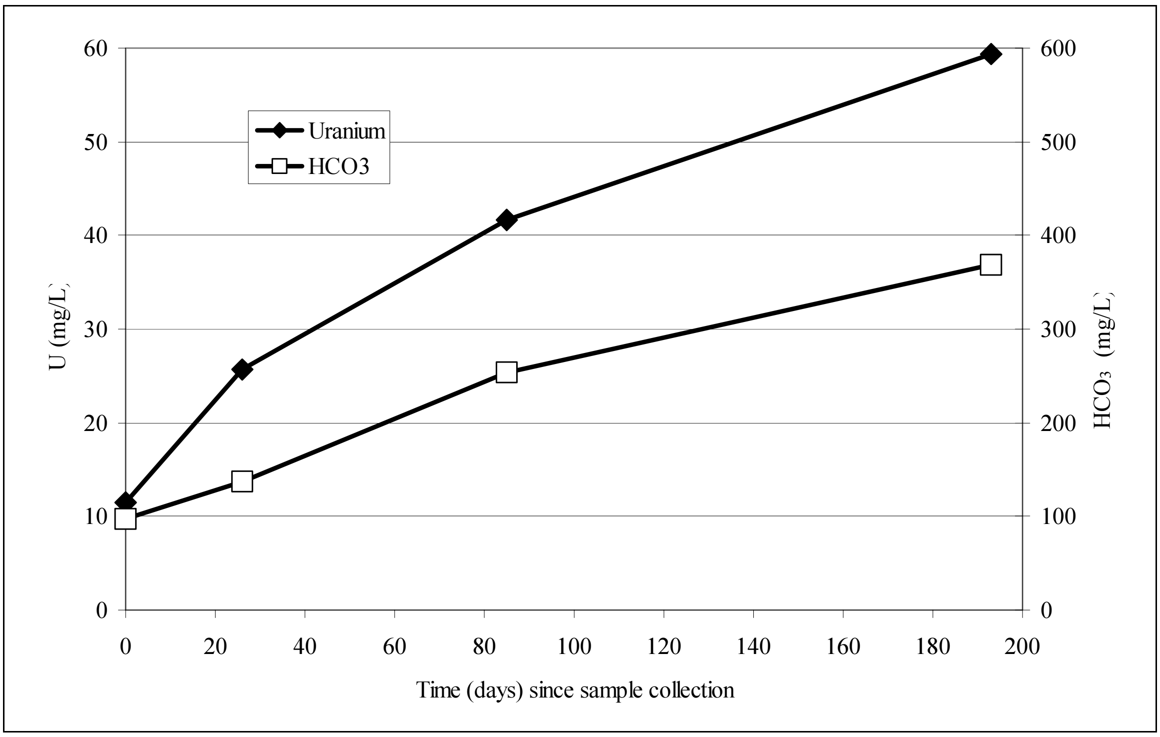

Fig. II-10. U and HCO_3 conctration versus time since sample collection for RL tailings

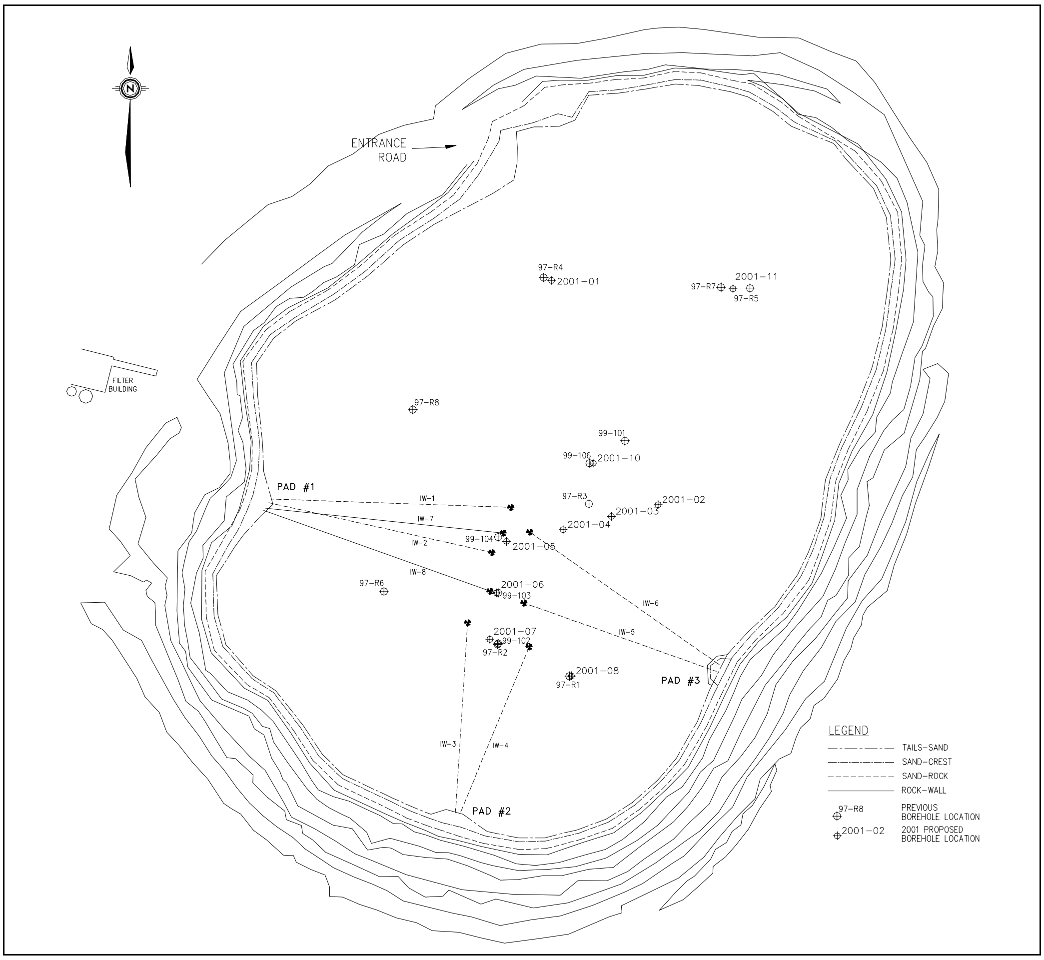

Fig. II-11. Plan view of the Rabbit Lake In-pit Tailings Management Facility showing the layout of tailings injection wells and locations of investigation boreholes (1997, 1999 and 2001).

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[II-2] MOLDOVAN, B., JIANG, D.T., HENDRY, M.J., Mineralogical characterization of arsenic in uranium mine tailings precipitated from iron-rich hydrometallurgical solutions, Environ. Sci. Technol., **37** (2003) 873–879.

[II-3] CAMECO, "1999 Rabbit Lake In-pit Tailings Management Facility Drill Program", May 2000 (2000).

[II-4] CAMECO, "2001 Rabbit Lake In-pit Tailings Management Facility Drill Program and Instrument Installation", June 2002 (2002.

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[II-6] DONAHUE, R., HENDRY, M.J., Geochemistry of Arsenic in Uranium Mill Tailings, Saskatchewan, Canada, J. Applied Geochemistry **18** 11 (2003) 1733–1750.

[II-7] CAMECO, "Follow-up to the 2001 RLITMF Investigation – U and ^{226}Ra in Tailings Porewater", May 2003 (2003).
[II-8] CAMECO, "Solution Pachuca pH Profile Adjustment Testwork", November, 1999 (1999).

ANNEX III. CHINA

STUDIES OF BENTONITE AND RED SOILS AS CAPPING OF THE URANIUM MILL TAILING IMPOUNDMENTS

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III-1. ABSTRACT

In this study, a conceptual model for the remediation of uranium mill tailing in south China is developed. Multiple-layer capping employing bentonite and local red soils at the Erqier and Shangrao uranium mill tailings impoundments were tested and the efficiency and cost of applying different capping materials were studied. These studies included determinations of the mineral composition and sedimentology of prospective capping materials, *in situ* radon and gamma surveys, measurement of infiltrating of the precipitation, as well as nuclide analyses in vegetation and local red soils.

The *in situ* gamma surveys showed that all three capping structure tested satisfy the national regulations with regard to gamma radiation. In general, capping material with high content of clay minerals can effectively prevent radon from escaping to the surrounding atmosphere due to clays' high adsorption of radon. The radon concentrations with thinner capping are higher than those with thicker capping. Vegetation on the cover surface is one of the important measures to achieve long-term stabilisation and isolation of uranium mill tailings. In South China, vegetation can play an active role to prevent erosion due to high precipitation, and to keep the remedied tailings matching to the character of the surrounding landscape. Nuclide concentrations in grasses were analysed in order to check whether the nuclides migrate from the mill tailings. Higher concentrations of U, Th and Ra were observed in grass growing on the mill tailings than in the surrounding environment. However, the nuclide concentrations are not elevated compared to the statistical nuclide concentration in soils in that province. This suggests that no significant migration of nuclides from the uranium mill tailings to the environment was observed in this study. The lower content of ^{40}K in grass growing on the capping was unexpected.

Some shallow borehole sampling under uranium impoundments at Erqier site was conducted. Analytical data of borehole samples indicate that uranium has migrated downward by about 1.5 m and that radium has migrated less than 2 m since 1963. This suggest that local soils and clay minerals that were utilised as bottom liners for the impoundments have successfully inhibited radionuclide migration.

Cost-effeciency analyses indicate that 80 cm of red soil inter-layered with 20 cm bentonite would result in efficency increase of 45.8% and a cost increase of 18%.

III-2. INTRODUCTION

There are many small-scale uranium mill facilities in China and large volumes of low activity mill tailings were often disposed off in a haphazard fashion, utilizing geomorphologic depressions or by filling-in valleys. As a result, a variety of environmental problems arose, such as radon emanation and the leaching of contaminants including radionuclides, heavy metals and arsenic, into surface and ground waters. So cost-effective geotechnical engineering measures must be employed to prevent or reduce the extent of these processes.

The objective of this project is to study the relative performance, both with respect to technological aspects and to cost efficiency, of bentonite and red soils as capping materials for uranium mill tailings. The scientific programme comprised four elements:

(1) characterizations of the chemical and mineralogical properties of bentonite and red clays;
(2) comparison of Rn emanations before and after capping and for various thickness of the capping layer using gamma-ray and Rn measurements in order to determine the optimal thickness;
(3) Species selection, vegetation and analyses of radionuclide contents of grass planted in the capping as well as that in surrounding area;
(4) Comparison of costs of applying the two different materials.

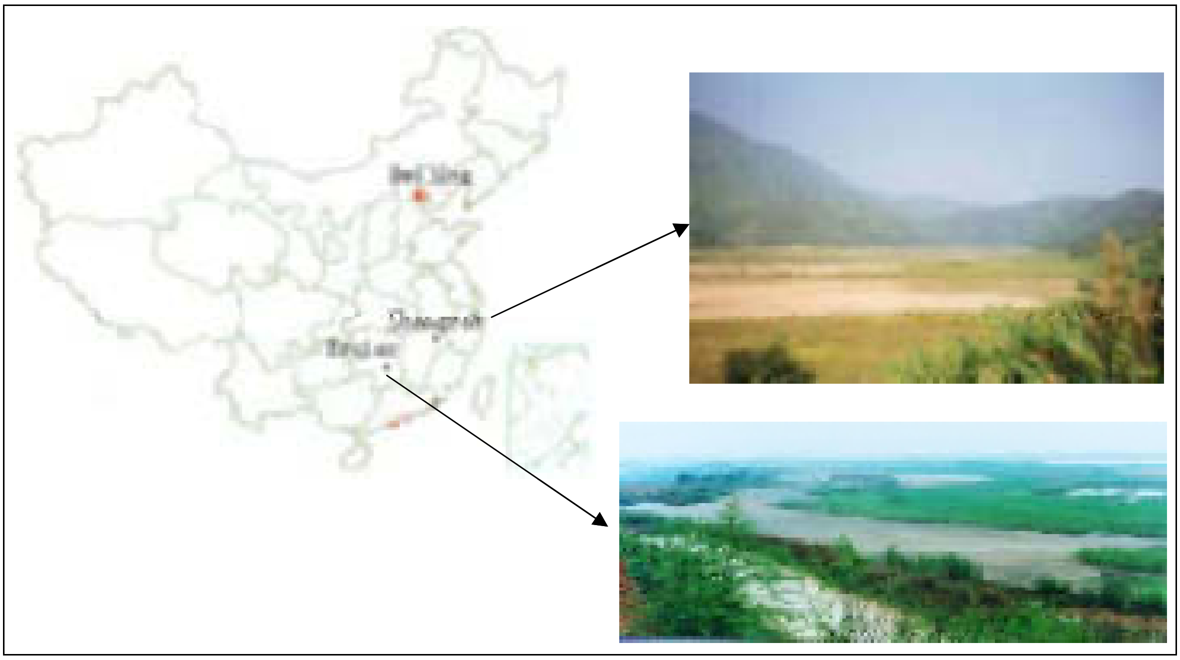

Fig. III-12. Location of two uranium mill tailing impoundments (Erqier and Shangrao).

The work object are the Erqier uranium mill tailing impoundments (Hunan Province, Southern China) and the Shangrao uranium mill tailing impoundments (Jiangxi Province, Southern China) (Fig. III-1).

The Erqier uranium mill is one of the largest of its kind in China and located 17 km south of Hengyang City. The underlying rocks are composed of lower Palaeogene amaranth feldspar-quartz sandstones, brecciae and mudstones. The local topography is characterized by hills. The uranium tailings management facility was built in a south-north-striking topographic depression. The engineered dams occupied about 1.7 km^2, with18.8 million tons of tailings slurry deposited since the start-up of facility in 1963. Tailings depths reaches up to 11 meters. The mill was decommissioned in 1994. A considerable amount of engineering and scientific effort is being focused now on the decommissioning of the tailings disposal area. The local climate is to be characterized as sub-tropical with high humidity. The average annual temperature is about 17.5~17.9ºC, and the recorded highest temperature in the 20th century was 40.8ºC. December to February is a season of low temperatures with a recorded temperature low of -10.3ºC. Annual average rainfall in the region is about 1233~1363 mm. Average annual evaporation is around 1394~1533 mm. The main direction of wind is southerly from June to August, and northeastern from September to May. The relative humidity ranges around 77~79%, and the humidity index ranges around 0.884~0.915.

The Shangrao uranium mill is one of the earliest uranium mills in China and was shut down in 1986. A systematic remediation is under way. The local topography is characterized by hills. The uranium tailings management facility was built in a topographic depression. The engineered dams occupied about 0.24 km^2. The Shangrao tailing management facility had received 2.33 million tons of tailings slurry since the start-up of tailings deposition in the beginning of 1960s. The local climate is to be characterized as sub-tropical with high humidity. The average annual temperature is about 17.8°C. Annual average rainfall is about 1973 mm. Average annual evaporation is 1421 mm. The main direction of wind is northerly with a frequency of 24.3%. The relative humidity is 76%.

III-3. WORK CARRIED OUT

(1) Development a conceptual model for the remediation of uranium mill tailing impoundments in south China;
(2) Field investigation and sampling at two uranium mill tailing impoundments and bentonite deposits;
(3) Conducting a multiple-layer capping (red soils and bentonite) operation at two uranium mill tailing impoundments;
(4) In-situ radon surveys on the surface of mill tailing before and after capping with local red soils;
(5) Chemical and mineralogical studies of capping materials (local red soils, bentonite and clayey soil);
(6) U, Th, and Ra analyses of soil samples collected in shallow boreholes;
(7) Species selection and re-vegetation on the surface of the mill tailings after being capped with a multiple-layer cover;
(8) Analyses of the radionuclide contents of grasses planted on the capping, as well as that in surrounding area;
(9) Comparison of costs of applying the two different types of material.

III-4. CAPPING OF URANIUM MILL TAILING IN SOUTH CHINA

III-4.1. Conceptual model

A conceptual model for the remediation of Southern China's uranium mill tailing has been developed. In this model, various factors have been considered, such as local climate (especially rainfall), effectiveness, cost and so on. The accurate thickness of each layer was determined according to respective study and field tests.

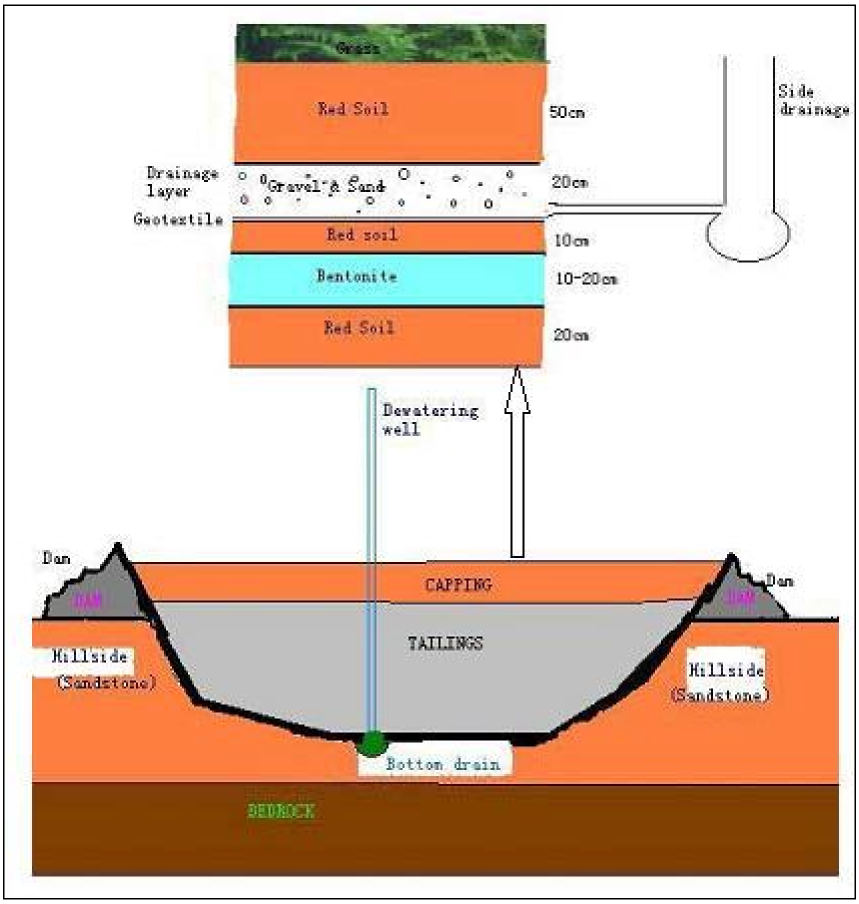

Fig. III-13. A conceptual model for remediation of uranium mill tailings in south China. (Top) Capping structure, including vegetable layer (grass), local red soil layer, drainage layer, geotextile and bentonite layer. (Bottom) Schematic diagram of uranium mill tailing impoundments.

III-4.2. Capping materials

According to the work programme of the IAEA Research Contract, a multiple-layer capping was designed and applied to reduce radon emanation from the uranium mill tailings. The multiple-layer capping is composed of local red soils and bentonite. Since the adsorption capacity of bentonite is superior to that of red soils, theoretically bentonite as capping materials would work better to decrease radon emanation rate and reduce the thickness of the capping needed.

In Shangrao mill, a preliminary capping test with 1 m clayey soil had been conducted seven years ago. Since the cost of clayey soil used as capping material is lower than that of red soil, it is useful for this project to determine the effectiveness of capping with clayey soil even through this part of work was not included in the project work plan.

Red soil widely occurs in the of South China, usually consisting of a uniform red earth, plinthictic and gravel layer. It is a sediment formed during the early Quaternary under the effect of natural factors and human activities, characterized by a dark-red weathering crust of several metres in thickness, acid reaction and illitisation [III-1].

Fig. III-14. The structure to(top right) and placement (bottom) of the multiple-layer capping, and distribution of measurement points (top left) at the Erquier uranium mill tailing impoundment.

Table III-1. Mineral compositions (wt%) for capping material.

Sample	Quartz	Feldspar	Hematite	Anatase	Clay minerals						
					Montmorillonite	**I/S**	**Illite**	**Kaolinite**	**Chlorite**	**M/V**	**K/S**
Shangrao site											
Red soils	47.8	1.3	2.2			18.5	18.5	11.7			
Clayey soils	53.9	1.1	1.5				18.7	13.5	3.0	8.3	
Bentonite	18.9	2.0		3.5	49.9			10.6			15.1
Erqier site											
Red soils	42.8	0.9	2.0	1.1			13.8	22.7	5.6	11.1	
Bentonite	25.4			1.1	73.5						

Note: I/S Illite/montmorillonite mixed-layer mineral
M/V Mica and vermiculite mixed-layer mineral
K/S Kaolinite and smectite mixed-layer mineral

The clayey soil is a yellow alluvial soil with somewhat more gravels than red soils and was formed during the Holocene. The clayey soil is poor and less cultivated in South China.

The mineralogical composition of the materials (local red soils, bentonite, clayey soils) used in field tests at the two uranium mill tailing impoundments, has been determined by XRD (X-ray diffraction) analyses. The instrument used is a D/max-2500 with CuK$_{\alpha}$ radiation (40 kV and 100 mA). The data are listed in Table III-1.

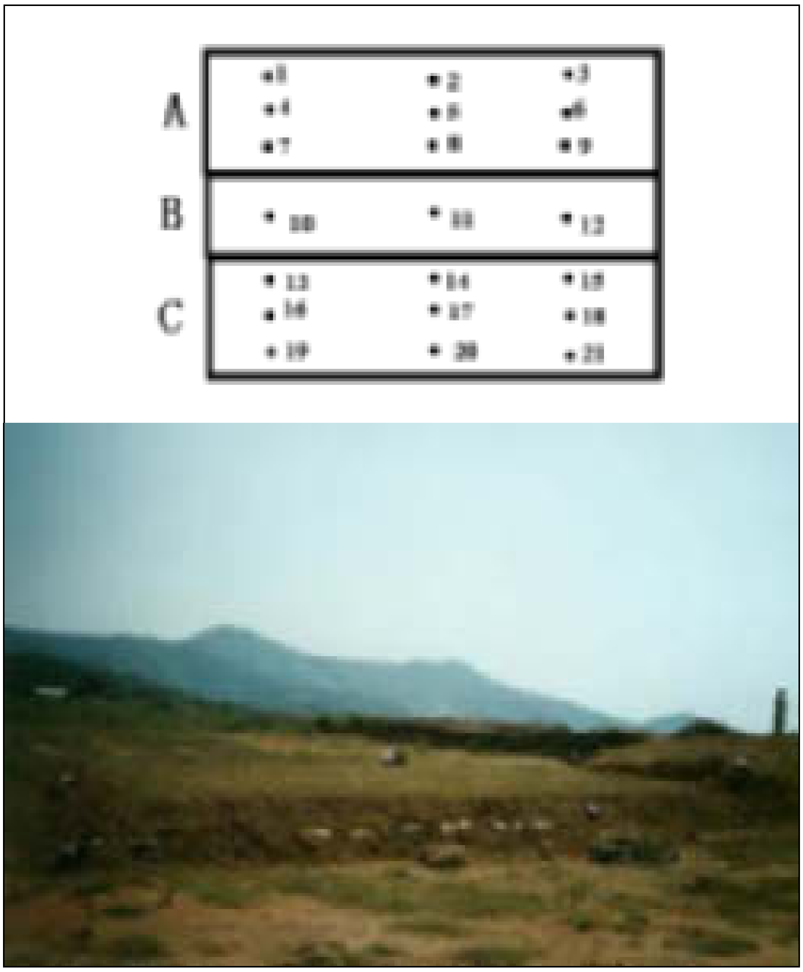

Fig. III-15. The distribution of measurement points (top) and placement (bottom) of the multiple-layer capping at the Shangrao uranium mill tailing impoundment.

III-4.3. Capping structure

In order to compare the effectiveness of different capping materials, a field-scale multiple capping was designed. The structure of capping and landscape of multiple capping is shown in Figs. III-3 and III-4.

III-4.4. Radon surveys on the capped tailings

In situ radon surveys and measurements of gamma radiation dose rates were conducted after the experimental capping placement. The instruments for field tests are shown in Fig. III-5.

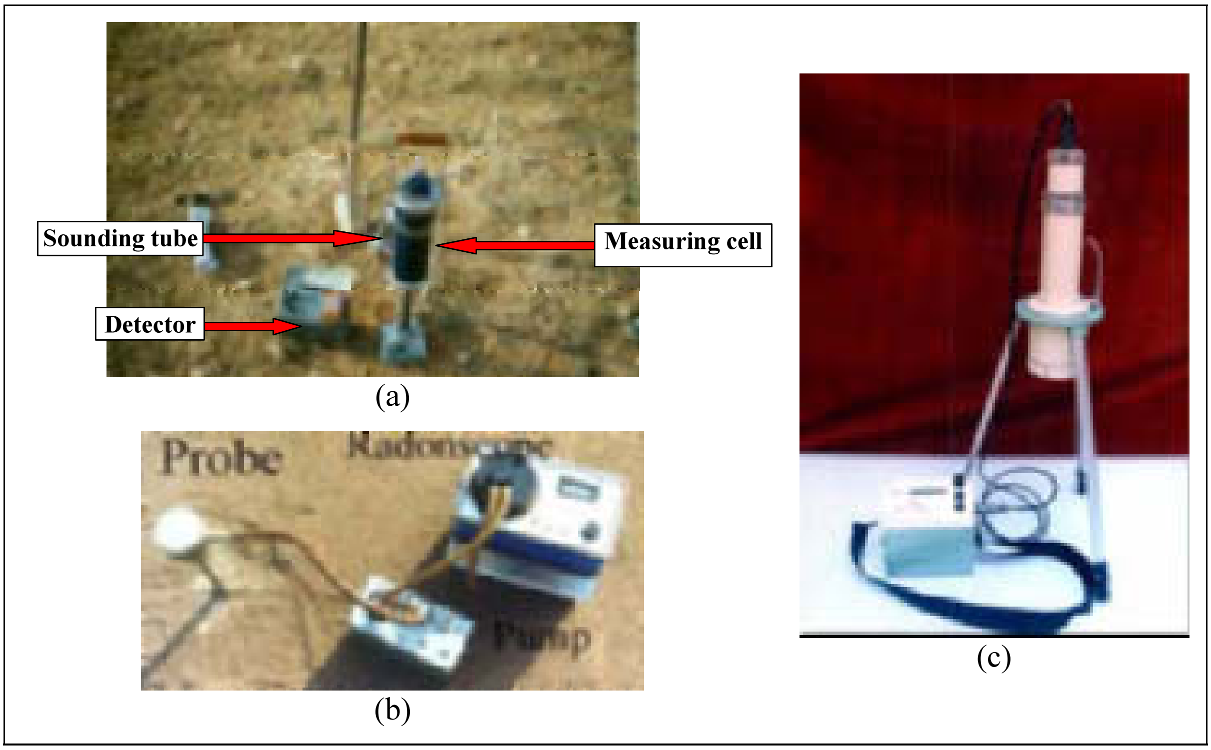

Fig. III-16. Instruments for field test: a,c-Instruments for the Shangrao Site; b-Instrument for the Erqier Site

Device 'a' consists of three parts: 1) a sounding tube, 2) a measuring cell, and 3) a FD-3017 type detector. The system is a portable, battery powered instrument for determining the radon content in the soil. In the initial measuring phase, air from the soil is pumped up through a sounding tube into a measuring cell. After the pumping phase the measuring phase begins. The detector is activated and the voltage to the measuring chamber is switched on. The charged radon daughters, formed by the decaying radon gas, are driven towards the detector by an electric field in the chamber. The detector registers the alpha radiation originating from the radon daughters. The pulses are counted and the result is shown on the instrument's display, the count plus a constant (determined experimentally, 262.5 Bq/m^3) equal the radon concentration in soil.

Instrument 'b' consists also of three parts: 1) a probe; 2) a DTB-7 type degassing pump; and 3) a FD-3016 type radonscope. The monitors were placed in the areas concerned for approximately 2 min. at a time and recorded the readings. Radon concentration in the uranium mill tailings and capping materials is measured using a scintillation cell. It is a 150 ml Al cylinder with inner walls coated with ZnS as detector. The air sample is drawn into an evacuated scintillation cell by pumping for 2 minutes, left standing for 2 minutes, and then counted for α-activity.

The gamma penetrating radiation dose rate on the surface in different capping structures have also been measured *in situ*. The instrument is a BH1303A portable X-ray/-γ-radiation detector (Fig. III-5).

III-4.5. Selection of re-vegetation species

The plant species selection is conducted based on the following considerations:

— Emphasis is given to indigenous, long-lived grasses;
— The grasses require little or no maintenance over the long term;
— The root of the grass developed vigorously thus preventing the capping from erosion.

Hence, *Digitaria sanguinalis* (L.) Scop was selected as candidate grass for the vegetation cover at the Shangrao site. *Digitaria sanguinalis* (L.) Scop is a low-growing spreading, hairy and pale green annual, bent and often rooting in lumps; frequently occurring along roadsides, in over-grazed pastures, lawns and gardens. Leaves are rolled in the bud-shoot. The sheath are compressed, pilose, green, but sometimes purplish-veined, split with hyaline margins. Auricles are absent. Collars are broad, distinct, sparsely hairy, divided by a middle rib. It is ligule membranous, 0.5 to 2.0 mm. long, acute, undulate or toothed, often reddish. Blades are 4 to 10 mm. wide, 5 to 15 cm. long, flat, soft, drooping, often puckered, sharp-pointed, green, not ridged, pilose on both surfaces with a few longer hairs at base on upper surface; mid-rib prominent on lower surface; margins scabrous.


Fig. III-17. Digitaria sanguinalis (L.) on natural plot (left) and on the capping (right).

III-5. RESULTS ACHIEVED

III-5.1. Gamma penetrating radiation dose rate

Gamma radiation dose rate is one of the important parameters to evaluate the effect of remediation of uranium mill tailings. After remediation, the gamma radiation dose rates should not be higher than $17.6 \cdot 10^{-8}$ Gy/h above the local natural background (Chinese standard: EJ993-96). A hand-held radiometer (Fig. III-5) was used to measure gamma radiation. Locations of measuring points are shown in Fig. III-4e (Point 1-21). The results for the Shangrao site are shown in Fig. III-7.

At the Shangrao site, the natural gamma radiation dose is $20 \cdot 10^{-8}$ Gy/h, thus the limit would be $37.6 \cdot 10^{-8}$ Gy/h. Before capping, the gamma radiation dose rate on the mill tailings was higher than $100 \cdot 10^{-8}$ Gy/h. The dose rates on the surface after capping for three blocks are lower than $31 \cdot 10^{-8}$ Gy/h (Table III-2). Therefore, the *in situ* gamma survey shows that all of three capping structure satisfy the national regulation with regard to gamma radiation.

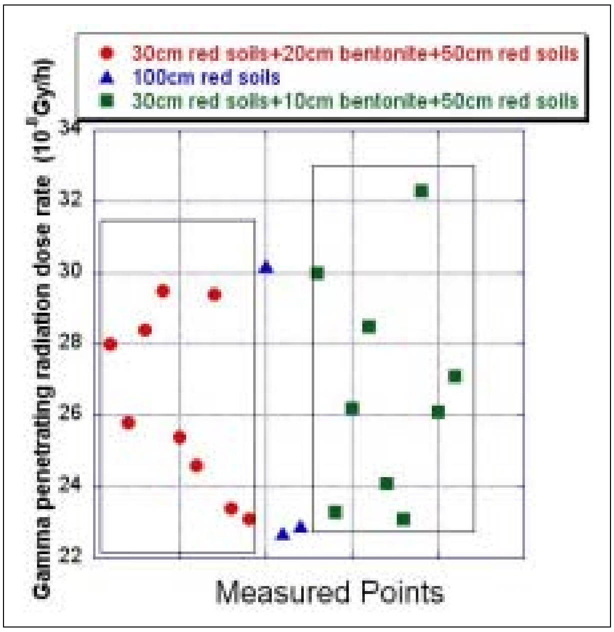

Fig. III-18. Gamma measurement at the Shangrao site.

III-5.2. Red soils capping

At the Erqier site, a red soil capping was constructed in 1996. The effectiveness of the capping was investigated by an *in situ* Rn survey and radionuclide analyses of soils sampled from the capping. The profile of the capping is shown in Fig. III-8. Uranium is analysed by laser exited fluorescence, Th is analysed by colorimetric analysis with a spectrophotometer, and radium is analysed by scintillation analysis (Radon daughter scintillation detector with active carbon adsorption). The results are shown in Fig. III-9.

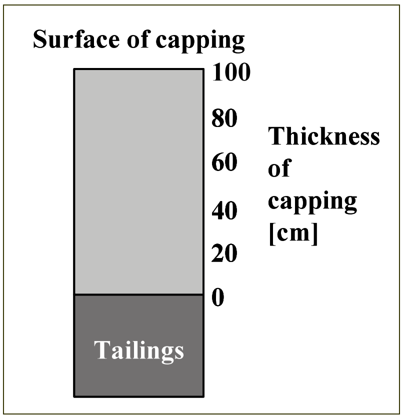

Fig. III-19. A sketch profile for radon measurement thickness.

In general, Rn content in the capping decreases to the capping thickness increases. The radionuclide content (U, Th, Ra) of red soil did not show consistent variation with depth. Generally the radionuclide concentrations decrease as the capping thickness increases, due to capillarity even through there were some exception. On the other hand, compared to multiple-layer capping field tests (see section III-4.3), the Rn concentration in red soils is about 10 times than that in the multiple-layer capping. Considering that Fig. III-9 shows the results from a capping constructed several years ago, there might be an accumulation of Rn in the capping, or in the other words, two months is not a sufficient period for radon equilibration after capping. The field test did not reveal significant differences between the compacted part and the uncompacted part. Theoretically, compaction should play an active role in preventing radon from emanation, however, due to the local high precipitation, uncompacted parts would keep more precipitation, which in turn prevents radon from escapping.

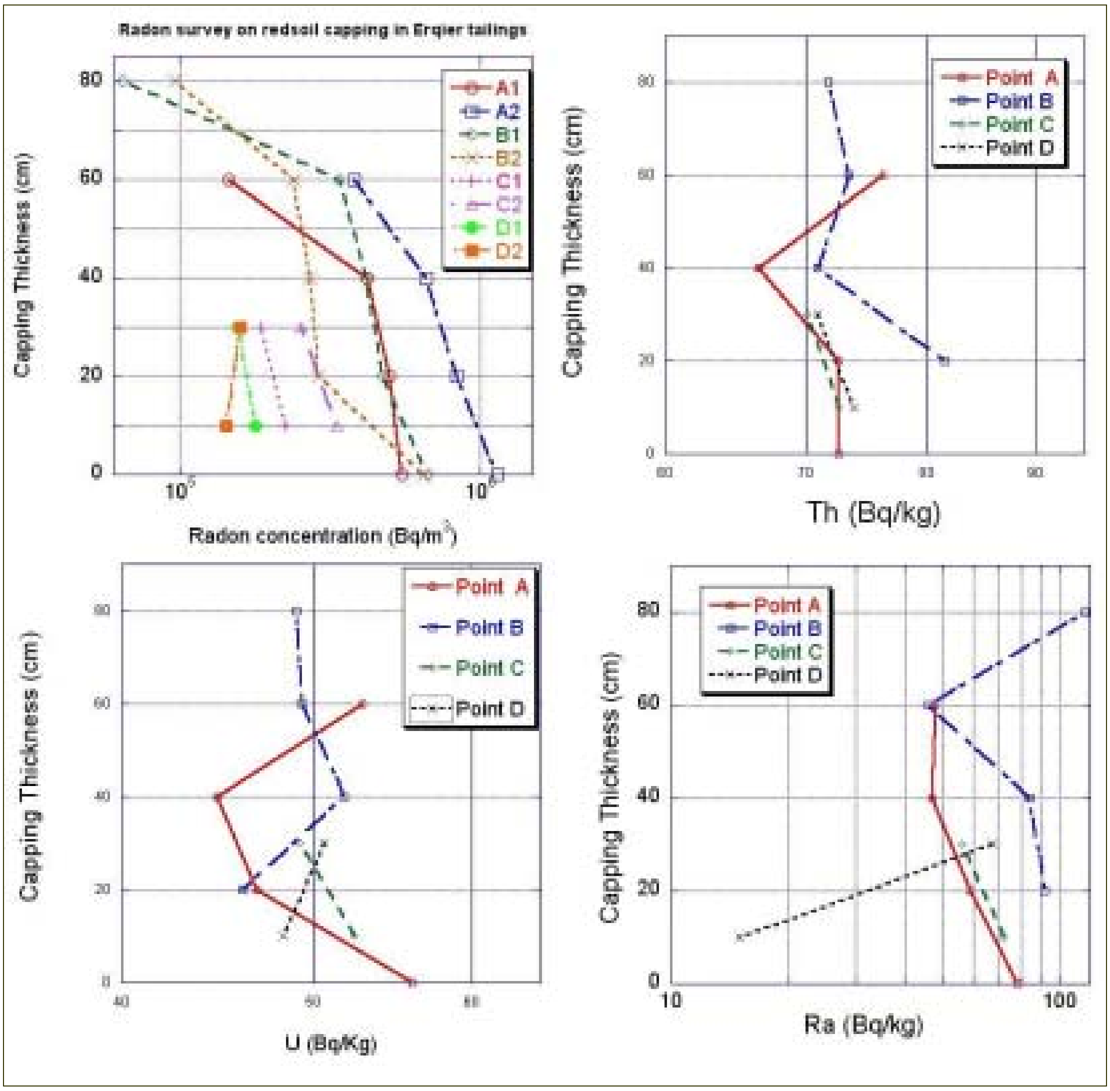

Fig. III-20. Effectiveness of capping with red soils at the Erqier site.

Note: Area A, C, E are located in the compacted zone, the thickness of capping soils are 80 cm, 50 cm, and 30 cm respectively;
Area B, D, F are located in the uncompacted zone, the thickness of capping soils are 100 cm, 50 cm, and 30 cm respectively.

III-5.3. Multiple-layer capping

The radon concentrations for thinner cappings are higher than that for thicker cappings at the two uranium mill tailings impoundments. The radon content with 80 cm capping is about one tenth, one third and one fifth of that with 40 cm capping in Blocks A, B, and C respectively. This to some extent might prove the higher effectiveness of multiple cappings with bentonite interlayer compared to ones without bentonite sub-layer (Fig. III-10).

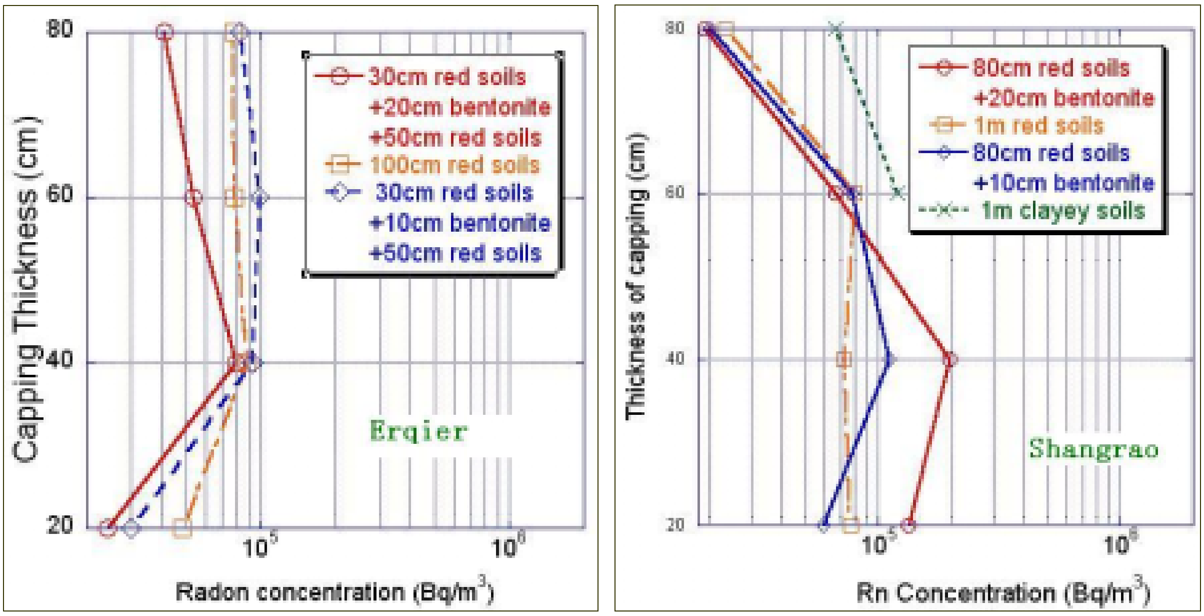

Fig. III-21. Radon concentration as a function of thickness of capping.

The water contents in the cover increases from the surface to the bottom of the capping (especially more than 50 cm from the surface of capping), since the cover can retain the water for a long period as a result of high precipitation at the two sites. This would explain the phenomenon that the measured radon content with 20 cm capping is lower than that with 40 cm capping. Radon can escape from the capping typically two mechanisms: 1) due to recoil, when the radon atom receives amomentum that enables it to travel a certain distance through a material; the recoil range is about 65 mm in air and 35 nm in clay [III-2]. 2) radon atoms not escaping the pore space by recoil may still be able to leave it by diffusion. If the soil water content increases in capping, the water films adhering to the grains effectively stop recoiling radon atoms in the pore water [III-3], which result in less radon collected by the sounding tube even though higher content of radon exists in the deep of cover.

In general, capping material with high content of clay minerals can effectively prevent radon from escaping to the ambient atmosphere due to clay's higher adsorption capacity for radon. The clay contents in the bentonite, red soils and clayey soils are 75.6%, 48.7%, and 43.5% respectively. Red soils have a little higher content of montmorillonite than the clayey soil. The montmorillonite has higher adsorption capacity than the other clay minerals and thus contribute to much better performance in preventing radon emanation. Bentonite is a naturally occurring clay consisting primarily of the mineral montmorillonite, whose unique crystalline structure is responsible for the clay's properties. The clay structure, when dry, resembles a negatively charged stack of plates. When the plates come in contact with a polar liquid, such as water, the plates absorb the liquid forcing them apart and this is the mechanism by which bentonite seals soil and retard radon emanation.

III-5.4. Migration of radionuclides from the impoundment

Shallow borehole sampling underneath the Erqier uranium impoundment has been conducted in order to study the infiltration. Uranium, thorium, and radium contents in the solids have been analysed. The results are shown in Fig. III-10. The average concentrations of natural radionuclides ^{238}U, ^{232}Th, and ^{226}Ra (area weighted) in soils in the Hunan province are 47.8, 54.2, and 59.4 Bq/kg respectively, with a range of (14.7-431.5), (9.7-437.8), and (20.3-296.8) Bq/kg respectively. Analytical data of borehole samples indicate that uranium has migrated downward for about 1.5 m and radium has migrated less than 2 m since 1963. This suggest that local soils and clays that were used in the bottom liners for the impoundment have successfully inhibited the radionuclide migration.

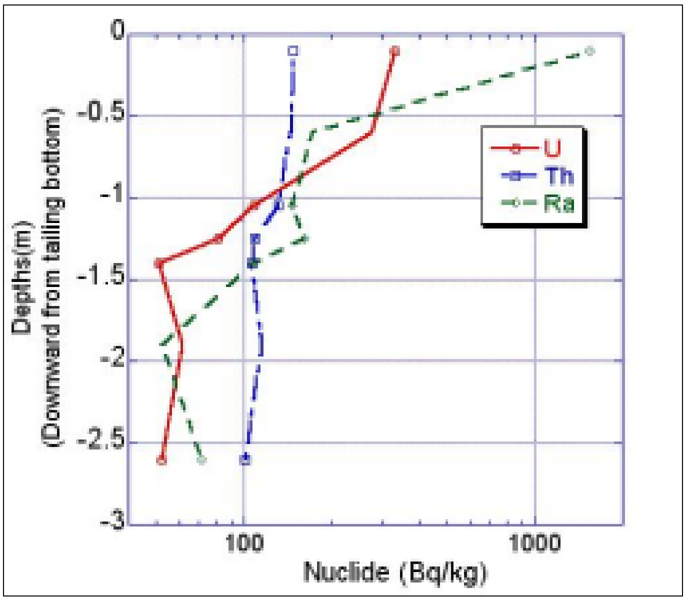

Fig. III-22. Radionuclide content in the solids of infiltration beneath the Erqier uranium mill tailing impoundment.

III-5.5. Vegetation cover and radionuclides analyses in grasses

Samples from grasses planted were collected twice (2 and 4 months after re-vegetation). The radionuclides (including U, Th, Ra, K) were analysed in the dry mass to evaluate radionuclide migration from the uranium mill tailings to the vegetation. The same grass species in the surrounding local environment were also collected and analysed as a reference. Uranium was analysed by laser-exited fluorescence, Th by colorimetry with spectrophometer, and radium is analysed by scintillation analysis (radon daughter scintillation detector with active carbon adsorption). The analytical data are given in Figs. III-12 and III-13.

The vegetation cover on the capping surface is one of the important measures to assure the long-term stabilisation and isolation of uranium mill tailings. At the Shangrao Site, the revegetated cover will play an active role to prevent the capping from being eroded due to high precipitation and to merge remediated tailings into the character of the surrounding landscape.

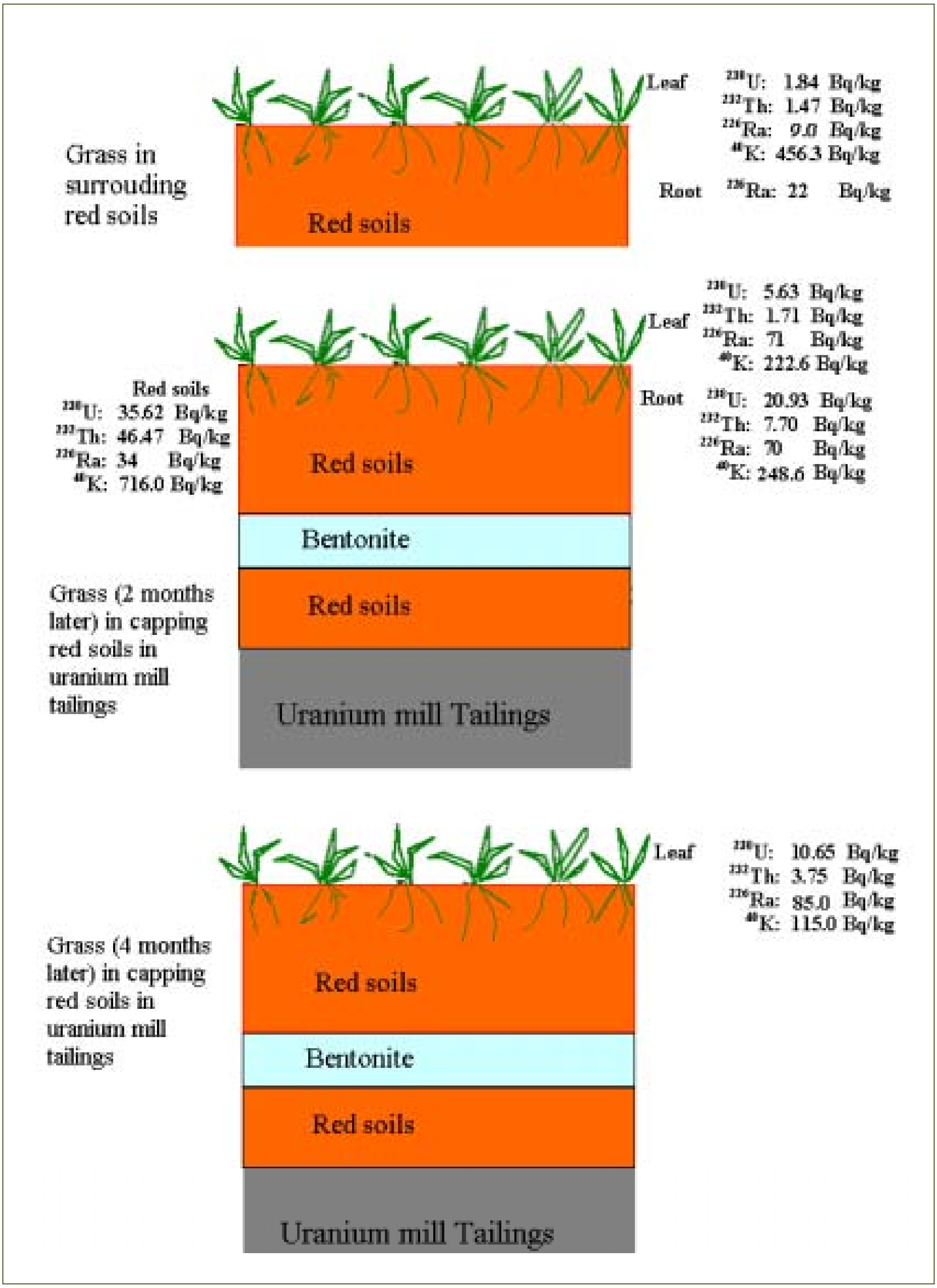

Fig. III-23. Radionuclide contents in grass and red soil.

The comparison of radionuclides contents in red soil capping and grass is seen in Fig. III-13(left). The radionuclides contents of ^{238}U, ^{232}Th, ^{40}K are lower than that in red soils; the ^{226}Ra content is about twice as that in red soils.

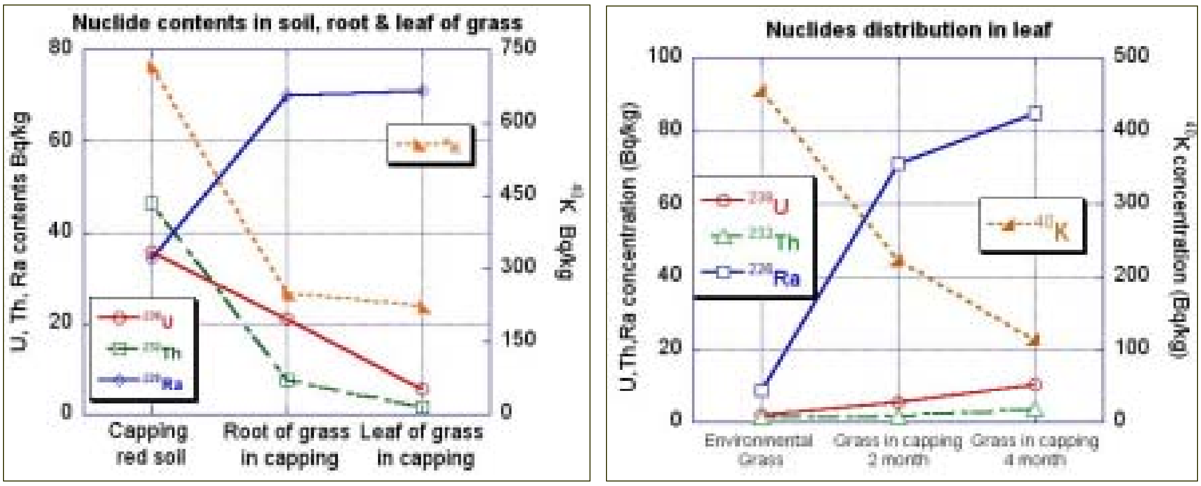

Fig. III-24. Radionuclide content in grass and comparison with surrounding grass.

The radionuclides in the grass were analysed in order to check whether radionuclides migrate from the mill tailings. The ^{238}U content in leaves is about about 2 times (2 months later) and 5 times (4 months later) higher compared to that in the surrounding environment. The ^{232}Th content in leaves is about 15% (2 months later) and 1.5 times (4 months later) higher compared to that in the surrounding environment. The ^{226}Ra content in leaves is about 7 times (2 months later) and 8 times (4 months later) higher compared to that in the environment; while the ^{40}K content in leaves is about one half (2 months later) and a third (4 months later) lower compared to that in the surrounding environment (Fig. III-13(right)).

According to field observation, the roots of grasses actually penetrated the capping only down to about 20 cm. The radionuclide analysis shows that ^{238}U and ^{232}Th concentrations in grass are higher than that in the surrounding soil, which is to some extent related to the mill tailings underneath. A possible transport mechanism might be capillarity. A similar phenomenon was reported by American researchers [III-4].

Even there are higher U, Th, and Ra concentration than that in the surrounding soil in both, the capping soil, and grass sowed, radionuclide concentrations are not higher than the statistical radionuclide concentration in soils of that province. The concentrations of the natural radionuclides ^{238}U, ^{232}Th, and ^{226}Ra fall in range of 14.7-431.5, 9.7-437.8, and 20.3-296.8 Bq/kg respectively, which suggests that no significant migration of radionuclides from the uranium mill tailings to the environment was detected in this study.

The lower content of ^{40}K in grass growing on the capping was unexpected. The further investigation will be conducted to explain this phenomenon.

III-5.6. Cost-effectiveness analyses

With regard to the Erqier uranium mill tailing impoundment, the result of cost-efficiency analyses is shown in Table III-2. The cost of different capping structures was estimated by referring to related regulations and prices of the different materials. The cost of the mutiple-layer capping at Shangrao site is similar to that in Erqier. The efficiency is calculated by comparison of Rn emanation rate with different capping materials.

Table III-2. Cost-effectiveness analysis for multiple-layer capping

Structure	Proctor density	Water content	Rn emanation rate [Bq/cm^2·s]	Efficiency	Cost
80 cm red soils inter-layered with 20 cm bentonite	1.51	17.0	0.39	+45.8%	+18%
100 cm red soils	1.54	16.0	0.72		
80 cm red soils inter-layered with 10 cm benotonite	1.58	16.8	0.56	+22.2%	+9%

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ANNEX IV. CZECH REPUBLIC

PREDICTING THE LONG TERM STABILISATION OF URANIUM MILL TAILINGS

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Diamo, S.P., Stráž Pod Ralskem

IV-1. ABSTRACT

The long-term stabilization and remediation of uranium mill tailings ponds is an important task for DIAMO. After uranium mining was stopped, DIAMO has to remediate more then 400 ha of tailings ponds at three locations. It is currently planned to cover the surface with low permeability materials with a slope of approx. 3% to protect the interior of the disposal facility from infiltrating rainwater. This entails to cover the free surface of these ponds with several hundred thousand tons of inert material. As a result of this load, the porewater from the tailings is expelled and the body of the impounded materials consolidates. Consolidation of tailings proceeds irregularly, depending on the internal structure of the tailings layers and on the progress of loading. The surface needs to be recontoured for a long time into the future.

The topic of the DIAMO project is to predict and optimise the final surface contour of the tailings pond body, and to determine the time schedule and locations for recontouring work. The K1 tailings pond in Dolní Rožínka (Southern Moravia) is a typical example for such task. The average thickness of the tailings layer is around 25 m and the average porewater contents varies from 25 up to 40%. In the years 1998-99 a PHARE pilot project was undertaken that aimed to predict the quantity and quality of drainage and infiltration waters as a function of time.

A new investigation programme (field, laboratory and modelling) has been implemented. The range of material properties and distribution of types of tailings was established. Orientation calculations of the tailings consolidation were made for fine slime zone. The results have shown that significant subsidence of the surface is to be expected after loading with inert material for the construction of an interim cover.

IV-2. INTRODUCTION

Uranium ore was mined from the Rožná deposit since 1957. A uranium mill is in operation since 1968 employing a carbonate extraction technology. Two tailings ponds were built as a repository for mill tailings. Pond K1, the bigger one, is still in operation, while pond K2 is recently used as a repository for remediation residues. Both ponds are planned to be restored after uranium production has ceased. A conceptual plan for the restoration of the ponds has been elaborated [IV-1] as well as the technical plan for their decommissioning [IV-2]. A risk analysis and environmental impact assessment (EIA) [IV-3] procedure are curently in the stage of public discussion with the aim to get the final approval from the government administration no later then at the time of closing of processing plant.

Two different variants for the restoration of the ponds has been proposed. A project concerning prediction of water balances and water chemistry of the ponds was completed as part of the PHARE programme ”Remediation Concepts for Uranium Mining Operations in CEEC – Phase B” [4]. The results obtained within this project were used as the basic criteria for evaluation of restoration variants and for the technical design of various remediation options.

The CRP project is focused on the prediction and optimizing of final contours of the tailings pond body and the time schedule and location of remediation work. Validation of the water balances and predictions of time over which treatment of drainage waters will be required are integral parts of this project. Field investigations, laboratory testing of undisturbed tailings samples, long term geodaetic monitoring and mathematical modeling form the elements of this project.

IV-3. GENERAL DESCRIPTION OF ROŽNA DEPOSIT

IV-3.1. The Rožná Site

The Rožná deposit is located in Southern Moravia, in the Czech-Moravian Highlands, some 55 km North-West of Brno (Fig. IV-1). Uranium mining from the Rožná deposit began in 1957. Uranium ore processing at the Rožná site the commenced in 1968 using a carbonate leaching method.

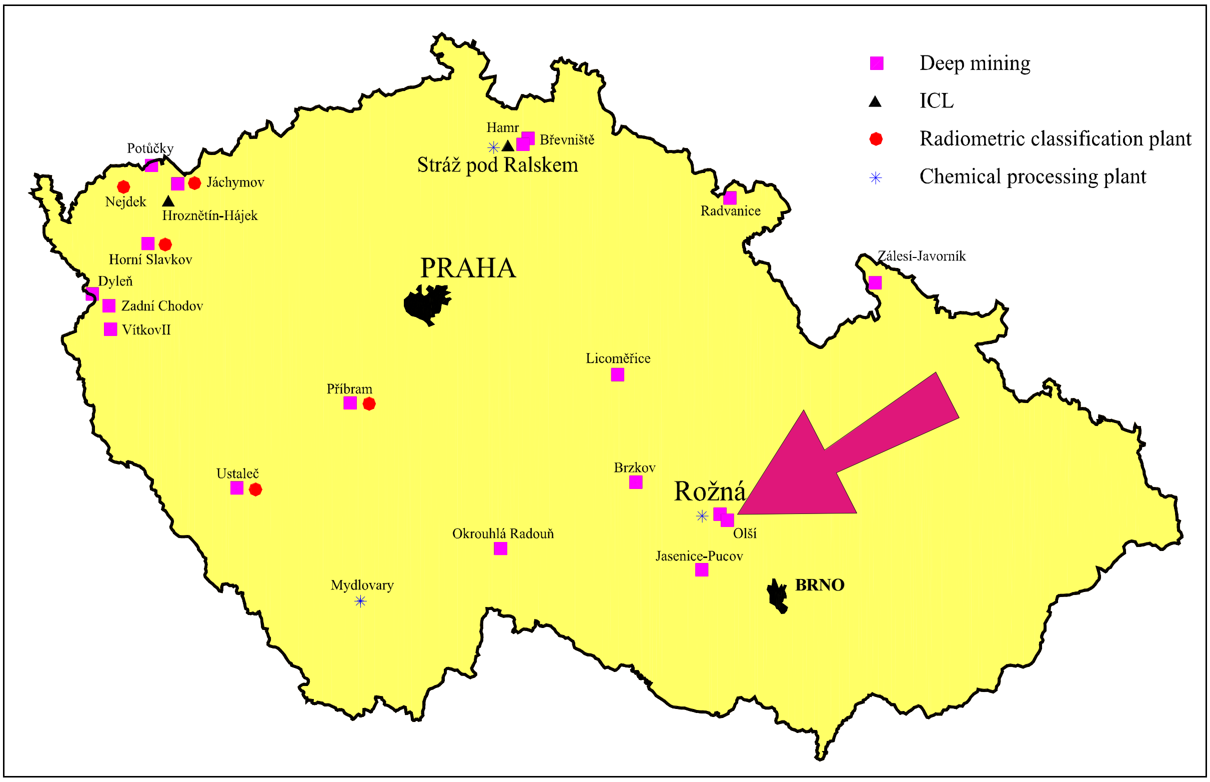


Fig. IV-25. Location of the Rožná site within the Czech Republic.

IV-3.2. Geology and hydrogeology

The deposit is located in a formation of metamorphic sedimentary/effusive rocks of Precambrian age, represented mainly by gneisses and amphibolites. Tectonic zones reach up to 10-15 km in length, their average width is in the order of several metres. Uranium ores (uraninite and coffinite) are present in depths below 600 m. The uranium mineralisation is of hydrothermal origin and it is confined to fissure structures. The uranium ores contain a significant amount of metallic sulfides, namely pyrite.

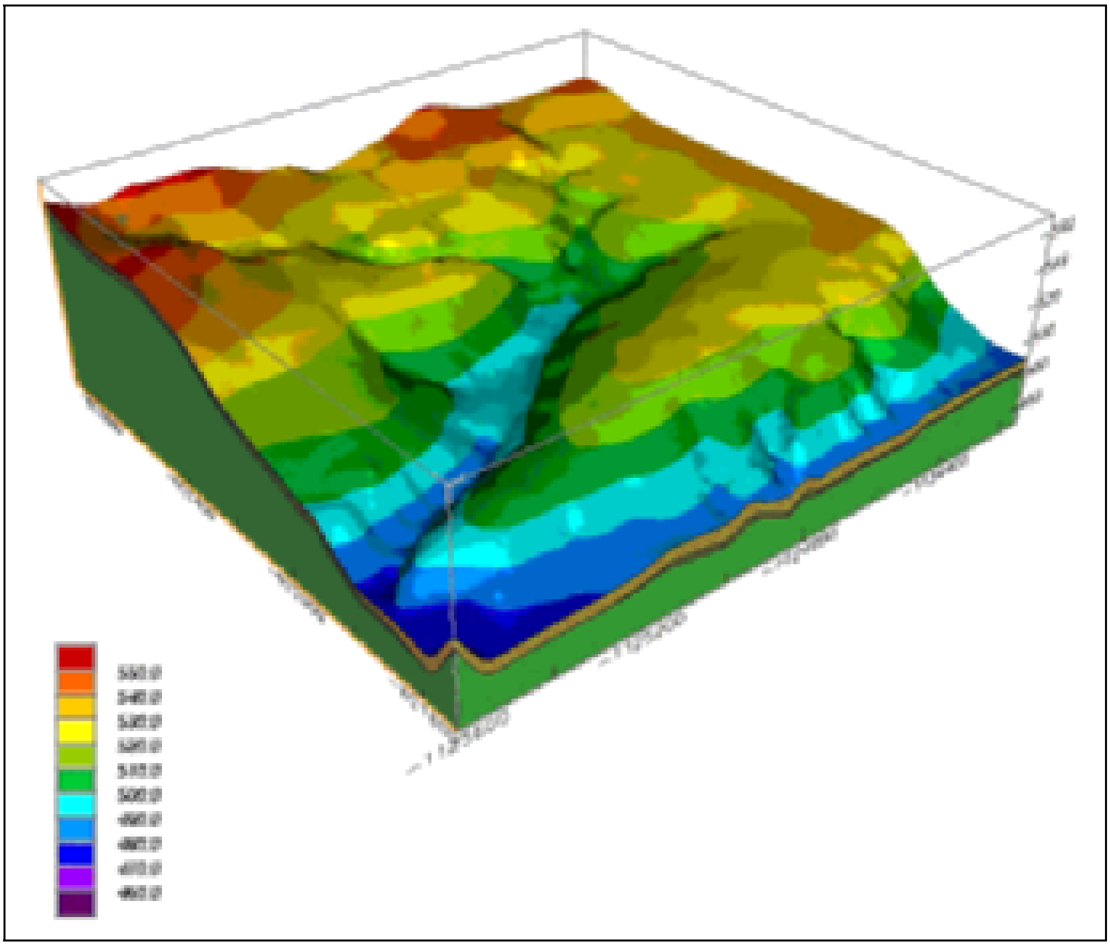


Fig. IV-26. The original contouring in the area of the K1 tailings pond.

Fig. IV-27. View of the tailings pond K1 Rožná – aerial photograph.

The K1 tailings pond is located near the shafts nos. R I and R II of the Rožná mine. The structure of the underlaying rocks is very simple. The underlying rocks, dipping 50° to the West, consist of gneisses and amphibolites with pegmatite and serpentinite lenses and dykes. The width of this tectonic zones is about 0.5 m. The metamorphic rocks are covered by clayey-sandy fluvial sediments. The thickness of the covering sediments varies with a maximum of up to 4 m. The underlying rocks are largely impermeable. Water is present only in fractures filled by crushed rock material. The covering sediments are highly permeable. Subsurface waters drain into Nedvedicka creek.

IV-3.3. Ore Processing

The processing technology is based on ambient pressure carbonate leaching with uranium separation from the pulp by ion-exchange. Uranium concentrate (yellow cake), containing a minimum of 65% of uranium, is the final product. The processing plant is located in a protected water resources zone and, therefore, was required to have a completely closed process water cycle in order to minimise possible negative influences on the environment.

Two tailing impoundments were built for the disposal of residues: K1 Rožná (built 1965 – 1968) and K2 Zlatkov (built 1978 – 1980). Both tailing ponds were originally of the valley type (Fig. IV-2), but the K1 pond was later reconstructed with a circumferential dam. The K2 tailing pond is currently not in operation and is used as a repository for remediation residues (Fig. IV-3).

The K1 pond is still in operation. The dam of this tailings pond is made up from waste rock without any sealing, heightened step by step from the fifth level with dams 3.5 m of high in the direction to the central part of the tailings pond. These dams are founded on the deposited tailings. A basic condition for safe operation of a tailings pond is a minimum horizontal distance of 15 m between the free water level and a dam made of waste rock alone. A sealing of the dam body is provided by sedimentation of fine-grained tailings on the inside face of the dam. The permeability of the dam was calculated from the drainage measurement to be 0.75 m per day (= 8.7×10^{-7} m/s). The tailing impoundment is protected against inflow of surface waters by diverting channels. Seepage waters from the tailings impoundment are collected by drains and pumped back into the pond.

Table IV-8. Technical parameters of the Rožná K1 tailing pond.

area	64.50 ha
Deposited material	$12.8\cdot10^6$ t
volume of free water	1 105 786 m^3
volume of deposited material	$9.62\cdot10^6$ m^3
total capacity	$11\cdot10^6$ m^3
volume of drained water per year	420 000 m^3
max. thickness of deposited material	48 m
max. height of dam	53 m

Taking into consideration the expected close-down of production, a new concept for heightening of the dams has been implemented. The inner part of the pond was divided by dams into sedimentation compartments and a water accumulation compartment (the central part of the tailings pond). Internal dams are running parallel to the circumferential dam at a distance of 20 m. The space between the dams was filled in with pulp and covered by waste rock up to the top level of the circumferential dam. Some technical parameters of the K1 pond (as of 1 January 2003) are collated in the Table IV-1.

IV-3.4. Water management

The uranium processing technology (carbonate leaching) operates in a closed water cycle. Technical measures have been implemented with the aim to collect seepage waters, to protect the pond against surface water inflow and others. All drained waters, as well as all seepage waters, are pumped back to the pond. The closed water cycle is not balanced at present. The total volume of free water increases every year. This volume increase is caused by a lower water content (37.5%) of the tailings against the projected one (60%) rather than by the imbalance between precipitation and evaporation. It is important to note that the water balance is also affected by groundwater seeping through geological fractures into the drainage system. This groundwater mixes with contaminated seepage water and increases the volume of waters necessary to be treated. The total amount of drainage waters is approximately 600.000 m^3 per year (approximately 420.000 m^3 from the K1 pond).

As mentioned above, also amount of ore processed affects the free water volume. Tailings deposited into the pond after processing contain about 0.375 m^3 of water per 1 ton of ore. The total volume of fixed water in the deposited tailings is in the order of 4.8×10^6 m^3.

IV-3.5. Water treatment

The excess of water in the system has to be compensated by discharging treated water. The chemical composition (Table IV-2) of the water does not allow it to be released directly into surface water recipient. A combined water treatment is being used – evaporation with crystallization of disodium sulphate for waters with high content of salts, and electrodialysis for water with low content of TDS.

Table IV-9. Chemical composition of free and drainage water of the Rožná K1 pond (for the year 2002).

Component	Average concentrations [mg/l] free water	drained water
pH	8.2	7.6
U	13.2	12.7
Ra [mBq/l]	1085	195
TDS	29805	15084
SO_4^{2-}	17431	8201
HCO_3^-	546	636
NO_3^-	1140	665
NO_2^-	1144	131
Cl^-	325	194
F^-	11	2
NH_4^+	214	45
Na^+	9427	5097
K^+	115	82
Ca^{2+}	345	191
Mg^{2+}	82	74

An evaporating unit with a maximum capacity of 210,000 m^3 per year was built in 1976. The treatment unit was complemented with an electrodialysis (EDA) unit of 65,000 m^3 per year capacity in 1984. Since 1976 some 4.9 million cubic metres of water have been treated and released into the Nedvìdièka creek (on the average 204,000 m^3 per year).

The origin of the sulphate ions is to be sought in the oxidation of pyrite during the processing of the uranium ore. The reaction is not completed in the mill and some residues of that mineral phase are deposited with tailings into the pond where the oxidation process continues for a long time. It results in conditions leading to additional leaching of residual uranium in the tailings.

Comparing the results of the chemical analysis of free and drainage waters shows that the concentrations of most components proportionally decrease with diluting by clean groundwater, except for uranium. This indicates that some leaching of residual uranium from the tailings takes place in deeper layers. It can also be documented in the vertical profile of chemical composition of the pore water (Table IV-3, Fig. IV-4). While the concentrations of uranium close to the surface of the tailings correspond very well with free water concentrations, at a depths between 15 and 25 m they are about 3 times higher, and in the deepest layers they are up to 7 times higher.

This problem led to a new concept for the water treatment. Beside the two operational systems, which will be reconstructed for a higher capacity, a third, new system based on uranium removal from seepage water by ion-exchange will be installed.

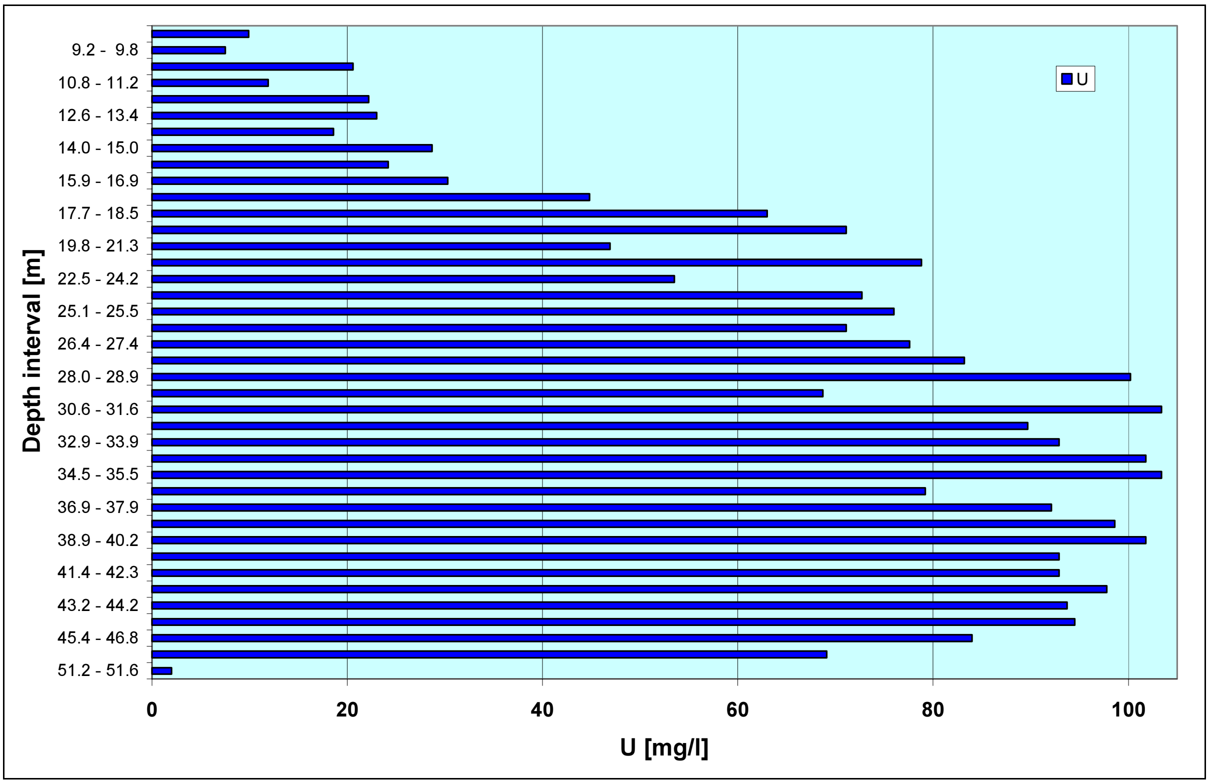


Fig. IV-28. Vertical profile of uranium concentrations in pore waters of Borehole KI – 1.

Table IV-10. Vertical profile of the chemical composition of pore water in Borehole KI – 1.

sample number	depth interval [m]	pH	redox potential [mV] *)	conductivity [mS/m]	TDS [g/l]	component concentration									
						NH4	Na	K	Mg	Ca	Mo	U	SO4	HCO3	NO3
						[m g / l]									[mg/l]
1-6	**8.8 - 9.2**	7.89	126	2613	28.20	172.2	6360	144.4	99.0	282.8	3.50	9.9	16970	562.5	890.4
1-7	**9.2 - 9.8**	7.80	124	2613	27.36	161.4	7780	157.6	96.6	97.1	4.75	7.5	16770	568.2	787.2
1-8	**9.8 - 10.8**	8.16	107	3118	27.84	216.1	8480	139.4	67.5	48.9	7.06	20.6	17840	1031	829.2
1-9	**10.8 - 11.2**	7.94	119	2849	27.72	201.8	8340	147.1	86.7	133.5	5.72	11.9	17550	832.9	852.0
1-10	**11.8 - 12.6**	8.53	127	3022	29.72	195.5	9740	126.3	42.2	19.8	7.06	22.2	18230	1770	860.4
1-11	**12.6 - 13.4**	8.56	115	3441	34.74	222.8	11770	140.2	33.1	11.3	8.77	23.0	21720	2666	874.2
1-12	**13.4 - 14.0**	8.67	112	4634	35.72	240.4	11500	152.1	36.0	12.2	8.21	18.6	21920	2603	870.0
1-13	**14.0 - 15.0**	8.69	56	3516	36.20	264.6	11990	151.9	18.6	6.8	8.44	28.7	22490	3617	576.0
1-14	**15.3 - 15.9**	8.60	76	3624	35.44	270.4	12470	141.8	21.2	7.2	7.90	24.2	22490	3280	648.0
1-15	**15.9 - 16.9**	8.68	69	3484	34.14	260.6	12260	142.0	16.0	6.5	7.72	30.3	22300	3399	471.6
1-16	**16.9 - 17.7**	8.66	67	3452	33.98	268.9	10360	163.8	14.9	8.1	8.04	44.8	21330	3600	206.4
1-17	**17.7 - 18.5**	8.49	99	3806	39.98	216.1	12340	148.1	31.7	7.8	10.19	63.0	25400	3192	136.8
1-18	**18.5 - 19.5**	8.48	86	4054	43.66	214.9	14770	145.2	34.7	14.4	10.49	71.1	27730	3058	20.4
1-19	**19.8 - 21.3**	8.62	76	3925	40.10	304.8	12220	128.5	22.0	14.0	8.52	46.9	25590	3883	26.4
1-20	**21.3 - 22.5**	8.66	89	4075	39.46	425.0	12770	108.8	20.3	4.3	8.08	78.8	24720	4912	116.4
1-21	**22.5 - 24.2**	8.60	77	3871	39.46	383.8	12450	123.2	20.7	14.6	8.39	53.5	25210	3970	115.2
1-22	**24.2 - 24.8**	8.60	94	3989	38.50	473.8	13450	131.5	22.7	8.4	8.54	72.7	24430	4607	404.4
1-23	**25.1 - 25.5**	8.59	86	3871	39.28	467.4	13700	127.7	21.4	9.4	8.56	76.0	24340	4575	517.2
1-24	**25.5 - 26.4**	8.59	82	3957	37.96	439.9	12440	131.5	21.2	10.4	8.98	71.1	23760	4165	612.0
1-25	**26.4 - 27.4**	8.59	95	3806	37.04	489.6	11900	138.8	19.4	10.1	9.11	77.6	22680	4396	868.8
1-26	**27.4 - 28.0**	8.68	81	3882	37.30	654.7	11120	165.6	17.4	8.1	8.76	83.2	21520	4993	1778
1-27	**28.0 - 28.9**	8.60	82	4054	38.36	875.4	10790	129.7	18.2	10.0	8.12	100.2	21140	5611	2395
1-28	**29.2 - 30.6**	8.66	104	3860	39.96	544.8	12370	215.1	24.2	10.3	8.86	68.7	23860	4224	1650
1-29	**30.6 - 31.6**	8.61	89	3871	38.12	875.9	10950	165.2	21.2	6.3	8.24	103.4	21330	5202	3510
1-30	**31.6 - 32.9**	8.60	104	3710	34.88	829.5	10930	145.8	18.4	8.5	6.29	89.7	19490	4714	2915
1-31	**32.9 - 33.9**	8.62	94	3720	33.86	774.9	10400	122.4	15.4	6.3	6.24	92.9	19200	4599	2624
1-32	**33.9 - 34.5**	8.56	76	3731	33.82	816.1	10480	114.1	23.8	6.1	6.08	101.8	19200	4681	2645
1-33	**34.5 - 35.5**	8.59	89	3763	33.56	786.0	10690	101.2	15.6	5.9	6.02	103.4	19010	4670	2618
1-34	**36.2 - 36.9**	8.61	86	3785	34.02	751.4	9860	216.5	14.5	6.8	6.63	79.2	19490	4403	2551
1-35	**36.9 - 37.9**	8.66	73	3753	33.30	802.4	10020	214.1	14.5	6.0	6.98	92.1	19200	4251	2336
1-36	**37.9 - 38.9**	8.59	85	3828	33.50	958.0	10950	148.1	16.2	5.9	7.88	98.6	19200	4681	2549
1-37	**38.9 - 40.2**	8.55	92	3935	34.52	1064.7	9880	135.7	18.2	6.6	8.30	101.8	20070	4669	2232
1-38	**40.2 - 41.4**	8.57	94	3871	35.20	975.2	11160	169.1	18.2	4.7	7.67	92.9	20170	4470	2677
1-39	**41.4 - 42.3**	8.57	92	3914	35.86	1022.6	11550	190.5	20.2	5.7	8.19	92.9	20750	4189	2660
1-40	**42.6 - 43.2**	8.51	104	3957	35.22	946.4	11700	161.2	22.2	6.7	8.02	97.8	20460	4332	2760
1-41	**43.2 - 44.2**	8.51	110	3882	35.28	866.6	11270	163.8	23.2	5.5	8.15	93.7	20650	4028	2196
1-42	**44.2 - 45.4**	8.50	103	3871	35.54	860.5	11170	161.4	25.0	6.0	8.28	94.5	20360	4033	2732
1-43	**45.4 - 46.8**	8.61	84	3796	34.74	724.4	11470	147.7	18.6	4.6	8.06	84.0	19760	4926	2292
1-44	**50.2 - 50.9**	8.45	94	3473	29.24	609.4	10150	128.8	49.4	8.6	4.79	69.1	16260	4427	1878
1-45	**51.2 - 51.6**	7.93	98	946	---	98.5	2800	97.0	54.5	101.8	---	2.0	5860	461.6	---
1-47	**51.7 - 51.9**	8.45	70	1688	---	83.3	2170	67.7	99.5	123.6	---	---	8740	2390	---

*) Pt vs. Ag/AgCl (3M KCl) system

IV-3.6. Geotechnical parameters and geodaetic measurements

The geotechnical parameters of the deposited material are the basic data for calculations of the stability of tailings impoundments. Under the PHARE project., core drilling through the deposited tailings was undertaken (Fig. IV-5.). Six boreholes (K1-1 to 6) were drilled around the K1 pond, five of them from the top of the internal dams, and one from the fifth stage of the main dam. Two types of samples were collected:

(1) samples for the separation of porewaters and the analysis of chemical composition of both, liquid and solid phases,
(2) undisturbed samples for laboratory tests for geomechanical parameters.

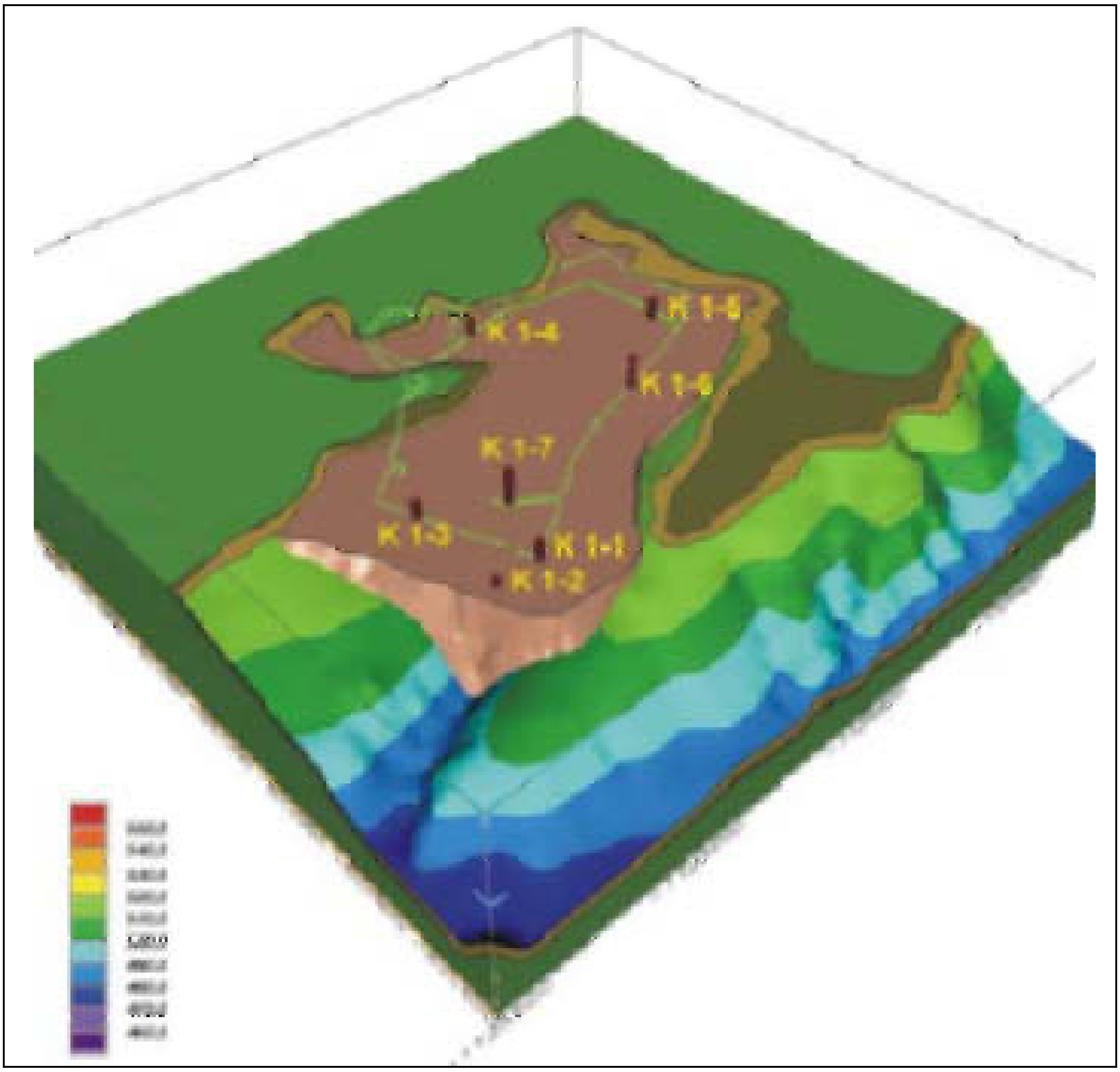


Fig. IV-29. Position of investigation boreholes at Rožná tailings pond K1.

The typical vertical profile of the circumferential part of the pond consists of vertical layers of coarse-grained 'sandy' sediments that are interlayered with fine-grained sludges. This depends on the former system of deposition. Such vertical profile is usually called a 'transition zone'. The average values of selected basic parameters for the deposited tailings are shown in the Table IV-4.

Table IV-11. Average values of geotechnical parameters of the Rožná K1 tailing pond.

Paramter	Average value	Unit
density	1753	$kg \cdot m^{-3}$
density of dry residue	1183	$kg \cdot m^{-3}$
moisture content of deposited material	37.5	%
porosity	58.5	%
saturation	97.5	%
permeability of fine-grained layers	6.9 - 7.83	$\times 10^{-8}$ $m \cdot s^{-1}$
permeabilty of coarse-grained layers	1.8 - 1.97	$\times 10^{-4}$ $m \cdot s^{-1}$

From the beginning of the tailing pond construction on, a geodaetic monitoring of the dam was undertaken at regular intervals. The vertical, as well as horizontal movements of geodaetic points located on each additional step of the heightened dam are monitored with great accuracy every year. The results of this monitoring gives a good idea about the consolidation of the deposited material during the operation of the tailing pond. Average vertical movements varied around 10 mm per year, some extreme values reaching up to 20 mm per year. The relatively low consolidation rate of the sludges is caused by the fact that the process of dewatering of the tailings has not yet started due to the water level in the central part of the pond being too high. An increase in the consolidation rates is achieved by pumping out the decant water and starting to load the interim cover material on the top of the pond. The prediction of the resulting settling process is the main goal of this project.

IV-4. REMEDIATION CONCEPT

The aim of the remediation of both of the tailing ponds is to blend the remediated ponds into the existing landscape. The remediation involves the dewaterig and treatment of contaminated waters (free water, porewater, seepage water, drainage water) from the tailings impoundments. The final goal of the remediation process is to protect the deposited tailings against infiltration of ambient precipitation, as well as against groundwater inflow. Protection against direct γ-ray exposure and radon emanation is an integral part of remediation activity.

Two variants of the remediation concept were originally investigated: After evaluation of all aspects (technical, environmental protection, deep rock protection etc.) and public discussion the ”A” variant would be implemented.

Variant A - Consecutive removal and treatment of excess free water is assumed in this variant. A combination of the currently operated treatment technology (evaporation + electrodyalysis) with ion-exchange for the removal of uranium would be necessary.

Variant B - Quick variant – pumping all free non-treated water into the deepest level of the Rožná mine after closing down of the mining operation. The currently operated treatment technology would be applied to the drainage water plus a new technology for uranium removal.

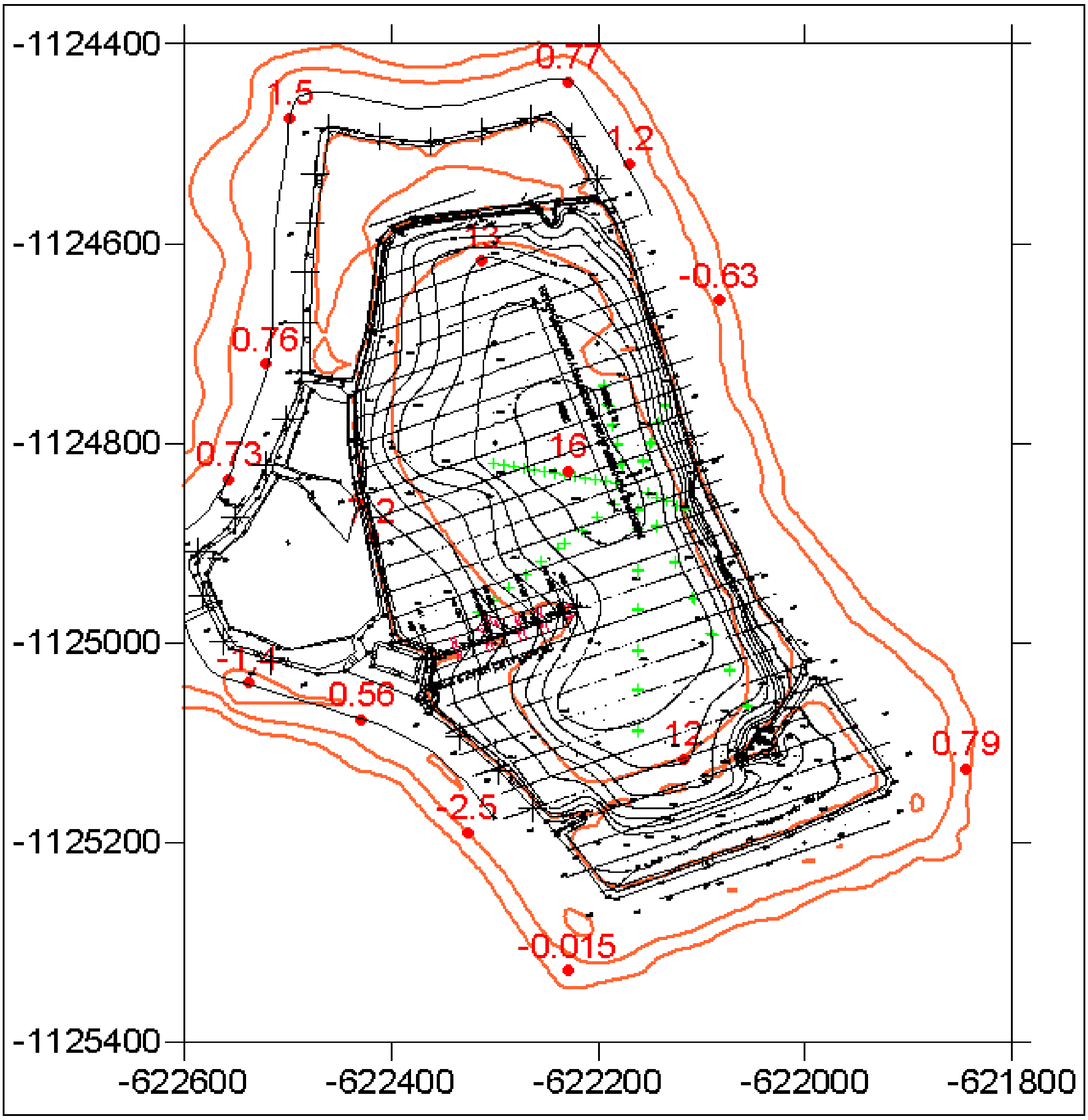


Fig. IV-30. Final contouring of K1 tailings pond. Red numbers indicate elevation or depression of final contour comparing the situation in June 2002.

The variants differ only in the way in which the free water is managed. The remaining elements of remediation are the same. The actual remediation will start, therefore, with the removal of free water. The K1 pond will be stabilised, reshaped and covered with a protective layer. During this period (approx. 8 – 10 years) the K2 pond will be used as a temporary reservoir for drainage water. After the covering is completed, a temporary basin will be constructed on the top of covered K1 pond and remediation of K2 will begin. Finally, both ponds will be covered with topsoil and be revegetated.

The remediation concept assumes a contouring of the top part of the K1 tailings pond by infilling the free space with inert material in order to achieve a maximum slope of 3 %. After lowering the water level in the impounded material, the upper part of the main dam, together with part of the dewatered tailings will be relocated closer to the middle of the pond with the aim to achieve an even slope of the impoundment. (Fig. IV-6).

After the infilling of the free space is completed, the whole surface of the impoundment will be covered by a protective layer. Two variants for the construction of this layer with different materials were investigated.

Preliminary calculations of radiation protection effects and construction requirements (frost-free foundation) lead to a required thickness of 0.8 m for this protection cover layer. Some data for different designs of the cover layer together with calculations of the shielding effects are presented in Table IV-5. The calculations of radiation protection effects were done for two recommended regulatory limits: 1 μGy per hour for γ-radiation, and 0.8 Bq/m·s for radon emanation. As can be seen from the results, all suggested cover layers are in compliance with regulatory limits. The final decision on which type of covering will be used depends on the actual situation at the time of carrying out of the remediation work. Several factors will affect this decision, e.g. availability of materials, transport distances, cost etc.

Table IV-12. Selected design variants of cover layer with calculated shielding effect for each partial layer.

Type of sealing	**Clay**			**Clay**		
Type of drainage	**Crushed rock**			**Synthetic drain**		
	Thickness [m]	Surface γ dose [$Bq/m^2 \cdot s$]	Rn emanation [μGy/h]	Thickness [m]	Surface γ dose [$Bq/m^2 \cdot s$]	Rn emanation [μGy/h]
Recent values	-	18	11	-	18	11
Sealing	0.6	0.018	0.034	0.6	0.018	0.034
Drainage	0.15	0.172	0.0122	0.018	0.018	0.034
Cover	0.45	0.0151	0.0005	0.6	0.013	0.0005
Top soil	0.20	0.0143	<0,0005	0.2	0.0117	<0,0005

Type of sealing	**Synthetic material**			**Synthetic material**		
Type of drainage	**Crushed rock**			**Synthetic drain**		
	Thickness [m]	Surface γ dose [$Bq/m^2 \cdot s$]	Rn emanation [μGy/h]	Thickness [m]	Surface γ dose [$Bq/m^2 \cdot s$]	Rn emanation [μGy/h]
Recent values	-	18	11	-	18	11
Sealing	0.07	0.36	9.71	0.07	0.36	9.71
Drainage	0.15	0.33	3.47	0.018	0.36	4.95
Cover	0.45	0.26	0.153	0.6	0.26	0.077
Top soil	0.20	0.24	0.038	0.2	0.235	0.019

IV-5. WORK CARRIED OUT DURING THE PROJECT

The specific situation at the Rožná K1 tailings pond poses a significant geotechnical problem that has to be solved before other remediation works can commence. The prediction of the consolidation of the deposited material after the surface dewatering and additional loading is the key issue with respect to the stability of the remediated pond. The successful loading of infill material will determine the final efficiency of any long term stabilisation and isolation of the pond because it will determine the success of the planned recontouring of the tailings pond body.

A complex set of field and labratory investigation works has been carried out with the aim to obtain data on fine slime zone, which is the most important part of the pond for future remediation. The results from this investigation programme were evaluated in close co-operation with WISMUT GmbH, using evaluation and modelling tools that have been developed by that company for the planing and design of tailing ponds remediation.

IV-5.1. Borehole drilling

A new, cored borehole K1-7 (Fig. IV-5) has been drilled from the top of the dividing dam with the objective to complement currently available data with others from the central part of the tailings pond. The currently available data do not describe the very fine-grained tailings located below the free water level. The borehole reached the original surface at a depth of 41.3 m from the top of the dam. The full suite of laboratory investigations on the core samples was completed, i.e. soil properties, geomechanical, geochemical tests etc. The soil physical properties were evaluated referring to the major layering of different tailings types. The typical value ranges of soil physical properties in the identified tailings layers in borehole K 1-7 are shown in the Table IV-5. An analysis of the results shows that the borehole has not struck the soft (slime) tailings area, but that it was located in the transition zone, i.e. in a part of the pond, where mixed layers of coarse and fine tailings have been deposited depending on the distance from discharge points [IV-5].

Table IV-13. Geomechanical properties of major tailings layers from borehole K 1-7

Depth below surface [m]	**consistency/ soil class**	**w_n [%]**	**e (v.ratio)**	**ρ_s [g/cm^3]**	**Φ [°]**	**C′ [kPa]**	**shear vane strength [kPa]**
11 – 13	pulpy/ clay, silty	28-34	0.99-1.07	2.70-2.80			10
13 – 20 (13 – 15,5)	weak to stiff/ silt, clayey, sandy layers	29-42	0.87-1.20	2.78-2.85	25.0 – 31.4 33.8 – 34.5	5 – 22 8 – 15	19-29
20 – 23.4	weak to stiff/ silt, clayey,no sand	40-46	1.18-1.31	2.81-2.84	31.4	5	25
23.4 – 33.3	stiff/ clay, silty	37-44	1.06-1.22	2.81-2.85	29.1 – 29.8	4 – 7	29
33.3 – 41.3	low cohesive/ sand, silty	20-31	0.62-0.83	2.72-2.80	37.3	16	57

IV-5.2. Fine slime sampling below the water level

Since the new borehole did not furnish a typical soft tailings profile, additional sampling of very fine unconsolidated tailings was undertaken. Using a special device (UWITEC Corer) for sampling below the water level, a total of five samples were collected. Only one of them (P3), however, turned out to be suitable for full scale laboratory tests, including the physical soil properties and oedometer test for the determination of the basic functions $e \rightarrow \sigma'$ and $e \rightarrow k$. The results obtained are typical for unconsolidated fine tailings settling in the fine slime zone furthest from the discharge point: the void ratio of fine tailings from sample P3 was e_{1kPa}= 2.65, the respective compression was determined to be C_c= 0.48, the stiffness modulus increased with loading from 0.1 MPa at 1.25 kPa to 4.5 MPa at a 200-400 kPa additional load.

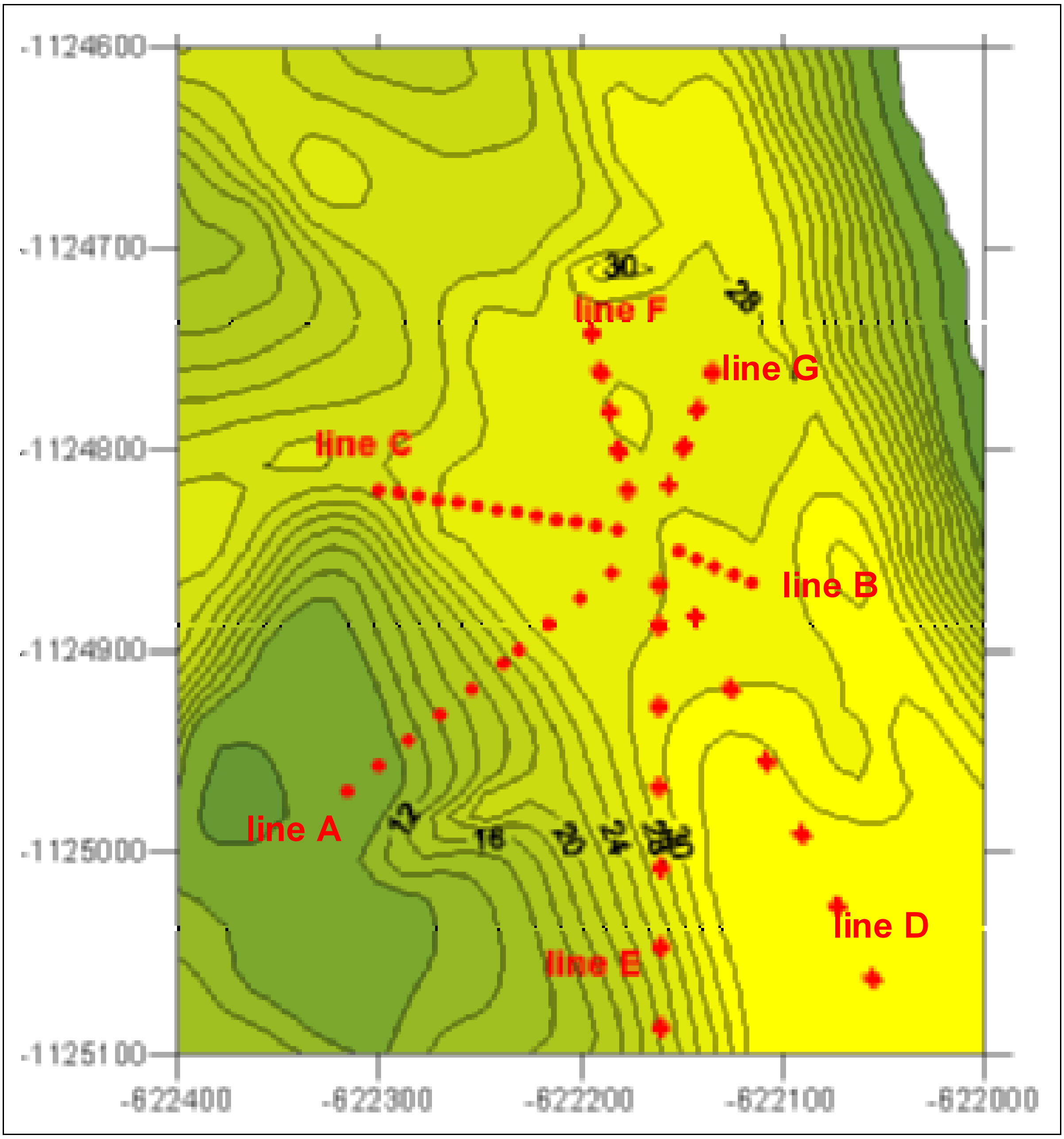


Fig. IV-31. Location of the CPT measurement points except for the body of the dam. Black contour lines indicate the total thickness of the tailings.

IV-5.3. Identification of fine slimes areas

IV-4.3.1 Cone penetration tests

For the evaluation of the potential settlement process it is necessary to define the occurrence of mainly fine slime zone. Two indirect methods have been used: (a) cone penetration tests (CPT), and (b) shear vane tests (SVT). Natural conditions (total depth of free water) and technical limitations of the devices used only permitted to carry out CPTs to a maximum depth of 12 m below the tailings surface, and SVTs to 7 m. The tests were undertaken at about 70 points (both for CPT and SVT) in the central part of the pond (Fig. IV-7).

The measured CPT profiles can be grouped into two categories:

— Type I: profiles showing cone resistance continuously increasing with depth
— Type II: profiles showing cone resistance readings significantly varying over depth.

Type I profiles indicate homogeneous material distribution over depth range at the measurement point. This type of profile mainly occurs along lines E and C and to some extent along line A; with line E profiles exhibiting a significantly slower increase in cone resistance with depth than line A profiles. It may be noted that readings along line E and some profiles along line C define an area of very low cone resistance down to greater depths. In extension of line E this would also include inner profiles of line F (having a layered structure). Within the area of measurements, these profiles apparently form a 'channel' consisting of very soft materials.

Profiles showing varying cone resistance (Type II) indicate layered structures within the tailings deposit. Measuring points along the lines D, G, B and F belong to the Type II. These profiles were obviously recorded in what is known as 'intermediate' zones that comprise alternating layers of sands and slimes.

Cone resistance distributions calculated from discrete measured values confirm this assumption. Figure IV-8 shows low cone resistance zones down to greater depths (ca.10 m) along lines E and C. As a consequence, the slimes deposit is to be expected to occur in these areas of the tailings pond.

It should be noted that the confidence interval of the calculated distributions is limited to the centre of the 'star-shape' formed by the lines. Structures beyond this area are not based on these recordings. They have to be interpreted as extrapolation errors as a consequence of missing measurements. Available CPT data indicate that the slimes zone is mainly located in the area around line E and around parts of lines C and F.

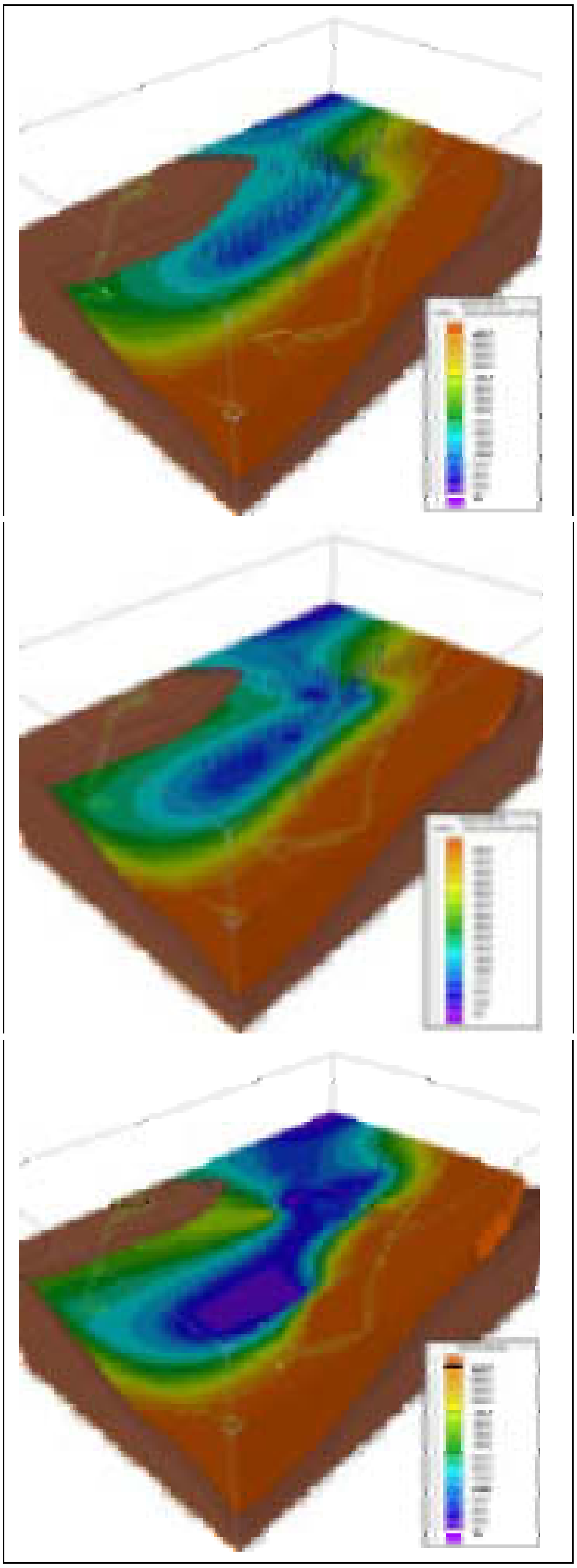
IV-4.3.2 Shear vane tests

For stability predictions, peak shear strength is of primary significance. The data in the near-surface area (up to 2 m below tailings upper edge) range from approx. 1 kPa up to more than 20 kPa. This indicates that either very different varieties of tailings or tailings in different states of consolidation were sampled. Sandy tailings typically exhibit shear strength values in excess of 5 kPa that rapidly increase with depth. Experience of Wismut GmbH shows that the shear strength of soft tailings near the surface is less than 5 kPa even after several years of self-weight consolidation; such situation requires very careful approach to remediation and cover placement. Figure IV-9 clearly shows two different distributions of depth profiles:

— shear strength increase with depth up to values of about 25 kPa (Line C), as well as
— very low rise in shear strength distribution (Line E).

Values recorded along lines A, B and C show very low shear strengths of less than 10 kPa down to a depth of 2 m below the surface. However, the shear strength at these measurement locations increases significantly with depth.

The shear strength data obtained are in good agreement with CPT results. Low values from CPTs at measuring points along line E, indicating a relatively homogeneous slime distribution with depth, are confirmed by very low shear strength values. Shear strengths measured along line E did not exceed 5 kPa. This is also the case for the three profiles VRF42, VRF62 and VRF82 located near the platform. These profiles are a direct extension of line E.



515 m a.s.l.

518 m a.s.l.

521 m a.s.l.

Fig. IV-32. Interpretation of the CP testing inside the body of tailings. Violet to blue colours indicates fine slime zone.

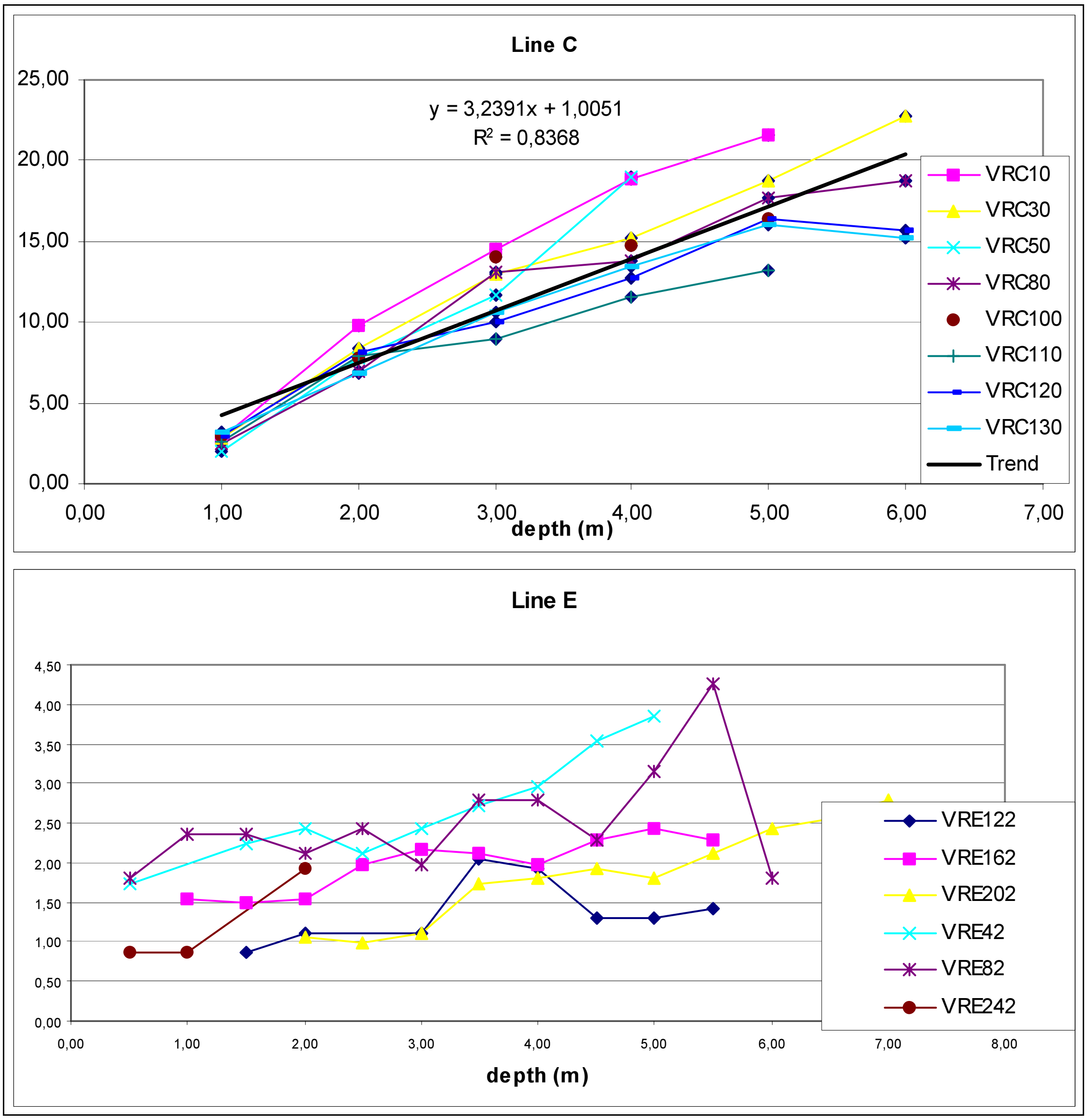


Fig. IV-33. Typical examples of two different types of vertical distribution of shear strength. Line C – sandy or intermediate zone; Line E – fine slime zone.

The information gained from the assessment of CPT data on the distribution of the slime deposit mainly around line E is corroborated by the distribution of the SVT values.

No materials characteristic of soft tailings could be obtained from the investigation programme carried out on samples from boreholes K 1-1 to K 1-7. In earlier Reports [IV-5, IV-6], the samples from these boreholes were assigned to the tailings beach or transition zone, respectively. A shallow sample taken in 2002 with an UWITEC Corer exhibits characteristics that are typical for soft tailings. In the absence of further samples taken at greater depths, it is assumed that these material properties would also be valid for the underlying tailings. This assumption is corroborated by results from CPTs that identified soft tailings along line E down to depths of about 10 metres [IV-6].

IV-5.4. Theoretical and modeling work

Oedometer tests results give the following e→σ'relationship:

$$e= -C_c \cdot \log \sigma' + e_0 \qquad \text{[IV.1]}$$

where $C_c = 0.48$, and $e_0 = 2.65$

Conductivity data were derived from samples taken from borehole K 1-7 in the transition area.. On the basis of the k→e relationship below (Eq. IV.2), a conductivity k of 10^{-8} m/s is assumed together with a void ratio e of 2 for the soft tailings area:

$$k= C \cdot e^D \qquad \text{[IV.2]}$$

where $C=5\times10^{-10}$ m/s, and $D= 4.5$

No void ratio *vs.* depth profile is available for the soft tailings area. The sample taken with an UWITEC Corer had a void ratio of e= 2.65. The average void ratio in the soft tailings area exceeds that one in the transition area around borehole K 1-7 of approx. e= 1.

IV-4.4.1 Modelling of the infilling history

The process of filling and self-weight consolidation was simulated for the locations of the fine tailings using the non-linear finite strain consolidation model FSCONSOL [IV-7]. The purpose of this modelling work is (i) to determine representative material functions (e→σ' and e→k_f relationships) for a given tailings type and (ii) to estimate the current degree of consolidation.

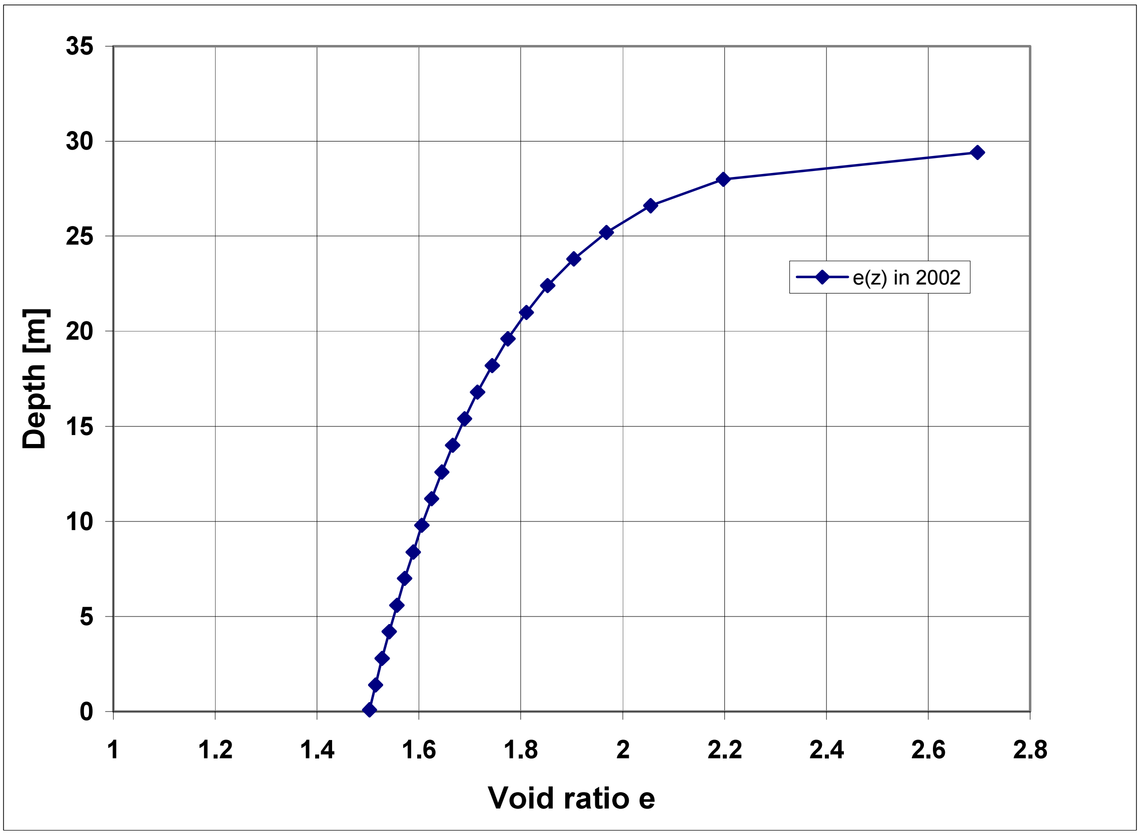


Fig. IV-34. Modelled void ratio vs. depth due to self-weight consolidation (without additional load) for the year 2002.

The consolidation model requires as input the infilling rate, slurry density (initial void ratio) and consolidation properties of the tailings (i.e. $e \rightarrow \sigma'$ and $k_f \rightarrow e$ functions). The rate of filling and initial dry densities of the tailings are estimated from the historic discharge records. Initial guesses for the non-linear material functions ($e \rightarrow \sigma'$ and $k_f \rightarrow e$) are taken from the laboratory analyses.

A deposition time of 34 years (1968 – 2002) is assumed for the thickest tailings deposits of about 30 m. Deposition rates were already assessed to be 3.5 kg/d·m^2 – 4 kg/d·m^2. A 10 years shorter deposition time for the thinner tailings profiles of about 20 m is assumed.

Applying the functions above to the 30 m thick tailings layer produces the void ratio depth profile shown in Figure IV-10, which develops due to self-weight consolidation, i.e. without additional load. The average void ratio is e= 1.8, giving an average hydraulic conductivity of 6.6×10^{-9} m/s.

IV-4.4.2 Estimated settlements due to additional loads

The plans for final contouring call for a heightening of the infilling by about 5 to 15 metres in the area of deep soft tailings layers. The resulting additional load varies from 100 to 300 kPa. It is assumed that a one metre layer of cover material results in additional load of 20 kPa. In order to assess load-induced settlements in the soft tailings area, settlements induced by additional loads of 100, 200, and 300 kPa were calculated for two tailings profiles of 20 m and 30 m thickness, respectively. From that, relative settlements can be calculated. Calculation results are listed in Table IV-7. The results indicate that expected relative settlements are in the range of 8% to 14%, depending on the thickness of tailings and additional loads.

Table IV-14. Calculated settling of two different tailings thicknesses, due to three different additional loads.

		Additional load [kPa]		
		100	**200**	**300**
Tailings thickness before loading	m	21.3		
Tailings thickness after loading	m	19.3	18.7	18.3
Relative settlement	%	9.5	12.3	14.1
Net increase in elevation	m	3.0	7.4	12.0
Tailings thickness before loading	m	29.6		
Tailings thickness after loading	m	27.1	26.3	25.8
Relative settlement	%	8.3	10.9	12.7
Net increase in elevation	m	2.5	6.8	11.3

The lines 'Net increase in elevation' in Table IV-7 indicate the actual change in surface elevation (compared to the year 2002) due to settlement plus the thickness of the additional load. It may be concluded that a planned thickness of the additional load of about 15 m will produce a change in surface elevation of approx. 11m. The designed final surface elevation of 542.45 m a.s.l. (Fig. IV-6) would drop to ca. 539 m a.s.l.

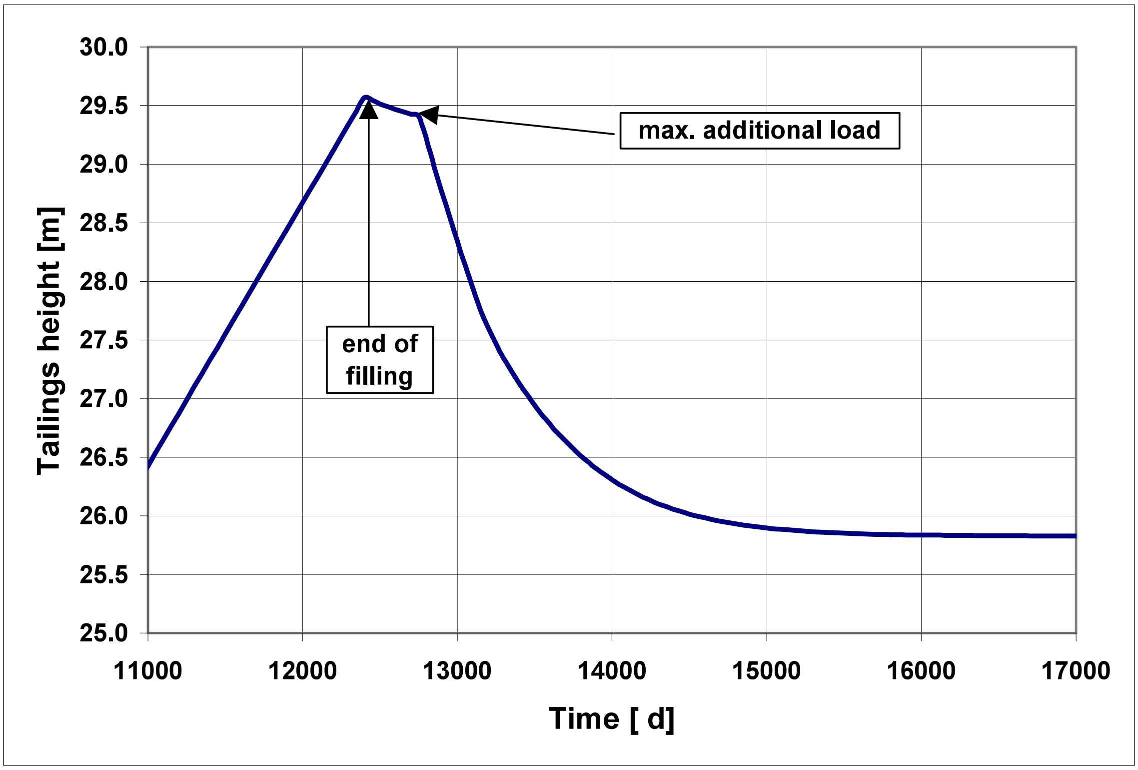


Fig. IV-35. Evolution of the settlement under an additional load of 300 kPa (equivalent to a 15 m cover). It is assumed, that the load increases linearly over one year from 0 to the maximum.

Considering the available materials parameters, any estimate of time evolution of the settlement will have a high degree of uncertainty. Laboratory tests are needed to determine conductivities of soft tailings within the relevant area of tailings pond K 1. A rough estimate is, however, possible: Following the emplacement of an additional of 300 kPa to be completed within one year, it will take about 2000 days for 90 % of the settlement to be achieved. This estimate does not take into account measures to accelerate settlement (e.g. installation of vertical drains). Figure IV-11 illustrates the time evolution of settlements for an load of 300 kPa. This load was emplaced and uniformly distributed over a period of one year. In assessing the length of the consolidation phase, it has to be kept in mind that the tailings pond is still active and there is no indication that the pond will be dormant during a couple of years afterwards. The calculation was done under the assumption that remediation and fill placement would begin immediately after the termination of tailings deposition.

IV-6. SUMMARY AND CONCLUSIONS

The currently available information supporting to the remediation concept of the tailings pond K1 at the Rožná site has been collated. The successful long term stabilisation and isolation of the pond is a significant geotechnical problem that has to be solved before other remediation work can commence. A complex set of field, laboratory and modelling investigations has been completed in order to be able to predict the time and technical progress of remediation of the pond.

A new concept for the impoundment of tailings will be implemented in the course of the next few years until the operation of the mill ceases by 2005. The construction of new inner dams have been started to make ‘bridges’ across the pond. The tailings are spread towards the centre of the pond with aim to cover very fine-grained tailings with sandy (or non-sorted) materials. This will be a way to achieve a more compact surface over the finest grained tailings in order to make easier the construction of interim covers after the disposal of tailings has ended.

The second effect of new progress of impounding is expected in order of partial reducing of potential settlement process after additional loading of interim cover material. A reduction of the potential deviation from the designed contours can be achieved by both, reducing the total thickness of the interim cover layer (i.e. additional load), and transformation of the fine slime zone into a state close to the intermediate (or transition) zone that exhibits smaller deviations from the contour as designed.

A secondary effect of the new impounding method is expected in form of a partial reduction of the potential settlement after additional loading of interim cover material. A reduction of the potential deviation from the designed contours can be achieved by both, reducing the total thickness of the interim cover layer (i.e. additional load), and transformation of the fine slime zone into a state close to the intermediate (or transition) zone that exhibits smaller deviations from the contour as designed.

Some additional investigations are envisaged after the impoundment of tailings has ended. It will be necessary to drill a new borehole from the top of one inner dam into the central part of the pond. The purpose is to obtain a full vertical profile for deriving geotechnical properties, namely for the function void ratio *vs.* permeability. This will then be the basic information for modelling of the progress with time of settlement process.

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ANNEX V. FRANCE

METHODOLOGY TO ASSESS THE RADIOLOGICAL IMPACT OF DISPOSALS OF URANIUM MILL TAILINGS AFTER REMEDIATION (SHORT TERM IMPACT)

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V-1. ABSTRACT

Disposal sites of uranium mill tailings are subject to remediation so as to ensure lasting protection of individuals and the environment. The options adopted for the remediation of a disposal for uranium mill tailings must be such that the current radiological and chemical impact can be reduced to levels that are as low as reasonably achievable using the best available techniques at acceptable cost.

The European Directive 96/29/Euratom requires that an effective dose limit of 1 mSv/y is met, whereas the former French regulatory limit was 5 mSv/y, leading to the re-assessment of the radiological impact of the repositories based on more realistic scenarios.

This study follows on from previous work undertaken by the government, COGEMA, and the Institute for Radiological Protection and Nuclear Safety (IRSN) that produced the 'Doctrine regarding the remediation of repositories for uranium mill tailings' [V-2]. On completion of this work, IRSN was asked to provide a detailed and concrete methodology for assessing a realistic radiological impact of disposals after remediation. This methodology only deals with impact assessments just after remediation. It does not tackle the questions of post remediation environment surveillance or disposal structures lifetime assessment, or long-term impact.

V-2. INTRODUCTION

The quantity of uranium mill tailings currently disposed of in France is in the order of 50 million tons, distributed over 20 sites (Fig. V-1), five of which hold more than 5 million tons (Table V-1). Due to the half-life of some radioelements ($t_{½}$ for ^{230Th} is 75,000 y, for ^{226}Ra it is 1,600 y) present in the tailings, the time during which a risk will be present, will be practically infinite, when considering a human life span.

The options adopted for the remediation of a disposal site for uranium mill tailings must be such that the current radiological and chemical impact can be reduced to levels that are as low as reasonably achievable using the best available techniques at acceptable cost. In any case, the objective of the assessment of the radiological impact of a repository for uranium mill tailings after remediation is to demonstrate the ability of the repository to ensure protection of the people, taking into account the current legislation.

The European Directive 96/29/Euratom requires that an effective dose limit of 1 mSv/y is met, whereas the former French regulatory limit was 5 mSv/y, leading to the re-assessment of the radiological impact of the repositories based on more realistic scenarios.

Fig. V-1. Former uranium mining and milling sites in France.

Table V-1. Inventory of uranium mill tailings at French disposal sites

Region	Department	Site	Mill Tailing Quantity [kt]	Radioelement ^{226}Ra [TBq]	[Bq/g]
POITOU CHARENTE	Vendée	La Commanderie	250	1	4.0
PAYS DE LA LOIRE	Loire Atlantique	L'Ecarpière	11,350	182.1	16.0
LIMOUSIN	Haute Vienne	Bellezane	1,556	49.1	31.5
		Brugeaud	5,782	131.2	22.7
		Croix du Breuil	554	3	5.4
		Lavaugrasse	5,681	142. 5	25.1
		Montmassacrot	737	19	25.8
		Jouac	1,347	79	58.6
	Creuse	La Ribière	196	0.85	4.3
AUVERGNE	Puy de Dôme	Rophin	30	0.31	10.3
	Cantal	Saint Pierre du Cantal	605	7.9	13.0
BOURGOGNE	Saône et Loire	Bauzot	16	2.9	181.2
	Saône et Loire	Gueugnon	185	10.4	56.2
LANGUEDOC ROUSSILLON	Lozère	Le Cellier	5,947	43.1	7.2
	Hérault	Lodève	5,223	175. 4	33.5
		Le Bosc	2,970	118. 1	39.7
MIDI-PYRENEES	Aveyron	Bertholène	476	7.6	16
RHONE ALPES	Loire	Bois noirs – Limouzat	1,300	74.6	57.4
ALSACE	Haut Rhin	Teufelsloch et Shaentzel	4	0.025	18.5

The following sections describe the previous methodology used for the radiological impact assessment of uranium mill tailings repositories, as it was practiced since 1990, and some of its results (Section V-2). The the lack of realism for the critical group and the lowering of the dose limit to 1 mSv/y led to a review of this methodology (Section V-3). Its application to the Lodève site is a good opportunity to appreciate the expected consequences of reducing the effective dose limit (Section V-4).

V-3. PREVIOUS FRENCH PRACTICE

V-3.1. Methodology

Authorization to close a mining site and management of uranium mill tailings repositories were governed particularly by the French Decree no. 90-222. This decree introduced the principle of added exposure, or the Annual Added Exposure Rate (French acronym TAETA) that corresponded to the difference between the Annual Exposure Rate due to repository after remediation ($TAET_{final}$) and the natural Annual Exposure Rate before it was operated ($TAET_{initial}$). This natural exposure is due to in particular the presence of natural radioactive substances on the site and in its immediate vicinity.

External exposure — Internal exposure

Inhalation — Ingestion

$$TAET = \frac{A \times t \times 10^{-6}}{5\,mSv} + \frac{[Dust] \times t \times 10^{-3}}{170\,Bq} + \frac{PAE_{Rn222} \times 10^{-6} \times t \times d}{2\,mJ} + \frac{PAE_{Rn220} \times 10^{-6} \times t \times d}{6\,mJ} + \frac{[^{226}Ra] \times b}{7000\,Bq} + \frac{[^{238}U] \times 10^{-3} \times b}{2\,g}$$

$$TAETA = TAET_{final} - TAET_{initial}$$

A	: γ dose rate ($nGy.\,h^{-1}$)
[Dust]	: long-life alpha emitters ($mBq.m^{-3}$)
PAE	: radon Potential Alpha Energy ($nJ.m^{-3}$)
[^{226}Ra]	: Volumic activity of ^{226}Ra ($Bq.l^{-1}$) in water
[^{238}U]	: Concentration of ^{238}U ($mg.l^{-1}$) in water
t	: Exposure time (7000 hours per year)
d	: Inhalation rate of man (0,8 $m^3 \cdot h^{-1}$)
b	: Water ingestion per year (2,2 $l.d^{-1}$ x 365 day = 803 l)
Bq	: Becquerel, equivalent to one disintegration per second
J	: Joule, for radon Alpha Potential Energy (EAP) in nJ (10^{-9}J)
Gy	: Gray, absorbed dose, quantity of energy (J) absorbed by 1 kg of material
Sv	: Sievert, radiance effect on the living tissue

Fig. V-2. TAETA calculation.

The calculation of TAETA was applicable to the most exposed persons, fixing exposure parameters (quantity of ingested water, inhaled air, time spent on the site). The radiological impact on the environment was considered to be acceptable when the TAETA for members of the public was less than 1, which corresponded to an added effective dose of 5 mSv.

Adding for each exposure pathway the ratio of the corresponding value of added exposure over a year and the annual limit (Figure V-1) allowed calculating the TAETA. The annual limits were defined as follows:

- 5 mSv for external exposure;
- 170 Bq for long-lived alpha emitters in the ^{238}U-chain, present in inhaled dust suspended in air;
- 2 mJ of potential alpha energy for short-lived decay products of inhaled ^{222}Rn;
- 7 kBq for ingested ^{226}Ra;
- 2 g for ingested uranium, the daily quantity of hexavalent compounds that can be ingested not exceeding 150 mg.

The Decree 99-222 defined a critical group by the following characteristics:

- exposure times: 7000 h/y;
- inhalation rate: 0.8 m^3/h;
- ingestion over 365 days of 2.2 l/d of water as sampled in the receiving watercourse immediately below disposals.

V-3.2. Available evaluations

The order of magnitude of the added effective doses for the main French tailings repositories that have been evaluated using this methodology is summarized in Table V-2 and Fig. V-3. All these evaluations remain below 5 mSv/y and meet the requirements of Decree 90-222.

Table V-2. Order of magnitude of effective added doses corresponding to a scenario of the type as defined in Decree 90-222, for a critical group under the immediate influence of the main uranium mill tailings repositories [V-1]

Site	External exposure [mSv/y]	Exposure by inhalation [mSv/y]	Exposure by ingestion [mSv/y]	Effective dose from the natural environment [mSv/y]	Added effective dose [mSv/y]
Bertholène	0.9	0.8	0.9	1.6	1.0
Bessines	1.7	1.6	0.2	2.7	0.8
L'Ecarpière	1	0.7	0.2	1.5	0.4
Forez	1.4	1.3	0.2	1.9	1.0
Le Cellier	1.5	0.6	0.3	1.8	0.6

The relative uncertainty of these results is generally around 30% due to the large variability of measured values of the radon activity per unit volume and the external exposure.

The limit of 1 mSv appears to be met. However, it should be noted that these calculations ignore the uncertainties mentioned above.

The new methodology detailed in Section V-3 mainly introduces the definition of realistic population groups and exposure scenarios that makes it possible to assess radiological impacts of uranium mill tailings repositories more objectively with regard to radiological protection objectives.

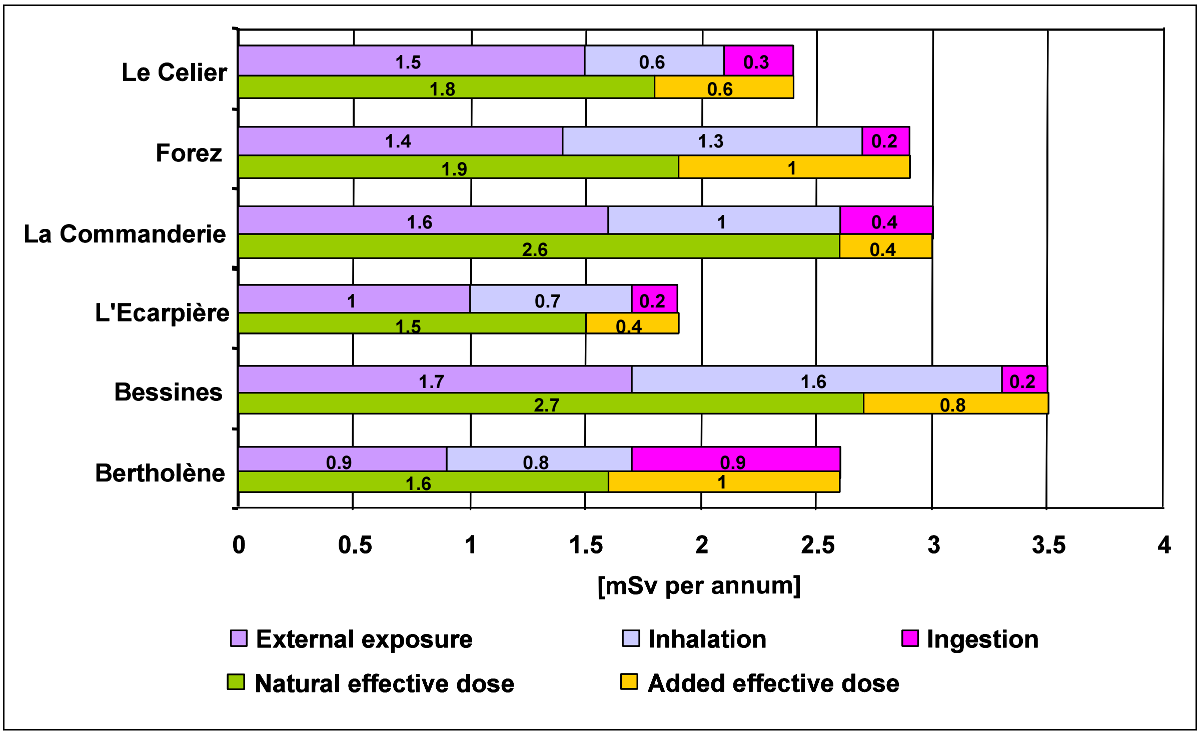

Fig. V-3. Annual doses and dose contributions at selected sites.

V-4. CURRENT METHODOLOGY FOR SHORT TERM IMPACT ASSESSMENT

V-4.1. Data collection

The description of the environment of the site and the site itself is the basis of the impact assessment. All the parameters of the environment that can possibly influence the result of the impact calculation should be examined.

V-3.1.1 The environment at the site

Local meteorological conditions

It is necessary to know accurately the climatic processes that will govern the atmospheric dispersion of dust and radon.

A wind frequency distribution as well as rainfall records should be provided.

All the data must be representative of an average year and should be established from data accumulated for several years.

Moreover, all exceptional events (particularly rain events) have to be compiled properly so as to take into account their influence on the water management.

It is of course necessary that all the data are collected at the site or from a point in the nearby environment.

Geology- hydrogeology

A detailed description of the local geology should be provided, as well as relevant data for the hydrogeology. All information that could help the understanding of groundwater movements should be provided. If a local groundwater model is available it should be presented.

Hydrographical system

Detailed descriptions of the various watercourses that cross the site should be presented. The flow (particularly floods and low water) of the watercourses, as well as drained surfaces, catchment areas, civil engineering features, and tributaries should be mentioned.

Water uses

Domestic and agricultural uses of water have to be clearly known and described. Possible uses for aquaculture, leisure, or tourism should also be described, if relevant.

Agricultural environment

Local cultivations, animal husbandry, and other activities linked with food production have to be described. Soil use is important to describe, as well as the origin of irrigation water. Fishing and aquaculture activities should be described if relevant.

Description of the human environment

Demographic data have to be supplied for the area subject to the impact of radioelements. These data should deal not only with the settlement but also with working populations and other populations (school-children, tourists…).

V-3.1.2 Site description

Description of the wastes

All potential sources of radioactivity have to be listed. For each radioactivity source, quantities, mass activities, physical and chemical stabilization treatments, mineralogy, and geochemistry have to be described.

Disposal structures- remediation options

The repository structures as well as the remediation options have to be described and their durability should be discussed.

Water management on the site

Water management objectives are to organize a selective collection of contaminated and non-contaminated water, and the prevention of erosion. The various systems used as well as their efficiency and durability have to be described.

Various workings on the site

A precise list of the various installations on the site (open pits, underground mines, waste rocks dumps) that may interact with the repository has to be developed.

V-4.2. Development of the site conceptual model and exposure pathways

V-4.2.1. Conceptual model

A conceptual model is a discursive and graphical representation of the situation. It represents a synthesis of the knowledge acquired on the site and its surroundings, and with respect to the environmental vulnerability.

The model helps to understand and to explain the situation in terms of contamination sources and actual as well as potential populations exposures pathways. Figure V-2 shows an example of a model representation using compartments.

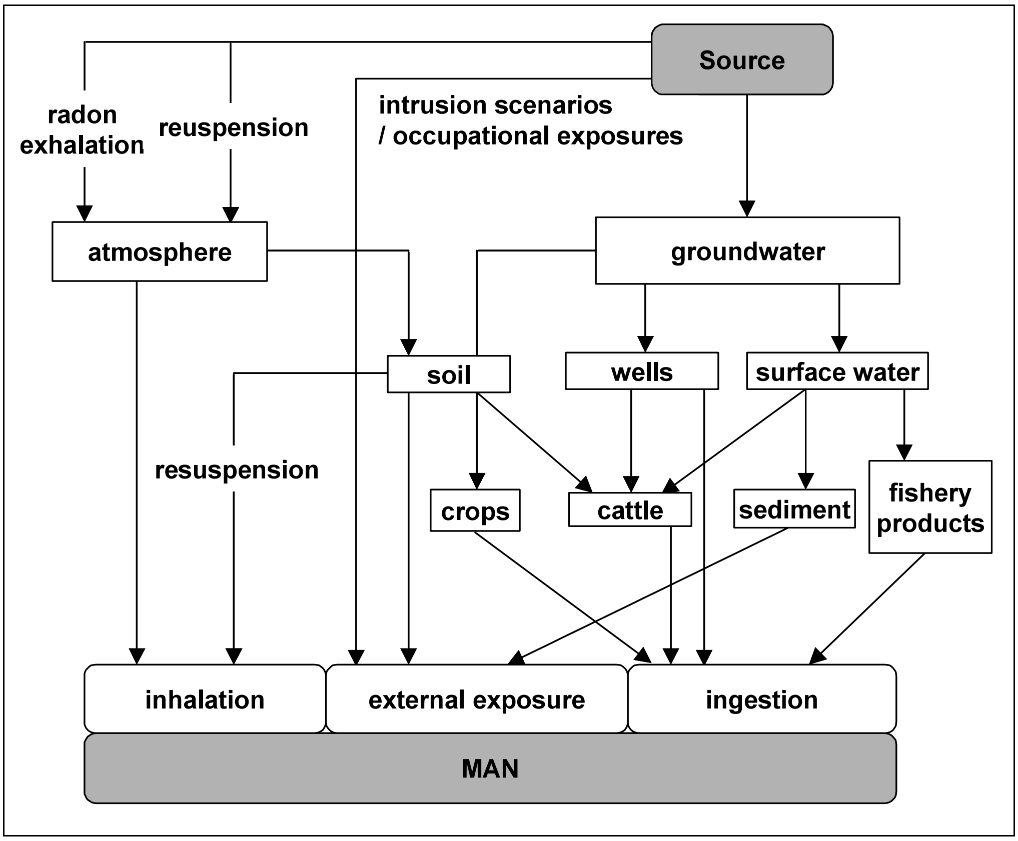

Fig. V-4. Example for the conceptual model.

V-4.2.2. Exposure pathways

External exposure

External exposure comes from particles deposited and from the radiations emanating from the site, radiation emanating directly from the site being generally highly predominant. It can be calculated using a measured average dose rate.

Internal exposure via inhalation

Internal exposure due to inhalation of short-lived daughters of ^{222}Rn and ^{220}Rn is a major pathway. It is calculated from the data on the volume potential alpha energy concentrations of short-lived daughters of ^{222}Rn and ^{220}Rn in the atmosphere. Dust coming from the site can also contribute to the internal exposure via inhalation, which is calculated using the volume activity due to long-lived alpha emitters in the dust in suspended in the atmosphere.

Internal exposure via ingestion

Contaminated water used for irrigation or watering animals can be a major contamination source. Ingestion of foodstuffs contaminated by the use of contaminated water is an internal exposure source. Contamination drinking water should also be taken into account, whether it comes from a well or from the distribution system, even if it may be a very secondary pathway.

V-4.3. Development of population groups and scenarios

V-4.3.1. Location of population groups

The objective is to determinate the geographic location of the inhabited area, residents of which receive the highest impact from the mining area under study. Since the inhabited areas around the site are generally large, it will be necessary to use a rationalised method.

V-4.3.2. Composition of population groups

The chosen groups have to be representative of the exposure of the whole surrounding population. The higher the impact is, the detailed the assessment should be, and the most refined the selection of population groups has to be.

People in a population group can be either adults or children, but in any one group, people should belong to the same age group. There is no methodology to determine *a priori*, i.e. before measurements in the environment and dose calculation are undertaken, which specific age group should be adopted. Actually, even if dose coefficients for the various age groups are well known, the result of the added dose calculation depends on

- activity schedules;
- frequented places;
- consumption of locally produced food;
- radioelements concentrations in the various consumed produces

Therefore, it is not possible to leave out any age group before a complete calculation of the added effective dose has been undertaken and it has been demonstrated that the level of exposure of this group is negligible compared to other groups.

V-4.3.3. Scenario definition

The attempt to be realistic, in accordance with Article 45 of the European Directive 96/29/Euratom, will lead to a determination of scenarios corresponding to real local lifestyles.

In order to perform a realistic assessment, activity schedules have to be as close as possible to the life and habits in the local environment at the site. Data concerning food consumption have to be given and should be accompanied by as many statistical data as possible. The origin of the food-stuffs has to be known in detail, in particular where they are cultivated locally.

When determining the diets for each population group, it is necessary to take into account the age group, the self-sufficiency level, and if possible the professional activity, the place of residence, etc.

V-4.4. Measurements in the environment

Radiological impact assessment immediately after remediation can be based on the monitoring network installed by the operator or by the authorities in the nearby environment. This network also allows to check the evolution of the source term.

The measurements have to be tailored to the exposure pathways that have been identified and to the local lifestyles.

V-4.4.1. Location of measurements and analysis

Outdoor measurements

These measurements concern the mean external exposure dose rate, the Potential Alpha Energy (PAE) of ^{222}Rn and of ^{220}Rn, and long-lived alpha emitters in suspended dust.

Measurements have to be made at the site at all the places, where contaminanted residues are disposed of, and if possible at a place on the site, where there is no disposal of contaminated residues.

In the nearby environment measurement stations should be installed at all locations inhabited by the population groups, and at all locations, where the population groups work and undertake leisure activities.

Measurements inside buildings

These measurements concern the Potential Alpha Energy (PAE) of ^{222}Rn and of ^{222}Rn.

Necessary measurement locations on the site are existing buildings that are or that could be used for other professional activities.

In the nearby environment measuring stations should be installed in some houses of the population groups, in some buildings where the respective people work (farms, schools…) and in some leisure buildings (gymnasium….).

Water

Sampling for analysis has to be carried out in catchment areas for agricultural use, at consumer taps, and if relevant in the swimming and fishing areas.

Food chain

The radionuclides to be analysed include at least: ^{238}U, ^{230Th}, ^{226}Ra, ^{210}Po, and ^{210}Pb.

Sampling has to be done in gardens, on fields, and in greenhouses, from products bought locally, and, if relevant, where e.g. wild fruit and mushrooms are gathered.

The objective is to get at least over a whole study year an averaged data set for the external exposure dose rate every quarter, an average value for PAE of ^{222}Rn and of ^{220}Rn, and monthly values for the activity of long-lived alpha emitters in the suspended dust. For drinking water, monthly concentration value for each radionuclide over the study year, and one concentration value for each radionuclide and each type of food locally produced, should be determined.

V-4.4.2. Presentation of results

Reporting should include:

- the location of the measurement, or of the sampling point, or of the cultivation, if it is a processed food product;
- a description of the measuring station (distance to the mining area, location on the wind rose, local topography, presence or absence of a ‘shield’ (e.g. a hill) between the site and the station, location with regard to the hydrological situation at the site);
- the sampling period;
- the climatic parameters that can influence the result;
- all the parameters that are used to transform the result of measurements into usable concentrations or dose rate (for example water content, sampled air volume).

V-4.5. Background radiological level

‘Natural’ radioactivity that comes from cosmic and telluric radiation.

If there is detailed information on the radiological state of the area before the beginning of mining operations, it can be used as background level. However, often environmental data are not precise enough and cannot be used. Some measurement stations should then be installed in the natural surroundings, in order to get some relevant data not influenced by the site.

Such stations have to be installed in a geological context similar to the geological context of the site, upwind and upstream the mining site, and on drinking water distribution systems different from those used by the studied population groups.

The objective should be to develop for each measurement station at the site a reference station in nearby environment that is representative of the natural surroundings.

V-4.6. Site impact calculation

Impact assessment is based on the determination of the total added effective dose resulting from the exposure of the various identified population groups. It corresponds to the difference between the current effective dose and the effective dose estimated for the initial situation.

V-4.6.1. Dose coefficients

Dose coefficients enable to relate incorporated quantities (through ingestion or inhalation) and effective doses. They depend on the age group. Dose coefficients that have to be used are issued in the European Directive 96/29 Euratom.

V-4.6.2. Sensitivity analysis – uncertainty

Generally a number of parameter are only imprecisely known play an important part in the dose assessment. Therefore it is necessary to estimate the uncertainty in the dose calculations.

The impact studies require an estimation of the uncertainty associated with the dose calculation results. Although a complete uncertainty analysis is not necessary, there must be undertaken a concise uncertainty analysis based on a sensitivity analysis of the parameters that highly influence the calculated doses, in particular those that have the highest uncertainty.

This process is necessary in order to be able to assess the highest possible dose received by population groups.

V-4.7. Determination of reference group(s) and monitoring networks

Population groups that receive the highest impact from a site are considered to be reference groups. Reference groups are monitored afterwards to ensure that the site impact remains acceptable.

If a population group with different lifestyle and habits is noticed afterward (for example after the building of new houses near the site, or a new camp site…), it is necessary to monitor its environment and make a calculation of the added effective dose in order to check if it could be a new reference group.

V-5. APPLICATION TO THE LODÈVE SITE

Lodève is a former uranium mining and milling site where production was stopped in April 1997; site remediation was completed in 2001. A four-person team operates the water treatment station, which processes water from the flooded mines and the uranium mill tailings impoundment, and it conducts environmental monitoring activities. The team also monitors and maintains the former production areas of the site, which will be converted into industrial zones in the near future.

Two methodologies were applied in the assessment of the radiological impact of the Lodève site in 2001 [V-3].

The first one was based on the scenario described in Decree 90-222 and the second one was based on realistic scenarios as described in Table V-3.

Table V-3. Description of the scenarios – time spent in various places [h/y].

Time spent [h/y]	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5	Decree 90-222 scenario
	Adult working far from the site	Retired people	Adult working near the site – agriculture	Adult working near the site – office	Adult working on the site – office	
Inside the house or at the office located near the site	5000	4380	5000	7200	5000	-
Outside near the site	1160	3980	3360	1160	1560	7000
Far from the site	2200	-	-	-	-	1360
Walking on the site	400	400	400	400	400	400
At the office, on the site	-	-	-	-	1800	-

The results (Table V-4) show that:

— the TAETA for the scenario according to Decree 90-222 and for all the locations of population groups is lower than 1;

— the added effective dose calculated for all the realistic scenarios and all the locations of population groups is lower than 1mSv/y.

Table V-4. Radiological impact assessment for the Lodève site for an adult – comparison between methodologies

Location of population groups	**Added effective dose [mSv/y]**					
	TAETA (Decree 90-222)	**(European Directive 96/29)**				
		Scenario 1	**Scenario 2**	**Scenario 3**	**Scenario 4**	**Scenario 5**
Saint-Julien	0.18	0.45	0.68	0.64	0.54	0.90
Saint Martin	0.18	0.46	0.69	0.66	0.55	0.91
Mas Lavayre	0.09	0.31	0.41	0.40	0.35	0.74
Capitoul	0.07	0.28	0.36	0.35	0.31	0.70
Mas Campagnard	0.20	0.41	0.74	0.68	0.45	0.87
Ferme de Treviels	0.20	0.43	0.74	0.69	0.50	0.90
les Hémies	0.09	0.27	0.40	0.38	0.30	0.71

However, TAETA close to 0.2 corresponds to an added effective dose close to 1 mSv/y. Application of the new methodology allows a better understanding of the short-term impact and a better choice of future reference groups. In fact, the added effective doses of adults not working on the site are less than 0.8 mSv/y. Closer attention has to be paid on adults working on the site (e.g. monitoring of the indoor air radon concentration).

V-6. CONCLUSIONS

The new radiological impact assessment methodology for impoundments of uranium mill tailings after remediation allows to demonstrate the ability of the repository to ensure protection for people, taking into account the current legislation.

In addition, through this impact study, reference groups, which are population groups that receive the maximum impact from the site, can be defined. Exposure pathways related to reference groups will be monitored afterwards to ensure that the site impact on man remains acceptable.

Moreover the impact study helps to define a monitoring network that aims to ensure that the impact on the environment remains acceptable.

It is essential to remember that the impact on man has to be re-assessed when population groups with different lifestyles are noticed afterwards (e.g., after the construction of new residences near the site, or a new camp site), or when the monitoring of the environment shows significant changes in the measured parameters values.

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ANNEX VI. GERMANY

DEVELOPMENT OF TECHNOLOGIES FOR IN SITU REMEDIATION OF CONTAMINATED SITES BY DIRECTED FORMATION OF NATURALLY OCCURRING SLIGHTLY SOLUBLE MINERALS

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VI-1. ABSTRACT

A novel technology is described that allowes the *in situ* treatment of areas contaminated by heavy metals or radionuclides, as well as the sealing of porous rock or soil formations. Solutions supersaturated with respect to sparingly soluble minerals (e.g. sulfates, carbonates, hydroxides) are prepared and used as a grout or penetration agent. Precipitation processes take place during the flow of such solutions through the treated soil or rock formation. These lead to an immobilisation of contaminants and a decrease in the permeability of the formation. Promising grouts are gypsum- or $BaSO_4$-forming solutions, as well as systems leading to the formation of $CaCO_3$. The results of the first large scale application of solutions leading to directed $BaSO_4$ formation in the former uranium mine Königstein of Wismut GmbH (Germany) are summarised.

VI-2. INTRODUCTION

Up to now, only a limited number of remediation technologies are known that allow a cost effective *in situ* treatment of soils, tailings or waste rock contaminated by heavy metals. In many cases, pump-and-treat methods are applied. Soil flushing, encapsulation and excavation followed by decontamination or disposal are other techniques used to remediate contaminated areas. All these techniques are extremely expensive and require long times. In some cases, secondary processes are induced, resulting in an additional release of contaminants. On the other hand, many natural processes are known that lead to the formation of sparingly soluble minerals. Such phenomena are typically coupled dissolution/precipitation processes. A well known method to reduce the acidity of tailings is overlaying with alkalinity generating materials such as limestone, dolomite or fly-ash. Dissolution of these materials, for example by rain, leads to alkaline solutions that can neutralise acidic zones. As result, secondary minerals such as iron or aluminium hydroxide and gypsum are formed. They can plug the flow paths and thus deeper zones are protected against oxidation and leaching. Similar reactions occur, when acid mine drainage penetrates into $CaCO_3$ containing soil formations. Dissolution as well as precipitation processes take place. It is well documented that the formation of gypsum and of hydroxides can result in clogging of flow paths [VI-1–VI-3]. Further solution transport is interrupted and infiltration of acid mine drainage is stopped. Although gypsum is characterised by a relatively high solubility, long-time stable sealing has been proven. Natural mineral forming processes require, however, long times and only small amounts of mass are transported. If new ways for *in situ* immobilisation processes to remediate areas polluted by heavy metals or radionuclides are sought, utilisation of processes similar to those occurring in nature is seen as a promising way.

Two different scenarios have to be taken into consideration:

(1) Immobilisation of dissolved contaminants, for instance radionuclides in pore water;
(2) Fixation of soluble or oxidisable contaminants, for instance protection of sulfidic minerals against leaching.

Although in many cases the same chemicals can be used, there are substantial differences between these two tasks. The hydraulic properties of porous formations are the main cause for this. It is rather difficult to achieve a homogenous mixing of two solutions in soils or porous rock formations. Mainly plug flow is induced and mixing takes place only in boundary layers. In the case of grouting, a special arrangement of injection and extraction holes is necessary. Under the second scenario, *inter alia*, the dissolution of heavy metal containing compounds must be prevented, i.e. the source term must be immobilised. Both, a reduction of the permeability of the acid generating domains, and the protection of reactive mineral surfaces against oxidation and leaching are possible ways to reduce the release of contaminants.

In all cases, the main aspect to be considered is, whether long-term immobilisation of the contaminants can be achieved. Particularly, the formation of sparingly soluble minerals is of importance in this context. Depending on the nature and the concentration of the contaminants, the formation of hydroxides, sulphates, carbonates or phosphates can be taken envisaged for the fixation of radionuclides and heavy metals.

First attempts to prevent the generation of acidic rock drainage by covering of reactive mineral surfaces with insoluble phosphates were described by Evangelou and Zhang [VI-4]. The concept of 'Neutral Barrier Technology' involves two procedural steps [VI-5]: First, the formation is permeated with a saturated $Ca(OH)_2$ solution. In the second step, gaseous CO_2 is injected. The calcite precipitates formed plug the flow paths and can also act as a reactive barrier. However, only small quantities of dissolved $Ca(OH)_2$ can be brought into the formation due to the low solubility of lime.

The objetive of the current project that is summarised here is to artificially induce precipitation processes thus reducing the permeability of soils, or immobilising contaminants. Metastable solutions are prepared that lead to the formation of sparingly soluble minerals, such as sulfates, carbonates or hydroxides.

VI-3. Possibilities to prepare mineral forming solutions

The utilisation of crystallisation/precipitation processes similar to those occurring naturally is only possible, if solutions can be prepared that lead to a directed formation of sparingly soluble minerals. This means, that the solutions have to contain in dissolved form the components of those minerals that are intended to be formed. Mineral formation then should take place during the passage of the solution through the formation that is being treated. There are different ways to prepare such solutions. Depending on the properties of the intended minerals, their respective solubility, and the quantities of solids required, the following options are possible [VI-6,VI-7]:

- Preparation of supersaturated solutions by using precipitation inhibitors;
- Preparation of supersaturated solutions by addition of compounds leading to a temporarily enhanced solubility;
- Construction of reactive systems. Solutions are prepared that contain components which release ions that then form insoluble minerals (for example carbonates) or that cause a change in the pH value.

By using precipitation inhibitors, it is possible to prepare solutions leading to $BaSO_4$, $CaSO_4 \times 2H_2O$ or $CaCO_3$ formation. The stability of the supersaturated solutions depends on the following factors:

- Composition and concentration of the inhibitor;
- Degree of supersaturation;
- pH and temperature;
- overall solution composition.

Without inhibitor:

$CaCl_2 + MgSO_4 + 2\,H_2O \rightarrow CaSO_4 \times 2H_2O$

$Ca(OH)_2 + H_2SO_{4(aq.)} + 2\,H_2O \rightarrow CaSO_4 \times 2H_2O$

fast, uncontrolled precipitation

In the presence of precipitation inhibitors:

$CaCl_2 + MgSO_4 \rightarrow Ca^{2+} + SO_4^{2-} + Mg^{2+} + 2\,Cl^-$

$Ca(OH)_2 + H_2SO_{4(aq.)} \rightarrow Ca^{2+} + SO_4^{2-} + 2\,H_2O$

temporarily stable solution

$\downarrow$

$CaSO_4 \times 2H_2O$

formation of layers, single crystals or sludges depending on the solution composition

Fig. VI-36. Fundamentals for the preparation of gypsum forming solutions.

Gypsum, a mineral occurring in many rocks, has a solubility in water of 2.5 g/l at 25°C. Gypsum precipitation occurs at once when, for instance, a solution containing $MgSO_4$ is mixed with a solution containing $CaCl_2$ and the resulting concentration exceeds the solubility limit. It is a fast reaction, leading to a fine-grained crystallisate of gypsum needles. However, if the mixing is carried out in the presence of small amounts of a precipitation inhibitor, clear $CaSO_4$ supersaturated solutions result (Fig. VI-1). The stability of the solution depends on the inhibitor used (Fig. VI-2). In general, increasing the inhibitor concentrations produces $CaSO_4$ supersaturated solutions with increased stability. At constant inhibitor concentrations, increasing $CaSO_4$ concentrations lead to faster crystallisation. Another way to prepare solutions supersaturated with respect to gypsum is to mix $Ca(OH)_2$ suspensions with dilute sulfuric acid. It is important that a precipitation inhibitor is present that is active in both, alkaline and acidic solutions. Mixing in a special mixing unit allows the preparation of a solution containing only Ca^{2+} and SO_4^{2-} ions. The stoichiometric mixture has an acidic pH. To obtain neutral solutions, it is necessary to add small amounts of dilute NaOH or KOH solutions. It is possible to prepare solutions from which up to 40 g/l $CaSO_4 \times 2H_2O$ can be precipitated. In the case of mixing alkaline solutions supersaturated with respect to $CaSO_4$ with solutions containing heavy metals or radionuclides, hydroxide precipitation as well as gypsum crystallisation occurs. Additionally, the inclusion of contaminants into the gypsum crystals formed has been observed. Different immobilisation processes can be induced by the selection of specific precipitation inhibitors, as well as by the addition of special precipitation agents.

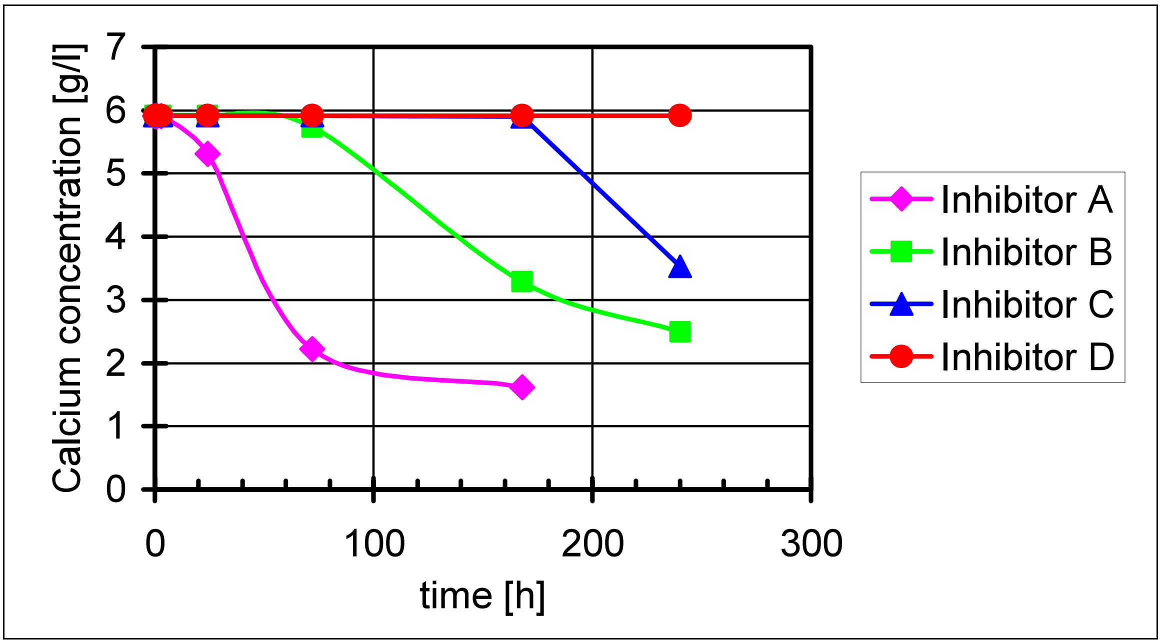

Fig. VI-37. Course of gypsum crystallisation from oversaturated solutions depending on the inhibitor concentration

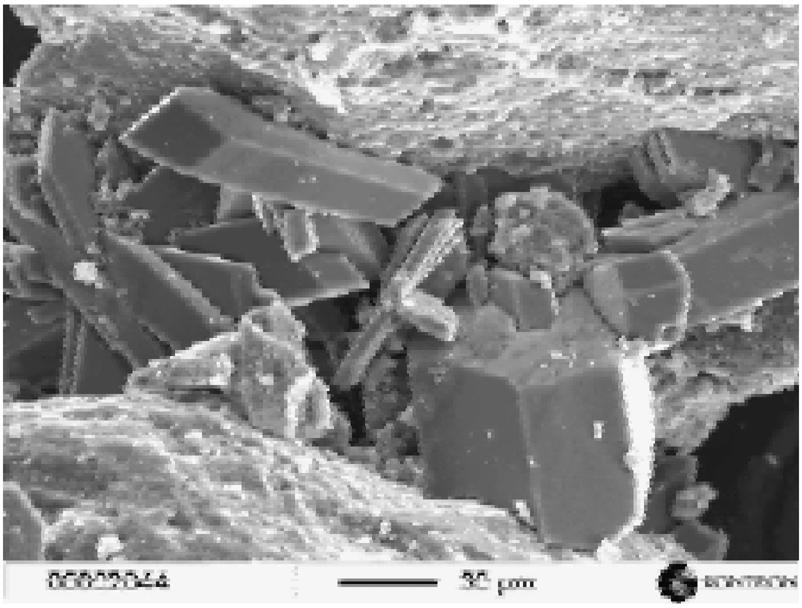

Fig. VI-38. Gypsum crystals formed in porous sandstone

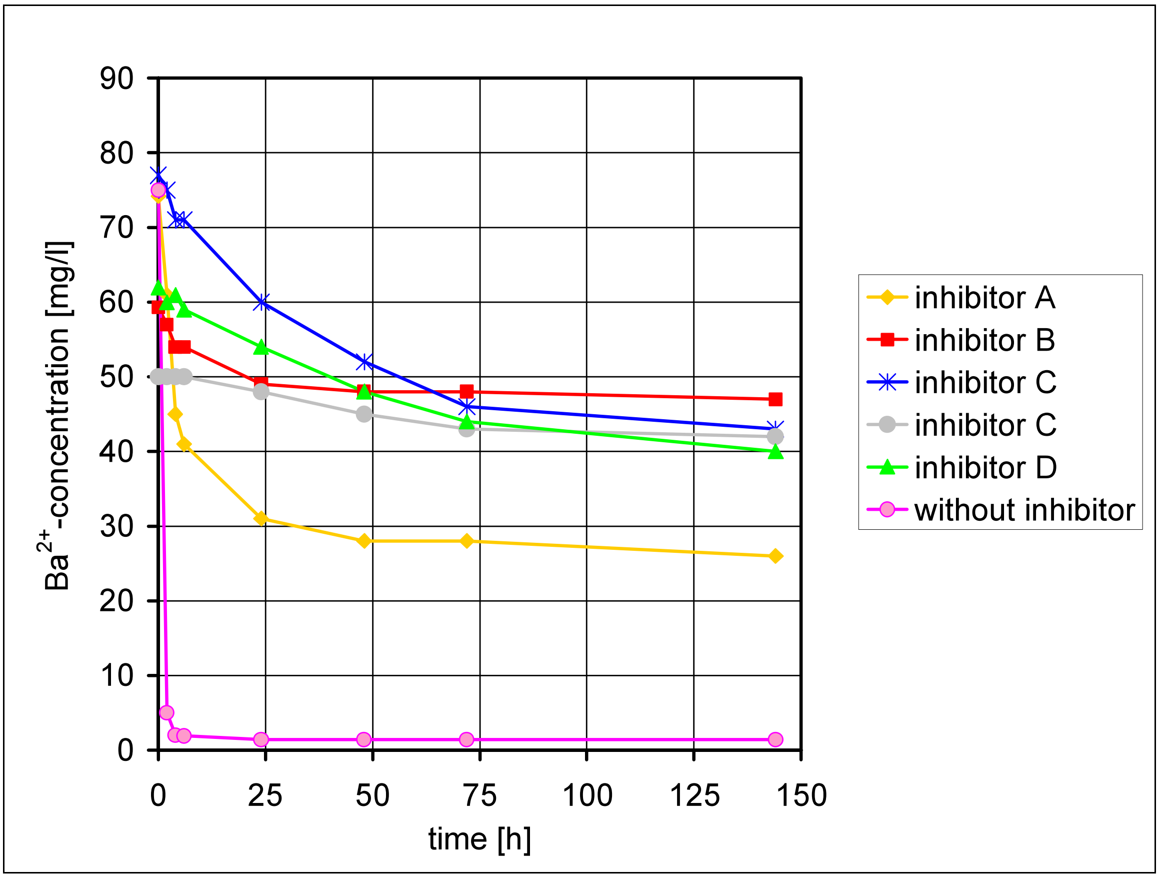

Fig. VI-39. Course of $BaSO_4$ precipitation from oversaturated solutions (inhibitor concentration: 0.8 mg/l)

Fig. VI-40. $BaSO_4$ layers formed on sandstone.

These grouting solutions can penetrate even into microfractures and very small pores. Already single gypsum crystals can cause a significant permeability reduction. They block flow paths and make further solution transport impossible. Solutions with a high stability can be used for treating areas far away from the point of injection. This is especially useful for achieving a large radius of injection and for ensuring a long life time of the injection point. Gypsum crystallisation leads to a step by step closure of flow paths in a manner similar to naturally occurring cementation processes.

In many cases a single injection will not be sufficient to achieve a closure of flow paths in a given case. Repeated injections may be necessary, leading to a step-by-step closure of flow paths, as precipitates form on the walls of the pore spaces. Such stepwise closure of open pores ensures a better depth penetration of the grouting solutions than could be achieved when pores would be closed completely at once. Figure VI-3 shows gypsum crystals grown within a porous sandstone. The crystals are strongly connected with the surrounding rock. Due to a blocking of the flow paths, further solution transport becomes impossible.

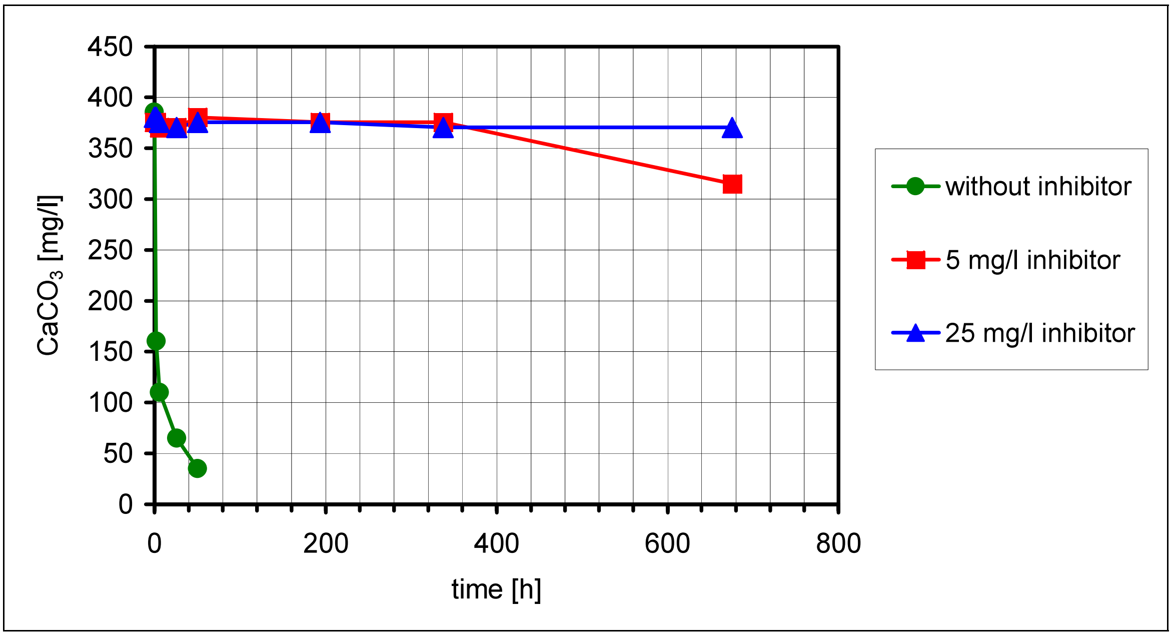

Fig. VI-41. Course of $CaCO_3$ precipitation from oversaturated solutions

$BaSO_{4(s)}$ forming solutions can contain up to 500…600 mg $BaSO_4$/l in dissolved form. This seems to be low in comparison with gypsum producing solutions, but one has to remember that the natural solubility of $BaSO_4$ is only 2.5 mg/l. This means, however, that solutions with concentrations more than 200 times above the natural $BaSO_4$-solubility can be prepared. Normally, mixing of Ba^{2+} and SO_4^{2-} containing solutions leads to a spontaneous precipitation of $BaSO_4$ (Fig. VI-4). Very fine crystals are formed that settle slowly. Due to the extremely low solubility product of 10^{-10} mol^2/l, even traces of Ba^{2+} ions are precipitated in the presence of SO_4^{2-} ions. This reaction is used in analytical chemistry as a highly accurate method for SO_4^{2-} determination. Mixing of $BaCl_2$ and sulfate ions containing solutions in the presence of an inhibitor allows to prepare clear, $BaSO_4$-supersaturated solutions. A favoured way to prepare 'pure' $BaSO_4$ solutions is to use $Ba(OH)_2$ solutions and dilute H_2SO_4 as starting components. Apart from the inhibitor the resulting grout contains only Ba^{2+} and SO_4^{2-}- ions, and no further components that are brought into the process have to be taken care of. A third way to prepare $BaSO_4$ forming solutions employs Na_2SO_3. The sulfite ion can be converted into sulphate by oxygen and thus, other oxidation processes are stopped. As a result, reducing conditions are created. The action of the $BaSO_4$ grouts is similar to that of $CaSO_4$ solutions. Decomposition of the inhibitor results in the formation of insoluble $BaSO_4$ precipitates. These precipitates form a protective layer on surfaces, including reactive minerals, and further dissolution is prevented (Fig. VI-5). An immobilisation of soluble contaminants is achieved by fixation in a mineral with a high stability under most naturally occurring conditions. It is possible to control the speed of the crystallisation process through the composition of the inhibitor and its concentration, or by the absolute degree of supersaturation.

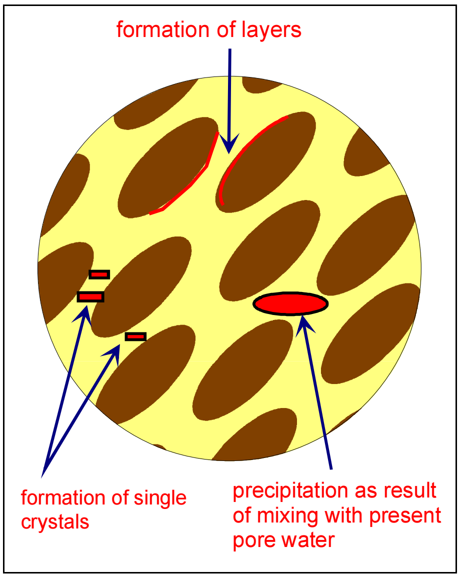

Fig. VI-42. Action of mineral forming solutions.

Fig. VI-43. Column experiments.

In a similar way it is possible to prepare $CaCO_3$ forming solutions by mixing of Na_2CO_3 and $CaCl_2$ solutions (Fig. VI-6). These are able, as discussed already for $BaSO_4$ and gypsum forming solutions, to form layers protecting reactive surface, to block flow paths or to immobilise dissolved contaminants (Fig. VI-7).

VI-4. Immobilisation of heavy metals by using $BaSO_4$-forming solutions

VI-4.1. Results of laboratory investigations

Column experiments with both, freshwater and $BaSO_4$-forming solutions, were used to investigate the differences between flushing and immobilisation. The columns used had a length of 1000 mm and a diameter of 300 mm, and were filled with approximately 60 kg of rock material from the Königstein mine (Fig. VI-8). A volume of approximately 23 l was required to fill the columns to the top. The solutions or water, respectively, were pumped from the bottom up at an average rate of 1.2 l/d. After filling, the columns were flushed continuously by water or immobilisation solution. After certain times the flow was stopped and the columns were emptied completely. The material remained in contact with air for up to half a year. After that, washing with water was started again. The immobilisation tests were carried out with a solution resulting in the formation of 350 mg/l $BaSO_4$. At an inhibitor concentration of 80 mg/l the stability was in excess of 96 hours. In order to increase the immobilisation capacity, small amounts of sodium silicate were added. Technical grade $Ba(OH)_2{\times}8H_2O$, Na_2SO_3, and sodium silicate solution were used. The feed solution was prepared in batches step by step by mixing solutions of the separate constituents. An alkaline and reducing solution was obtained. In the case of mixing with acidic, heavy metal containing solutions precipitates of slightly soluble hydroxides or hydroxysulphates were formed, depending on the mixing ratio.

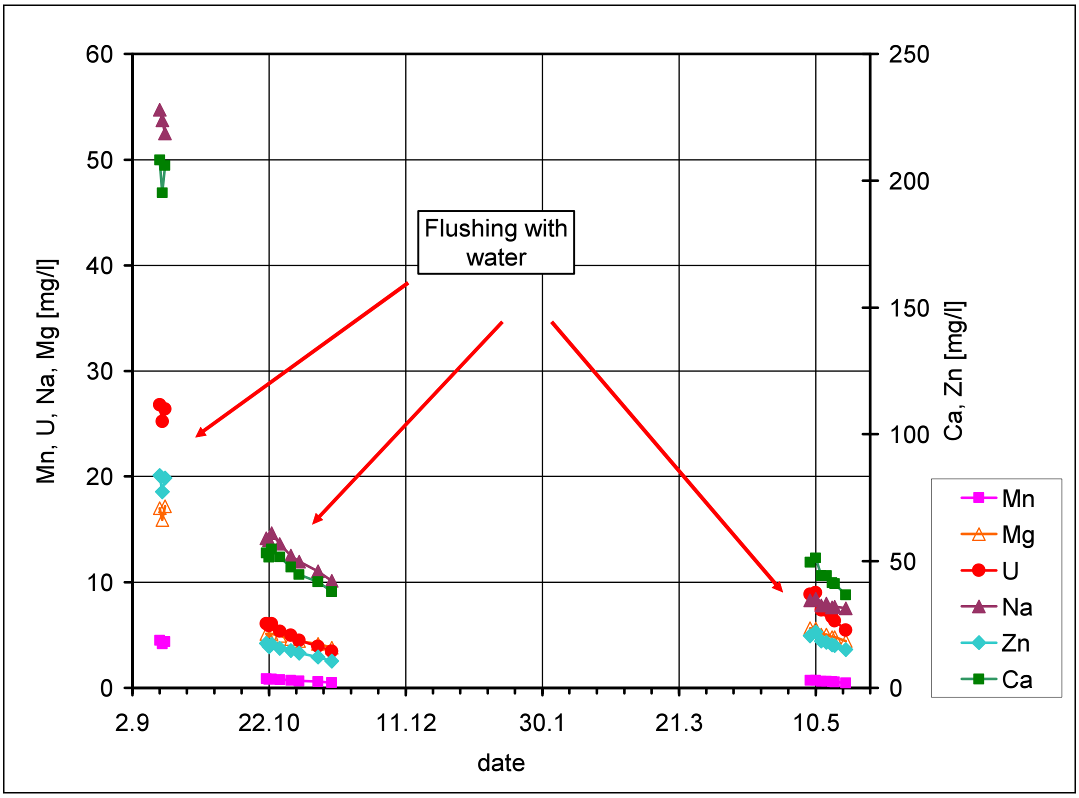

Fig. VI-44. Development of effluent concentrations during flushing of the column with water (the flushing was interrupted after 5 days, and continued after 45 days and 6 months respectively).

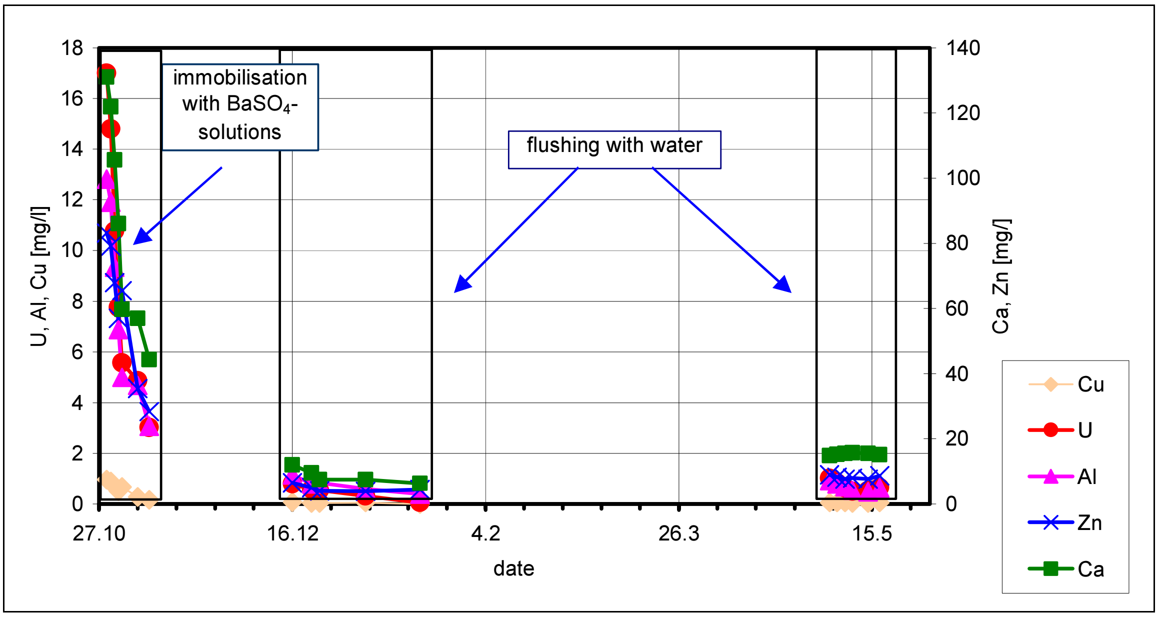

Fig. VI-45. Development of effluent concentrations during and after immobilisation with $BaSO_4$-forming solutions.

Figure VI-9 shows the concentrations of selected constituents during the flushing of the column with water. It is clear that concentrations decrease only slowly with time. Long flush times, or high volumes of water, are required to reduce the concentrations of metals in the effluent. Contact with air induces oxidation processes. As result, the second or third washing step also produces solutions with high contaminant concentrations. In contrast, the application of $BaSO_4$-forming solutions leads to effluent solutions with rapidly decreasing concentrations (Fig. VI-10). Four weeks after permeation with $BaSO_4$-forming solutions, the column was also flushed with water. Effluent concentrations remained at a low level. Obviously, oxidation processes were prevented due to the formation of insoluble layers consisting of $BaSO_4$, hydroxides and other secondary minerals. The same was observed during a second flushing with water, five months later.

VI-4.2. Results of field tests in the former uranium mine Königstein of Wismut GmbH

The application of solutions supersaturated with respect to $BaSO_4$ was tested in the former uranium mine 'Königstein' of Wismut GmbH (Germany). In this mine, uranium was recovered *inter alia* by *in situ* leaching of the sandstone host rock with dilute H_2SO_4. A detailed description of the mining operations carried out at Königstein is given in [VI-8]. The mine borders on to the National Park Saxon Switzerland and the distance to the river Elbe is 600 m only. The ore body is located in the so-called 4th sandstone aquifer, the deepest of four hydraulically separated aquifers in the cretaceous basin. The 4th an 3rd aquifer are separated by a 10 to 30 m thick aquitard that is perforated by natural faults and man-made connectivities due to the mining activities. The 3rd aquifer is an important groundwater resource for the region and of high environmental and economical significance.

Uranium production started in the 1960s employing conventional underground mining. Due to decreasing ore contents, a step by step switch over to specially developed underground *in situ* leaching technologies was carried out. One leaching technology used is based on 'hydrostatic' or infiltration leaching of blasted low-permeability rock in hydraulically isolated blocks. These blocks had volumes in the range of 100,000 to 1,000,000 m^3. In order to construct such blocks, sections of the ore containing horizon were blasted and then separated by dams from the other mine area. The blocks were leached with a solution of 2 to 3 g/l H_2SO_4. During the *in situ* leaching period, altogether 130,000 t of sulphuric acid were brought into the mine. Additionally, an unknown amount of sulphuric acid due to pyrite oxidation was released within the mine. All this has resulted in a substantial change of the geochemical status of the deposit. A high level of mobile contaminants is present in the mine, consistion of mainly sulphate, heavy metals, and natural radionuclides.

Uranium mining was stopped in 1990. Since that time, the closure of the mine was prepared. A step-wise, controlled flooding will be carried out over the next few years. In order to prevent the spreading of contaminants, and in particular the contamination of the 3rd aquifer due to rising water levels, a control and collection system for the flooding waters has been constructed. The collected water is pumped to the surface and treated in a dedicated water treatment plant. Due to the high content of mobile and leachable contaminants, the occurrence of contaminated water is expected for long times. From an ecological, as well as an economical point of view, it is very important to reduce the time by which the applicable water standards can be met, and to minimise the amount of discharged contaminants. *In situ* immobilisation by applying $BaSO_4$ forming solutions was investigated as one possible alternative for reducing the total amount of contaminants released, as well as limiting the time over which this occurs.

A ‘block’ situated in the southern part of the mine was selected for the field test. The block was leached in the 1980s, after that, no further treatment was carried out. It was shown that it is possible to prepare without problems large quantities of $BaSO_4$ forming solutions under conditions typical for mining. Over a period of six days a total of 3,540 m^3 $BaSO_4$ forming solution was brought into the block. The average solution composition was 115 mg/l SO_3^{2-} and 200 mg/l Ba^{2+}. 300 mg SiO_2 were added as sodium silicate solution. Approximately 1,200 kg $BaSO_4$ could be formed. The solution was characterised by an extremely high stability. In the absence of suspended solids, no precipitation occurred within 120 hours.

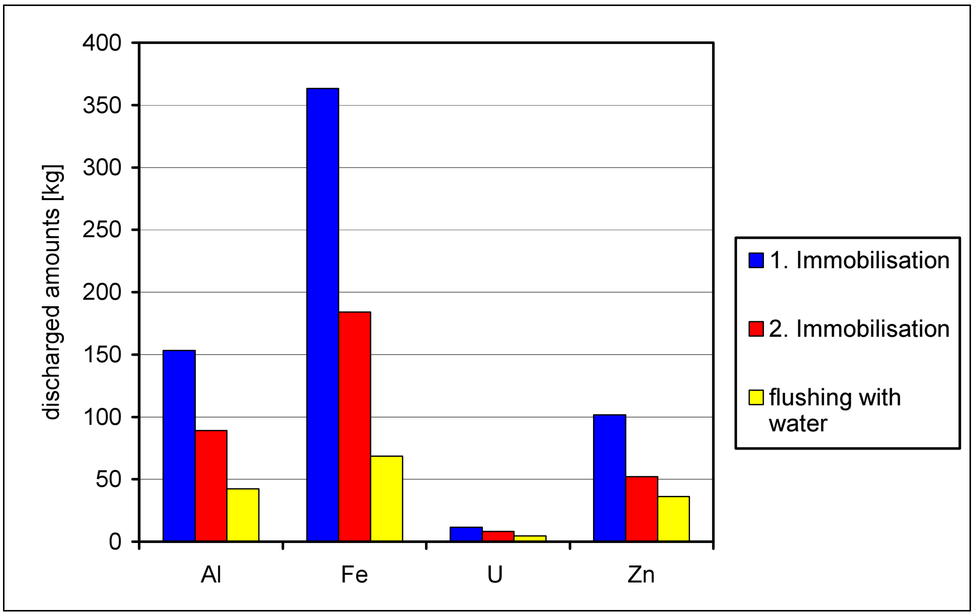

Fig. VI-46. Total amount of contaminants discharged during the various stages of the field test.

The solution remained for ten days in the block. After that time, the solution drained with a initial volume flow of 20 m^3/h. In a second step, the block was filled again with the $BaSO_4$ forming solution. After draining once more, the block was flushed with freshwater in order to demonstrate the stability of the neoformed secondary minerals.

The amounts of contaminants discharged during the different treatment periods are summarised in Figure VI-11. A step by step reduction is visible. The first immobilisation has resulted in solutions containing a total of 11.6 kg uranium, 363 kg iron and 101 kg zinc. After the second treatment with $BaSO_4$ forming solutions, a total output of only 8 kg U, 184 kg Fe and 52 kg Zn was observed. The flushing of the treated block with freshwater did not lead to an increased discharge of contaminants. To the contrary, a further reduction of the total load was observed. This demonstrates the stability against leaching of the immobilisates. Otherwise a drastic increase should have been observed.

To assess the effectiveness of the immobilisation, it is necessary to compare the discharged amounts of contaminants with those that would be expected when washing the test field with water. The results of a flushing experiment on a block located close to the test site were the basis for such calculations. During this experiment, the block was filled with water and emptied several times. Based on the results obtained, it was possible to predict the expected development of contaminant concentrations in the case of flushing the test field with water. Figure VI-12 shows the development the U concentrations when flushed with water and after

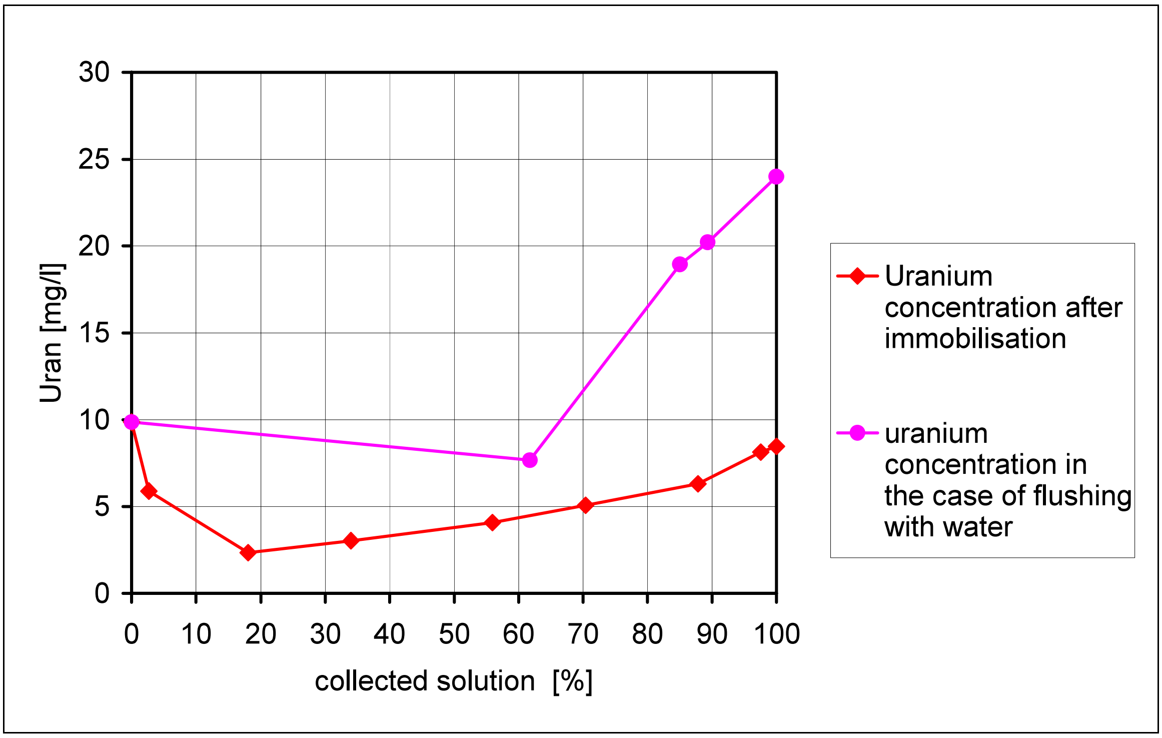

Fig. VI-47. Comparison of the change in effluent uranium concentrations after immobilisation and for flushing with water only.

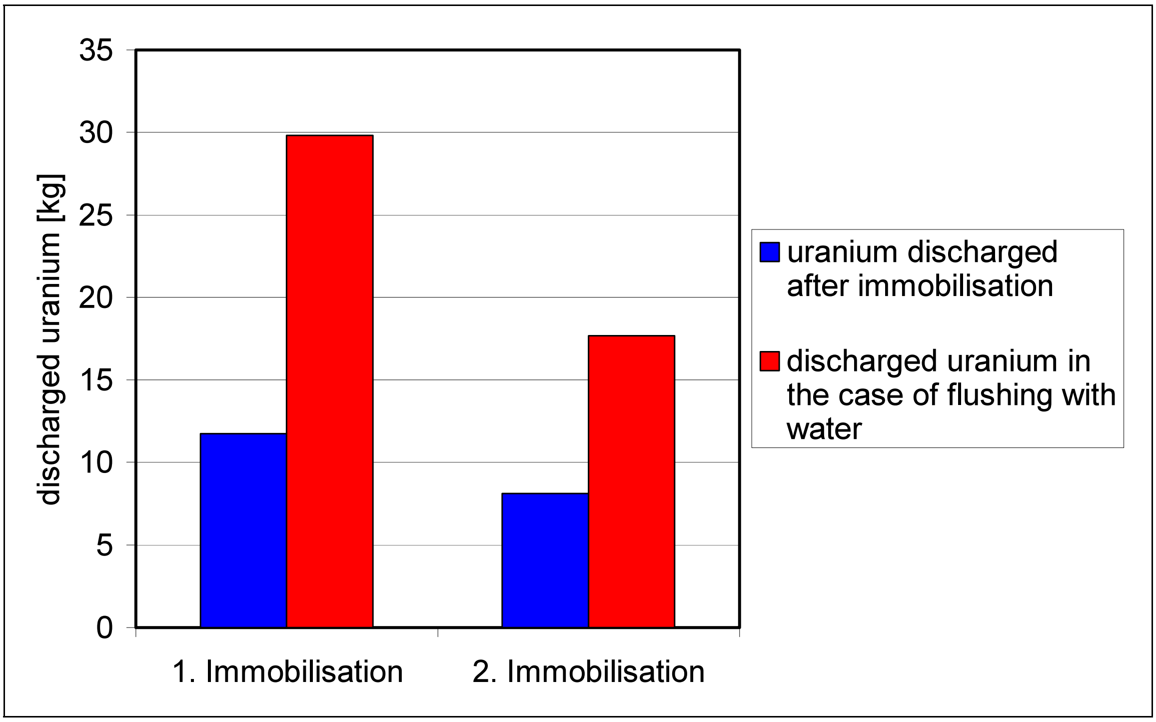

Fig. VI-48. Comparison of the amounts of discharged uranium in the case of immobilisation and for flushing with water only.

Fig. VI-49. View of the grout plant used for the preparation of the $BaSO_4$ forming solution.

the first immobilisation with $BaSO_4$ solution. The main difference is that theimmobilisation has resulted in a significant drop of the concentrations, whereas a washing would produce solutions with slowly decreasing concentrations only. The effect of the immobilisation is visible not only in the development of the concentrations, but also in the total amounts of discharged contaminants. For both immobilisation stages the discharged amounts of contaminants are between 50 and 70% lower than would be expected in the case of flushing the block with water (Fig. VI-13).

VI-4.3. Large scale application

Based on the results from these field tests, the decision was taken to apply this newly developed technology to selected areas of the Königstein mine. Approximately 100,000 m^3 of $BaSO_4$ forming solutions were prepared and injected between December 2001 and May 2002 into three different blocks. The blocks were prepared for uranium leaching before 1990, but the operation was suspended when uranium mining was terminated. As the rock had been in contact with air and moisture for more than 10 years, flooding would cause the formation of highly concentrated acidic solutions. Treatment with $BaSO_4$ forming solutions was aimed at inhibiting oxidation processes and reducing contaminant discharge during the flooding process at a later stage. As in the test fields, the solution was based on $Ba(OH)_2$, Na_2SO_3, water glass, and a precipitation inhibitor. Figure VI-14 gives an impression of the grouting plant used. $Ba(OH)_2$ solutions were mixed within a pipe with water and an the inhibitor-sodium-sulfite-sodium-silicate solution. In this way, up to 50 m^3/h of supersaturated solution could be prepared. Samples taken from different locations in the blocks demonstrated the successful use of $BaSO_4$ forming solutions.

VI-5. SUMMARY AND CONCLUSION

The use of mineral forming solutions offers many possibilities to immobilise contaminants in permeable formations. Both, the protection of reactive mineral surfaces against oxidation or leaching, and the immobilisation of dissolved contaminants is feasible.

The application of $BaSO_4$-silica forming solutions is regarded as a promising approach to immobilise heavy metals *in situ* within porous formations, for example in former *in situ* leaching areas. $BaSO_4$ and silica layers covering reactive mineral surfaces, as well as secondary precipitates, such as hydroxides or hydroxysulfates are formed. Because of the extremely low solubility of barytes, long-term stable immobilisation is achieved. Trouble-free preparation of $BaSO_4$ forming solutions is feasible under field conditions [VI-9,VI-10].

The technology of sealing and immobilisation by controlled crystallisation processes can find many applications, both for the remediation of contaminated areas and for solution of geotechnical problems. The technology combines many advantages such as:

— Sealing and immobilisation is achieved exclusively by naturally occurring minerals. The immobilisation process typically is coupled with a permeability reduction;
— There are many options to direct the precipitation of the minerals and thus to control sealing and immobilisation;
— Different technologies such as spraying, penetration grouting, pressure grouting, infiltration from ponds or reservoirs, can be used to transport the mineral forming solutions into the area that is to be treated;
— All grouts or penetration agents are pure solutions with a viscosity similar or equal to natural groundwaters. A good penetration or infiltration even of soils with a low permeability is obtained;
— The treatment with mineral-forming solutions can be combined with other remediation technologies, such as soil flushing or pump and treat processes;
— It is possible to construct reactive barriers within polluted areas by an *in situ* technique.

The applicability of directed crystallisation processes to immobilise contaminants in tailings depends on the characteristics of the tailings itself. In all cases in which an exchange of the porewater is possible, the use of supersaturated solutions offers the possibility to fix soluble components and to overlay reactive surfaces with insoluble minerals of a high stability. If the consistency of the tailings is so that grouting or infiltration, for example from trenches, is impossible, crystallisation processes can be used to encapsulate the tailings. In other words, the migration of heavy metal containing plumes can be prevented by sealing of potential flow paths.

Possible applications of the technology developed might include the remediation of rock formations treated by acidic as well as alkaline *in situ* leaching techniques and inhibition or reduction of acid rock or mine drainage. The technology is environmentally friendly and is easily adaptable to local conditions.

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ANNEX VII. KAZAKHSTAN

DEVELOPMENT OF METHOD OF COVERING RAISING DUST BEACHES OF RADIOACTIVE WASTES STORAGE OUT OF OPERATION

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VII-1. ABSTRACT

“Ulba Metallurgical Plant’ PJSC (Ulba) is an enterprise with more than 50 years of history of manufacture of different products involved in the nuclear fuel cycle. These activities entailed the construction of various tailings pond for the storage or disposal of processing sludges.

The design of such ponds usually assumes a balance between tailings input and water evaporation so that the material remains moist at all times during the operational period. However, after a significant reduction of the production volumes at Ulba a lowering of the free water level in the waste storage pond occurred. The extent of the lowering of the water level is in the order of metres and the area of ‘beaches’ formed as a consequence is in the order of hectares. The drying out of these beaches gives rise to contaminant dispersal due to wind erosion.

This project investigated various methods for stabilising the beaches, thus protecting against wind erosion, preventing the infiltration of atmospheric precipitation, and providing a certain mechanical durability. Materials investigated for forming covering or cementing layers were water glass (Na-silicate), clays, cement, magnesium carbonate, magnesium oxide, combinations of these components, as well as organic polymers. A particular problem was the applicability on geomechanically very unstable, semi-liquid surfaces.

Also discussed is the need for and feasibility of relocating the tailings.

VII-2. INTRODUCTION

The ‘Ulba Metallurgical Plant’ PJSC (Ulba) is an enterprise with more than 50 years of history of manufacture of different products involved in the nuclear fuel cycle. At different times there were products of thorium oxide, natural U_3O_8, enriched uranium dioxide fuel for VVER and RBMK reactors of Russian design, and nuclear fuel from reprocessed uranium. In addition, for a long time Ulba has been producing beryllium metal based materials and products. At present, Ulba is a single facility with the complete cycle of beryllium production from ore concentrates to finished products of beryllium and its alloys and compounds.

All these production processes were designed in such a way that during the long period of their operation liquid waste has been collected in special storage ponds. Figure VII-1 shows the layout of Ulba’s disposal pond system.

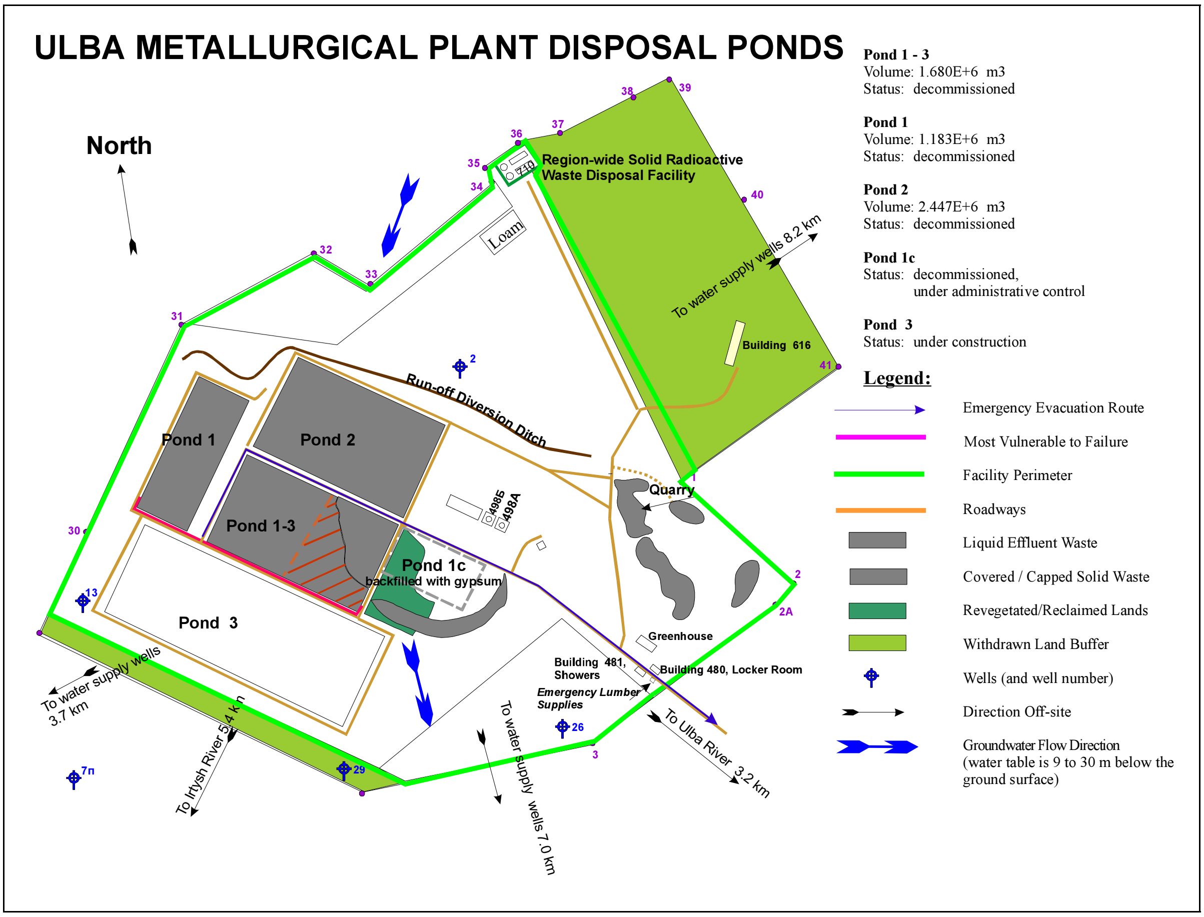

Fig. VII-50. Ulba Metallurgical Plant disposal ponds

Fig. VII-51. Pond No. 1

Fig. VII-52. Beach formation at Ponds No. 1-3.

Currently, two waste storage ponds, pond No.1 and pond No.2, are in use. Pond No.1c was taken out of service and re-cultivated in the 1970s by covering it with a thick layer of clay. At present, pond No.1-3 is not used due to the malfunctioning of its bottom liner. Pond No.3 is a new waste storage pond that is under construction. The depth to the groundwater underneath the pond is 9-30 m. The distance from the pond to the town water supply wells is 3.7 – 7.0 km, that to Irtysh river 5.4 km and that to Ulba river 3.2 km. The Figure VII-1 also shows the groundwater monitoring wells.

Figure VII-2 is an image of the currently used pond No. 1. As can be seen, the water balance is intact. The balance between waters discharged into the pond together with the waste, atmospheric precipitation input, and evaporation rate at the pond surface are monitored regularly. The calucalated balance indicates that no water losses from the tailings pond into underlying aquifer occurs.

After the drastic drop of production outputs due to the end of the cold war and disintegration of the Soviet Union in th 1990s, the quantity of liquid waste disposed into the ponds was reduced drastically. It resulted in a draw-down of water level in one of the ponds and formation of beaches from the production waste slurries. It has been noted that the water level dropped by about 5 metres resulting in the formation of beaches with an area of about 5 ha. Fig. 3 shows images of the pond where beaches have been formed.

The composition of the beach material is very complex. The material contains radioactive particles, toxic compounds of beryllium, as well as thorium and uranium. There is an imminent danger of atmospheric contamination due to the wind erosion of the beach material. The increase of beryllium concentrations and volume activity in the air in the pond area in the periods of higher winds was repeatedly noted.

Various ways of suppressing windblown erosion and dust generation were investigated by cementing or coating the beaches. This problem of forming coatings at the beaches is both, very critical and urgent, and at the same time very complicated. The main problem is to develop a process of coating that can be deployed on these instable and gel-like materials.

A multiple layer cover was envisaged that would be deployed in a stepwise fashion.

VII-3. RESULTS OF THE WORK

VII-3.1. The impounded material

Development of suitable cover compositions has started with studies of the chemical composittion of the ‘beach’ sediments, i.e. the impounded material. Four representative samples of the sediment from different locations on the ‘beaches’ were taken. A special metal tube with a spiral drill was used for sampling of the sediment profile to 1 m depth. The bulk chemical composition of the dry sample is given in Table VII-1.

Table VII-15. Bulk chemical composition of the dry 'beach' sediments

Sample No.	Composition of dry sample [%]							Water content [%]
	Ca	Mg	Al	Fe	Si	SO_4^{2-}	Be	
1	16.0	20.8	5.0	1.0	8.4	21.3	0.01	28.9
2	15.7	14.8	6.1	1.6	9.7	21.2	0.01	13.3
3	12.1	8.07	1.4	0.67	21.2	28.35	0.2	42.1
4	25.2	14.6	0.5	1.7	3.3	20.1	0.2	50.1

The chemical analyses indicated a quite high inhomogeneity of the ‘beach’ sediments, but this is not surprising considering the wide variety of materials disposed off during the principal production phases of the facility. This inhomogeneity means that it will be hardly possible to use the sediments as a component of any layer of the cover. If the cover material were developed on the basis of a sample taken at one location of the ‘beach’, one may obtain quite unacceptable results for other parts of the ‘beaches’ where the sediment composition does not correspond to the initial sample. As a first stage, a solidifying compound for the surface layer or the sediment was developed. Different binding and filling materials were tested, such as water glass (Na-silicate), clays, Portland cement, magnesium carbonate, and magnesium oxide. Tests were performed by placing a layer of the material to be tested on a layer of the sediment taken from a ‘beach’ of the storage pond. The depth of penetration of the test material into the sediment was estimated visually together with the appearance of the surface layer after drying. Experimental results are given in Table VII-2.

Table VII-16. Experimental results of solidifying liquid process waste sediments

No.	Cover material to be tested	Penetration into the surface layer	Surface layer appearance after drying
1	Clay water suspension (Solid:Liquid= 1:1)	No	Clay layer is cracking irrespective of thickness (0.5-1.5 cm)
2	MgO water suspension (S:L= 1:1)	No	Strong setting of MgO layer free of cracks
3	Water suspension of a clay-sand mixture (1 part clay + 2 part sand + 1 part water)	No	No cracks at layer thickness 2 cm
4	Water suspension of fine ground $MgCO_3$ (S:L= 2:1)	No	No cracks in $MgCO_3$ layer
5	Water glass ($Na_2O \times mSiO_2$, m = 2.7) density = 1.15g cm^{-3}	Complete	Strong layer
6	Water glass ($Na_2O \times mSiO_2$, m = 2.7) density = 1.39g cm^{-3}	Complete	Smooth and strong layer
7	Suspension of clay in water glass (m= 2.7, density =1.39 g cm^{-3}) S:L= 1:1	Partial	Smooth, strong and shiny layer
8	Suspension of fine ground $MgCO_3$ in water glass (density =1.39 g cm^{-3}) S:L= 1:1.5	No	Smooth and strong layer
9	Suspension of MgO in water glass (density = 1.39 g cm^{-3}) S:L= 1:1.5	No	Smooth and strong layer
10	Cement water suspension S:L= 1:1	No	Smooth and strong layer
11	Cement water suspension with sand (1 part cement + 1-3 parts sand + 1 parts water)	No	Strong layer irrespective of sand amount

A wide spectrum of easily accessible and cheap materials were chosen for initial testing. The clay is extracted from a pit near the ponds. Sand is cheap in the region around Ulba. A $MgCO_3$ deposit is easily accessible to Ulba and experience of producing active MgO from $MgCO_3$ is available. Water glass is also produced by Ulba in small amounts. The mixes on the basis of Portland cement usually have a high durability and stability. Thus, a wide variety of materials with different properties were captured and subject to testing for use as solidifying agents.

Clay water suspensions, a suspension of a clay-sand mixture, and the suspension of clay in water glass solutions were investigated. The results in Table VII-2 show that clay based suspensions do not form a sufficiently strong cover.

Fig. VII-53. Geotechnical failure and beach destruction after clay emplacement.

Rather strong coatings were produced when using suspensions of magnesium oxidein water. Fine ground magnesium carbonate, however, does not produce sufficiently strong of coatings. MgO has very high chemical activity caused by the very small particles size (less than 2 microns) and the ability to react with silicate forming strong compounds. $MgCO_3$ has no such ability and, hence, can not be used as binder.

Quite good results were obtained at this stage with water glass based coatings. Almost all experiments with water glass produced a rather high coating strengths, irrespective of any filler (magnesium oxide, magnesium carbonate, 'beach' sediment) used. Once exception was clay that resulted in a reduction of the coatingsstrength when used as filler.

As one would expect, a high strength of the coating was obtained with cement based samples.

Those samples that had a strong coating formed in the above experiments underwent testing for weathering resistance. The testing procedure consisted of immersing samples in cold water for 7 hours, drying at 40 to 60°C for 17 hours, repeating the immersion in cold water for 7 hours, after which they were placed in a freezing chamber for 17 hours at -10°C. [VII-1]

The experimental results showed that samples prepared on the basis of a cement-water suspension, suspensions of a cement-sand mixture, and samples treated with water glass of 1.39 g/cm^3 density remained strong and weather resistant. Other samples disaggregated either while immersed in water or in the freezing chamber (Tab. VII-3).

Table VII-17. Laboratory tests for atmosphere resistance

Item	Coating samples composition	Loss of weight [%]	Description
1	Soil + cement (25%) + water	23.4	Sample easily destructed
2	Clay + cement (25%) + water	5.1	Sample easily destructed
3	Clay (45%) + lime (45%) + cement (10%) + water	3.9	Strong sample without cracks
4	Loam + cement (5%) + water	29.0	Sample easily destructed
5	Loam + cement (10%) + water	1.8	Strong sample without cracks
6	Loam + cement (15%) + water	0.5	Strong sample without cracks
7	Loam + cement (20%) + water	0.3	Strong sample without cracks
8	Loam + lime (5%) + cement (5%) + water	27.6	Sample easily destructed
9	Loam + lime (10%) + cement (5%) + water	11.2	Sample easily destructed
10	Loam + lime (15%) + cement (5%) + water	13.2	Sample easily destructed

At this first stage of the work the best results were obtained by cement based compounds, i.e. suspensions of cement in water and suspensions of cement with sand in a 1:3 ratio. Therefore, the possibility of making such coatings cheaper by reducing the cement quantity and by substituting the sand with soil, clay, or loam that are cheaper to obtain was studied. The clay and the loam pit belongs to Ulba and is located close to the tailings pond. Table VII-3 gives the results of laboratory tests for wheathering resistance of a number of clay, loam and cement based coatings. The best results were obtianed with compounds based on a mixture of equal quantities of clay and lime with admixture of not less than 10% of cement, and with loam based compounds with an admixture of not less than 10% of cement. Substitution of a part of

Fig. VII-54. The process of application of polymer coatings.

cement in this compound with lime resulted in a drastic reduction of the strength and the wheathering resistance.

The total quantities of materials needed for coating the beaches with optimum compounds was estimated and respective figures are given in Table VII-4.

Table VII-18. Estimation of the required quantities of materials to construct covers

Item	Surface layer composition	Material				
		Clay [t]	Loam [t]	Lime [t]	Cement [t]	Water [m^3]
1	Clay (45%) + lime (45%) + cement (10%) + water	555	-	555	123.3	1184
2	Loam + cement (10%) + water	-	1009	-	112.1	404
3	Loam + cement (15%) + water	-	953	-	168.2	415

The experimental results showed that there is no real alternative to cement based coatings. At the same time, these coatings are very expensive, and as noted before, no method of application of such coatings to materials with low mechanical strength is available. Based the extensive experience in re-cultivation of ponds taken out of service by covering them with a layer of clay, soil, or loam of 80-100 cm thickness, experiments were undertaken to gradually cover the beaches with loam. The projected thickness of the layer should have provided sufficient mechanical strength for filling the whole beach. In fact, about 20% of the beach surface over most dense part could be covered. However, as soon as it was attempted to cover the more remote parts of the pond bank, at a distance of 3-4 meters, shear failure of the underlying tailings occurred with ensuing destruction of the monolith plate. Figure VII-4 illustrates the effect of the shear failure and the resulting beach destruction. Part of the beach cover has dropped into the tailings and lifted the the top part of the beach, resulting in deep faults. Thus, it is evident that this method of re-clamation was not feasible.

Generally speaking, the main problem is the low shear strength and load-bearing capacity of the beach material and, therefore, the emplacement of covers. In consequence, it will be difficult to solve the problem of the long-term stabilisation of the pond up to complete dewatering. Therefore, further investigations were focused on the development of coatings that do not necessarily possess a long service life but that eliminate problem of dust generation during the time until complete dewaterin is achieved.

In consequence, the making of polymer coatings to prevent dust generation was studied and a number of field experiments were carrried out. Figure VII-5 shows the process of applying polymer coatings on part the beach to evaluate their wheathering resistance under natural conditions.

VII-4. GROUNDWATER CONTAMINATION PROBLEMS

The tailings pond that was taken out of service gives rise to another problem, which is the contamination of underlying aquifers with radioactive and toxic substances. The bottom liner of the pond is malfunctioning, allowing the release of contaminants with the ensuing risk of detriment to the drinking water resources in the Ust-Kamenogorsk area. Figure VII-6 is a map of groundwater contamination with chlorides and sulfates in the Ust-Kamenogorsk area.

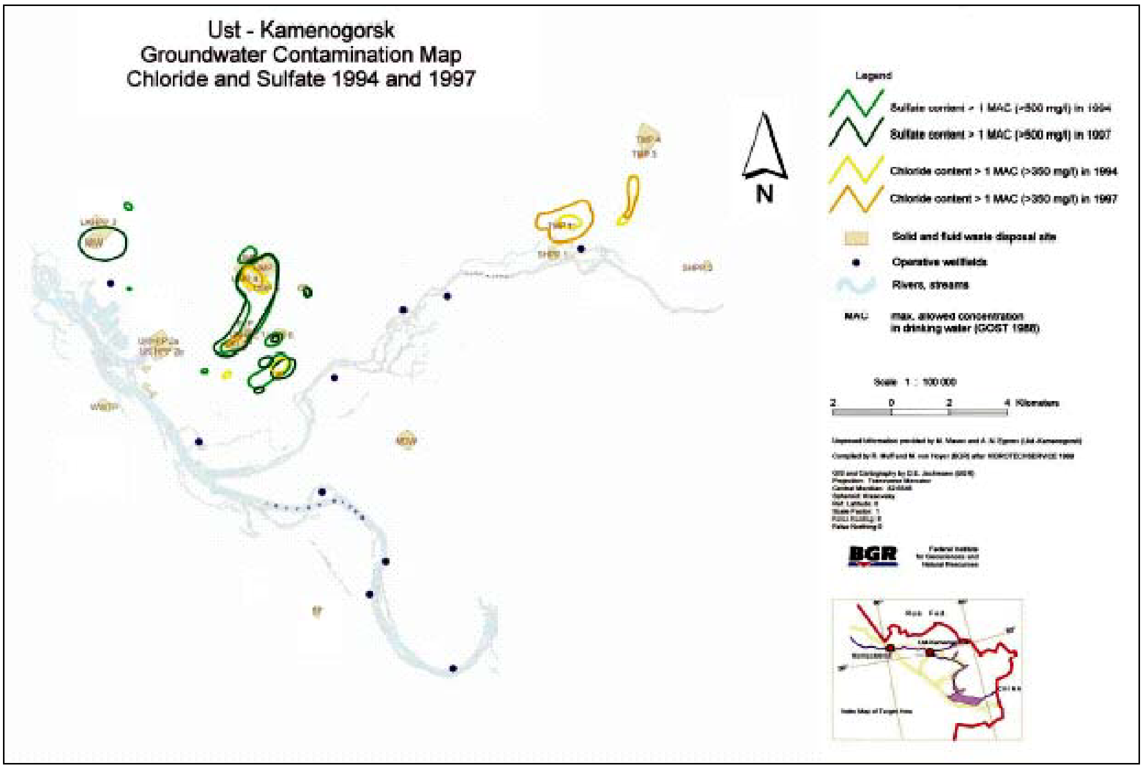

Fig. VII-55. Map of chloride and sulfate contamination in the Ust-Kamenogorsk area [VII-3].

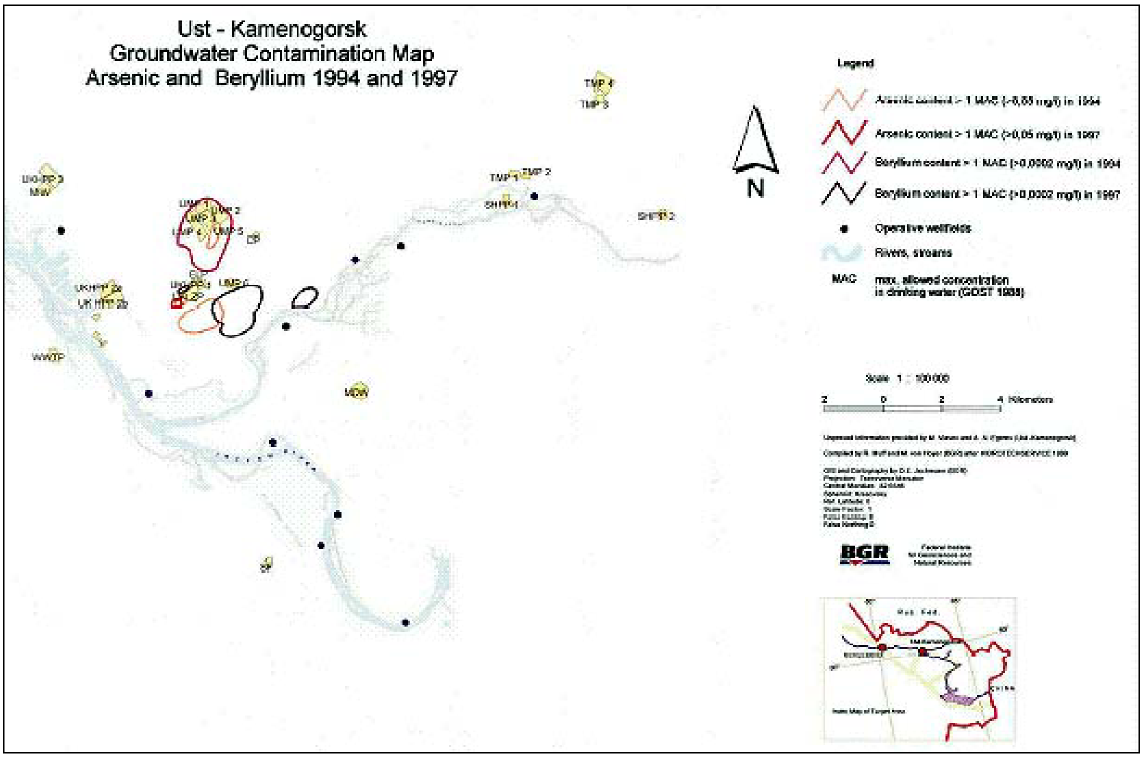

Fig. VII-56. Map of As and Be groundwater contamination in the Ust-Kamenogorsk area [VII-3].

Unfortunately, statistical data on the dispersion of radioactive and toxic substances in the groundwaters are rather limited. However, it is clear that radioactive and toxic contaminants are migrating together with sulfates, which are a component of the impounded wastes.

Figure VII-7 shows the concentration of beryllium and arsenic in the groundwaters. The tailings pond appers to be an active source of groundwater contamination.

The Table VII-5 gives some results of groundwater monitoring in the region of the tailings pond. The Table shows isolated excesses of content of components under control in subterranean waters in comparison with the drinkable water standard (MAL= maximum acceptable level) adopted for Kazakhstan [VII-2]. It was registered that the values of specific alpha and beta activity of subterranean waters and the content of the whole number of harmful and dangerous substances exceed those fixed in standards.

Table VII-19. Some of groundwater monitoring results

Variable	**Unit**	**MAL [VII-2]**	**1**	**2**	**3**	**4**
Solid residual	mg/l	1000	17390	3370	1230	6900
NO_3	mg/l	45	127	487	162	1240
Cl	mg/l	350	360			
SO_4	mg/l	500	10950	1490	510	3270
NH_4	mg/l	2	375	2.5		36
As	mg/l	0.05	0.06			
F	mg/l	1.5				
Cu	mg/l	1				
Pb	mg/l	0.03	0.17	0.05	0.03	0.11
Zn	mg/l	5				
Mn	mg/l	0.1	0.4			
Sr	mg/l	7	9.1			7.8
Be	mg/l	0.0002			0.0003	0.0004
Li	mg/l	0.03	9.4	0.04		3.2
Hg	mg/l	0.0005				
Z_c			556.1	21.5	8.3	172.5
$Z_{c\ 1\text{-}2}$			321.5	3	2.5	113.5
α	Bq/l	0.1	3.9	2.7	0.5	2.6
β	Bq/l	1	1.9	1		1
$Z_c\ \alpha\beta$			41	28	5	27

VII-5. DRAINAGE WATER MANAGEMENT SYSTEM

Taking into account the complexity and scale of the problem a project to prevent groundwater contamination at Ust-Kamenogorsk was developed. This project consists in the building of a new and state-of-the-art tailing pond for the industrial wastes together with a number of supporting engineering measures. Figure VII-8 is a conceptual diagram of the tailings pond and the patterns of contaminanat release to the underlying aquifer.

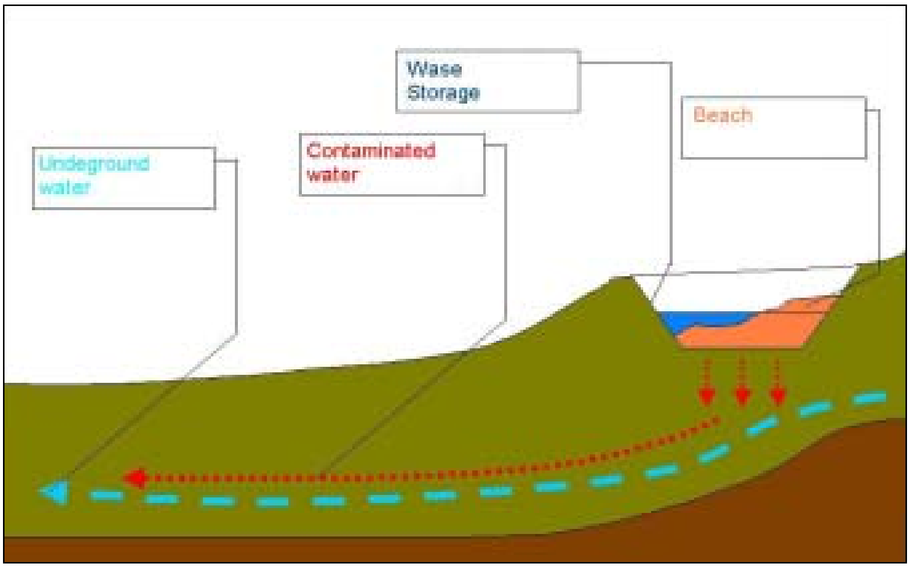

Fig. VII-57. Mechanism of groundwater contamination.

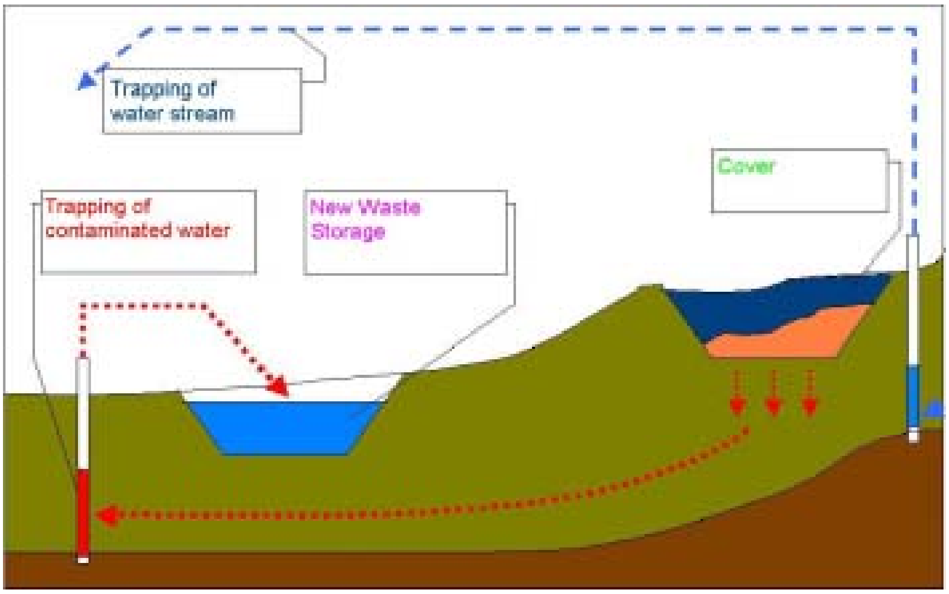

Fig. VII-58. Trapping of underground water.

The project consists of trapping of the main part of the groundwater upstream of the old pond and diverting it on the surface into the river Irtysh. The contaminated groundwater will be collected from wells downstream of the new pond then pumped into it. Contaminated dekant water from the old tailings pond will also be pumped to the new pond. After dewatering of this old pond it will be possible cover it by one of the known methods for building cappings.

VII-6. SUMMARY AND CONCLUSIONS

Thus, the following plan of action for taking the pond out of service service and for providing for its long-term stability was proposed:

(1) Mathematical simulation of groundwaters flows and the migration of radioactive and toxic substances;
(2) Construction of a new tailings storage pond, using state-of-the-art methods for liner construction;
(3) Determination of the optimum arrangement of wells for trapping clean and contaminated groundwaters based on mathematical simulation;
(4) Drilling of wells for trapping contaminated waters and transfer to the new pond;
(5) Transfer of the dekant water from the old pond to the new storage pond;
(6) Construction of a cover providing for the long-term stability of the old pond.

An important condition of the project success as a whole, is the realisation of a programme for process improvement to substancially reduce the amount of liquid waste generated. The main aspects of this program are the introduction of new technologies generating less waste, improvement of the water circulation pattern in the facility, cooperation with the other enterprises of the region on recycling and re-use of wastes, and the development of methods to reduce the amount of harmful components in the wastes.

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[VII-2] GOST 2874-82, "Standards for drinking water quality", Moscow (1982).

[VII-3] BUNDESANSTALT FÜR GEOWISSENSCHAFTEN UND ROHSTOFFE, Proposals for the remediation of contaminated groundwater and hazardous industrial waste sites in the Ust-Kamenogorsk area, Kazakhstan, Technical Cooperation Project No. 99.0795.7, BGR, Hannover (2001).

ANNEX VIII. REPUBLIC OF KOREA

REMEDIATION OF URANIUM MILL TAILINGS USING NATURAL AND ORGANO-CLAYS

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VIII-1. ABSTRACT

Several experimental studies have been conducted for the remediation of uranium mill tailings using natural and organo-clays. A few sorbents were also tested for the non-uranium radioactive species such as cobalt and caesium. Natural clays showed pH dependent sorption characteristics for uranium, indicating the clay surface and uranium species changed with pH variations. The organo-clays showed reduced sorption capacity for uranium. This result indicates that uranium species and organic parts of the organo-clays are competing on the the sorption sites. Therefore, for tailings containing only uranium should be isolated with just natural clays. However, sites contaminated with radioactive materials and toxic organic contaminants would be better treated with organoclays. In the last study period, sorption studies for cobalt were conducted with a few organic sorbents such as *Undaria Pinnatifida* (a sea weed). *Undaria Pinnatifida* showed very high sorption capacity for cobalt. Other sea weeds are under investigation and other radioactive species such as caesium will be tested.

VIII-2. INTRODUCTION

In recent decades, the use of natural and modified sorbents has been considered for the remediation of mining and mill tailing sites. Natural clays (e.g. smectites, bentonites, alumino-silicates, montmorillonites) and clays (e.g. montmorillonite) modified with e.g. the cationic surfactant hexadecyltrimethylammonium (HDTMA) have been used to reduce the mobility of heavy metals and radionuclides. Stabilization of uranium mill tailing using natural and modified clays may prevent migration of residual uranium and reduce the biosorption and ‘bioavailability’ of uranium for uptake by plants, earthworms and microorganisms. In this research, the potential of natural and organo-clays for immobilizing contaminants in uranium mill tailings was investigated. The sorption and desorption characteristics are assumed to be the main controlling factors determining the applicability of these sorbents in the immobilization of such residues and for the *in situ* remediation of contaminated sites.

The projects focused on the characterization of the sorption of uranium species on natural and organically modified clays (organo-clays). Experiments were conducted to investigate sorption characteristics of selected sorbents. The sorption data generated were used to calibrate various sorption models. Laboratory studies were conducted to determine the immobilization capacity of selected sorbents for the treatment of uranium mill tailings

Organic contaminant sorption on an organo-clay has been of significant research interest and as a new technology attracted many soil and environmental scientists and engineers over the last decade. Many studies have been reported with organo-clay, suggesting several potential applications, such as solidification and stabilization of waste effluents, secondary containment for gasoline storage tanks and hazardous waste landfills liners [VIII-1-VIII-5]. Organoclays, which are clays modified by insertion of an organic cation into the interlayer or ion exchange with inorganic cations, such as Ca^{2+}, Na^+, and K^+, in the interlayer spacing. The organic phase provides high sorption capacity for organic contaminants, which partition to this phase.

Two different organo-clays were prepared: montmorillonite modified by exchanging 50% and 100% of the CEC with HDTMA. To test the sorption characteristics, pH was varied as the most important environmental factor. The studies of competitive sorption and desorption of uranium and organic contaminants are still ongoing. The effect of humic material will also be studied. The applicability and economic feasibility of using such clays for in situ treatment of the uranium mill tailings will be analysed based on the experimental results and numerical modelling.

A few biosorbent were also tested for low level radioactive liquid waste which produced from nuclear power plant.

VIII-3. SORPTION STUDY WITH NATURAL AND ORGANO-CLAYS

VIII-3.1. Materials and Experimental Methods

Montmorillonite-KSF™ (Aldrich Chemical Co.) was washed with distilled water at 60 ºC for removal of organic impurities and the removal was monitored by UV of the drained water. The clay suspension was finally filtered through a 0.22 µm membrane filter, and the filtrate was checked for impurities by UV-spectrophotometry (Hewlett Packard, 8452A). The modified montmorillonite was collected by settling, dried for 24 hours, and stored in a brown bottle. The cation exchange capacity (CEC) for this clay was determined to be 50.4 meq/100 g clay. 50% and 100% of the CEC was exchanged with the cation used as an organic modifier, hexadimethylammonium (HDTMA). The details of the procedures can be found in [VIII-7]. The average particle diameter of the collected montmorillonite was about 15 µm.

The HDTMA was obtained as 25 wt% aqueous chloride solution from Aldrich Chemical Co. and used as received. Natural uranium was purchased from Sigma Chemical Co. as uranyl nitrate ($UO_2(NO_3)_2$) and used without further purification. ICP standards were prepared in acid (HNO_3 or HCl) to give a 1000 mg/l U(VI) stock solutions.

Sorption experiments were performed in batches using 40 ml amber vials. Water to clay ratio was 10:1 w/w%, and the mixtures was open to a $CO_{2(g)}$- atmosphere during sample preparation and sealed with Teflon lined caps. The solution pH was controlled with pH buffers and monitored during at the beginning and at the end of the experiments. After 24 hours of sorption, the solid phase was separated by centrifugation and aliquots were taken from the aqueous phase for the determination of uranium concentrations in filtered and unfiltered samples. The aqueous solution was vacuum-filtered through 0.45 µm PTFE filters and the uranium concentrations were measured by Inductively Coupled Plasma Atomic Emission (ICP-AE). The sorbed amount of uranium was calculated from the initial and final uranium solution concentrations.

The data could be best represented with a nonlinear Freundlich sorption isotherm:

$$q = K_F C^N \tag{1}$$

where q ($\mu g \cdot g^{-1}$) = the amount sorbed, and K_F ($\mu g \cdot g^{-1}$)/($\mu g \cdot cm^3$) and N (dimensionless) = Freundlich parameter. N reflects the curvature in the isotherm and may represent the energy distribution of adsorption sites.

VIII-3.2. Result and discussion

A set of experiments was conducted without any pH buffer. The pH of the solution varied between pH 3-4 over the experimental period. The non-modified clay exhibits a higher sorption capacity for uranium than that of organoclay (Figure VIII-1). The solid lines on the figure represent the Freundlich model fitting and a good fit is observed. The organoclay modified with 50% or 100% HDTMA showed a decrease of about 50% in the sorbed amount. This result can be explained thus that the sorption sites for mainly cationic uranium species on the clay are negatively charged and that some of the negatively charged sites were occupied by the organic cation. Thus sorption capacity for uranium is decreased as the clay is modified with organic cations. However, some sorption sites including edge sites that would be available for uranium.

Figure VIII-2 to VIII-4 show sorption isotherms of uranium for different pH values (pH 6-10). Figure VIII-2 shows the sorption isotherm of uranium on non-modified clay. At pH 6, the sorption capacity was the largest and the sorption amount decreased as the pH increases. This phenomenon was repeated for the 50% HDTMA and 100% HDTMA-modified clays and can be explained by the speciation of uranium(VI): it exists at pH of 2-4 mainly as UO_2^{2+} ion, regardless of the presence of carbon dioxide. UO_2^{2+} is converted to $UO_2(OH)^+$ around pH 5-7, At even higher pH-values the dominant species will become $UO_2(OH)_2{}^0{}_{(aq)}$ and $UO_2(OH)_3^-$.

Table VIII-1. Freundlich model parameters for sorption of uranium onto unmodified clay and HDTMA-clays.

Clay	Sorption			
	pH	K_F	n	R^2
Montmorillonite	6	9.7028	0.2535	0.9866
	8	1.6879	0.2695	0.9887
	10	0.0208	0.9619	0.9893
50% HDTMA Montmorillonite	6	9.3107	0.1534	0.8809
	8	1.8381	0.2347	0.9945
	10	0.0601	0.7367	0.9854
100% HDTMA Montmorillonite	6	1.2078	0.3543	0.9817
	8	2.2306	0.2098	0.9983
	10	0.3816	0.4311	0.9705

Therefore, the cationic U species at low pH have higher sorption affinity for the clay and at high pH uranium is changed into to neutral or anionic species. This leads to low sorption affinities for unmodified and organo-clay. The same lower overall sorption capacity of the organically modified clay can be observed in Figures VIII-2 to VIII-4, as was observed in Figure VIII-1 with unbuffered solutions. Figure VIII-2 to VIII-4 indicate that the pH effect is higher for the unmodified clay, while the effect is diminished as the ratio of HDTMA exchange increases. For the unmodified clay a significant difference was observed between pH 6 and pH 8 (Fig. VIII-2), while this difference is much smaller for the modified clays

(Figs. VIII-3 and VIII-4). A summary of the fitted Freundlich model parameters is given in Table VIII-1.

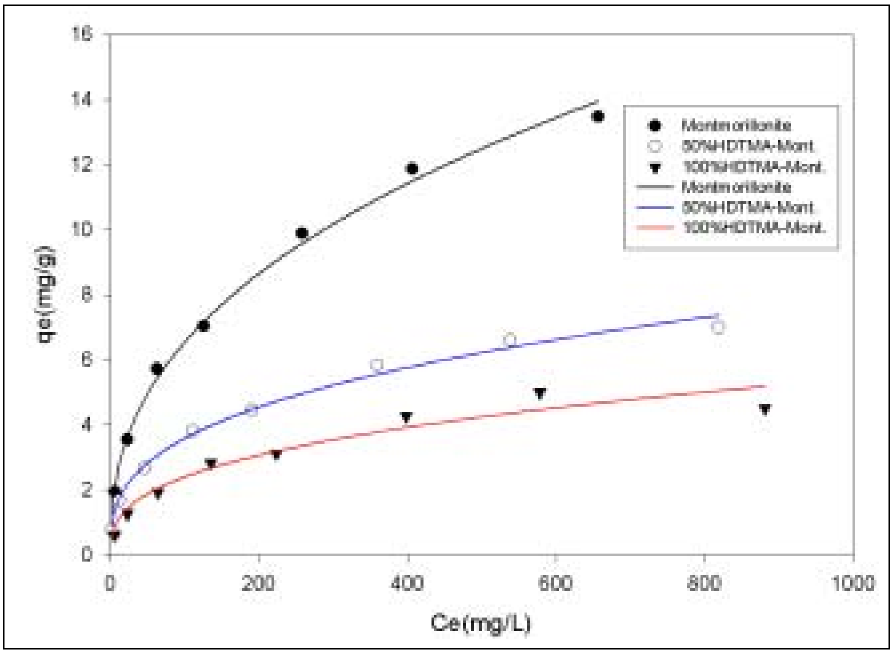


Fig. VIII-1. Sorption isotherm of uranium without pH buffer. Lines represent Freundlich isotherms.r.

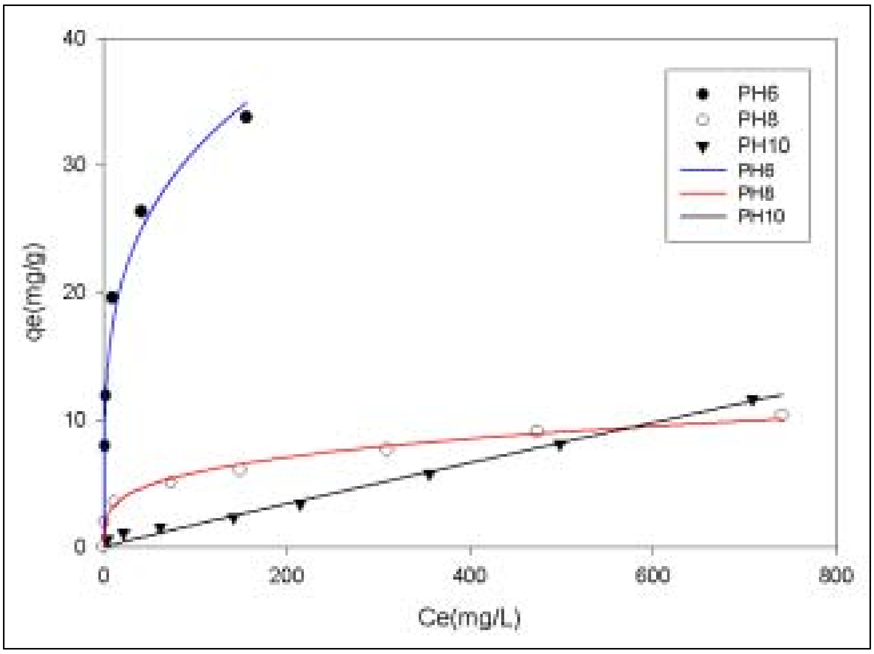


Fig. VIII-2. Sorption of uranium on montmorillonite at pH 6-10. Lines represent Freundlich isotherms.

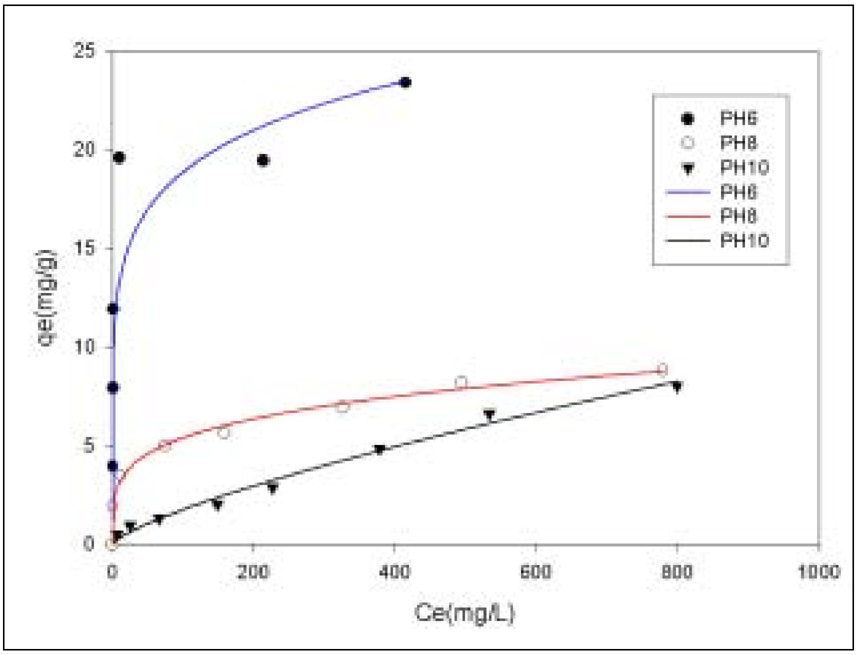


Fig. VIII-3. Sorption of uranium on 50% HDTMA-montmorillonite at pH 6-10. Lines represent Freundlich isotherms.

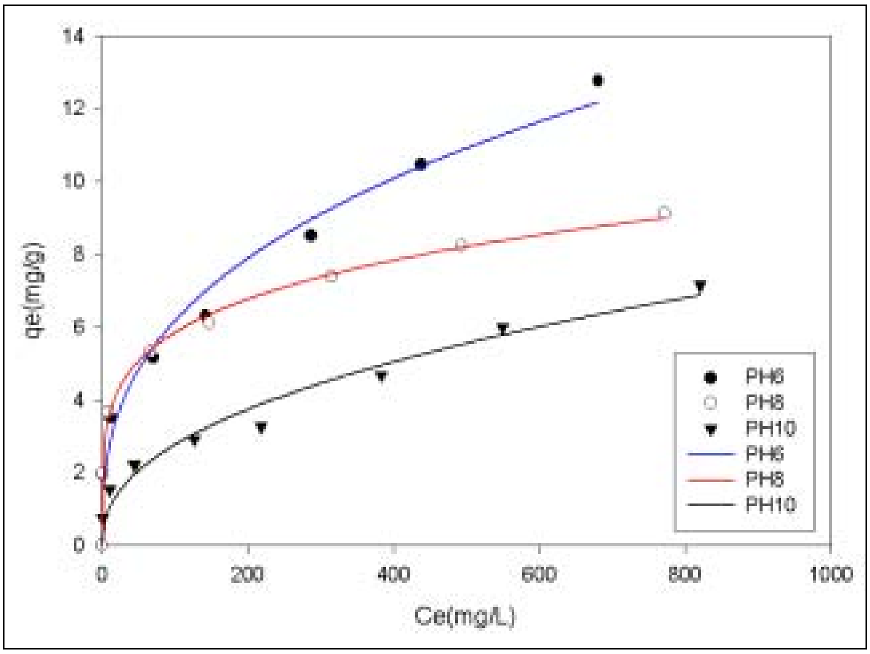


Fig. VIII-4. Sorption of uranium on 100% HDTMA-montmorillonite at pH 6-10. Lines represent Freundlich isotherms.

VIII-4. Evaluation of competitive binding effects

The results indicate that the sorption mechanism are not same for all of the tested clays. On the unmodified clay, cations, such as Na^+, Ca^{2+}, or K^+, can be exchanged for uranium, but on the organoclay, the organic cation will not readily exchange with the uranium cations. The Freundlich model is not intended for evaluating competitive sorption effects.

To analyse multicomponent competitive adsorption behaviors, inter alia the Langmuir Competitive Model (LCM) [VIII-8], the Ideal Adsorption Solution Theory (IAST) model could be used [VIII-8]. LCM is an extended form of the Langmuir model that allows predictions of the amount of solute *i* adsorbed per unit weight of adsorbent, $q_{m,i}$, in the presence of other solutes.

$$q_{e,i} = \frac{q_{m,i} b_j C_{e,j}}{1 + \sum_{j=1}^{n} b_j C_{e,j}} \tag{2}$$

where C_e is the equilibrium concentration of solute *i* in a mixture consisting of *n* components, while $C_{e,j}$ are the equilibrium concentrations of all the adsorbing solutes in the mixture. The constants b_i, b_j and $q_{m,i}$, q_{mj} are determined from single-solute systems.

VIII-5. Experiments with biosorbents

Biosorbents are well known as high sorption capacity for heavy metals. They could apply for radionuclides including uranium. In this study a few biosorbents were tested for fission products such as Co, Sr, Cs. Biosorbents might use with organoclay to increase the sorption capacity multicontamination.

VIII-5.1. Materials and methods

Three sorbents, *Chlorella*, *Spirulina*, and *Undaria Pinnatifida* were studied for the cobalt removal from liquid waste. The liquid waste was prepared from non-radioactive compounds to avoid exposure to the laboratory workers. $NaNO_3$, KNO_3, $Ca(NO_3)_2$, $Co(NO_3)_2$, $CsNO_3$ were purchased from Aldrich Chemicals. All the chemicals were used as received from the vender without further purification. Co(II) stock solution was prepared by dissolving $Co(NO_3)_2$ into nano-pure water. The sorbents was pH adjusted and the cobalt stock solution was added. All the kinetic and isothermal tests were conducted in a temperature controlled shaker (VS-8480SF, Hanil) at 200rpm and 25ºC. 0.1 N of HCl was used for the pH adjustment. The solid to liquid ratio was 150 (0.2 g sorbents and 30 ml nano-pure water). The sample solution was filtered with 0.22 µm syringe filter or centrifuge for 10 minutes at 1700 rpm (Hanil, MF600). The prepared solution was ananlyzed with AAS (atomic adsorbed spectrometer, Varian Spectra AA 250 Plus). The q_e was calculated with a following equation.

$$q_e = (C_0\text{-}Ct)V/M \tag{3}$$

where C_0: initial concentration, C_t: concentration after equilibrium, V: volume of liquid, and M: weight of the sorbent. The sorption isotherm was fitted with the Freundlich model.

VIII-5.2. Results and discussion

Chlorella and *spirulina* have showed low sorption capacity for Co and Cs in preliminary experiments. Hence, these two sorbents were not subject to more detailed studies.

VIII-5.2.1. Kinetic study for Undaria pinnatifida

Figure VIII-5 shows the result of kinetic studies with Co. Equilibrium was achieved within 30 minutes. The quick attainment of equilibrium is essential for a compact design of a reactor. Good cadidates of sorbent should have fast kinetic properties. However, all the sorption isotherm experiment were given 2 hours to ensure the equilibrium.

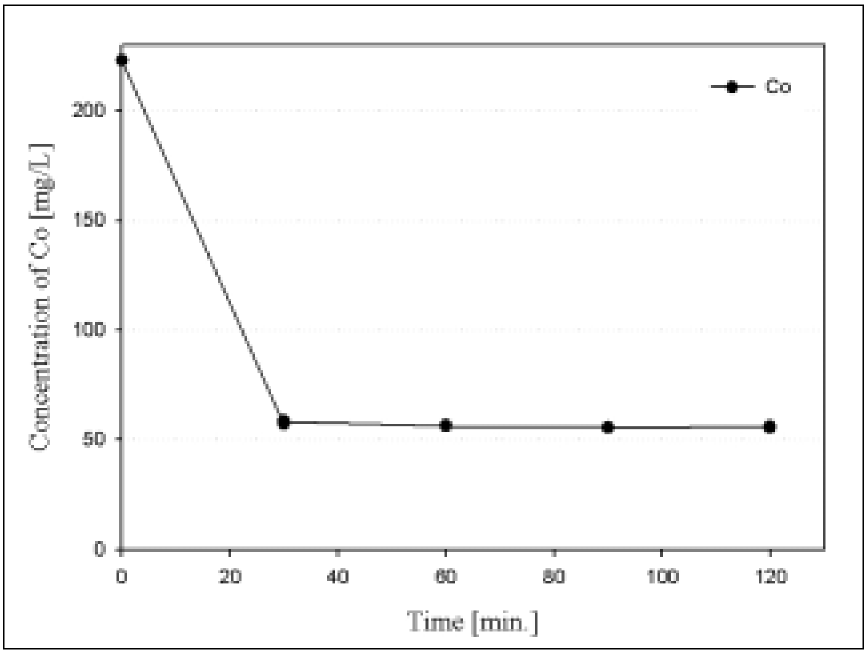


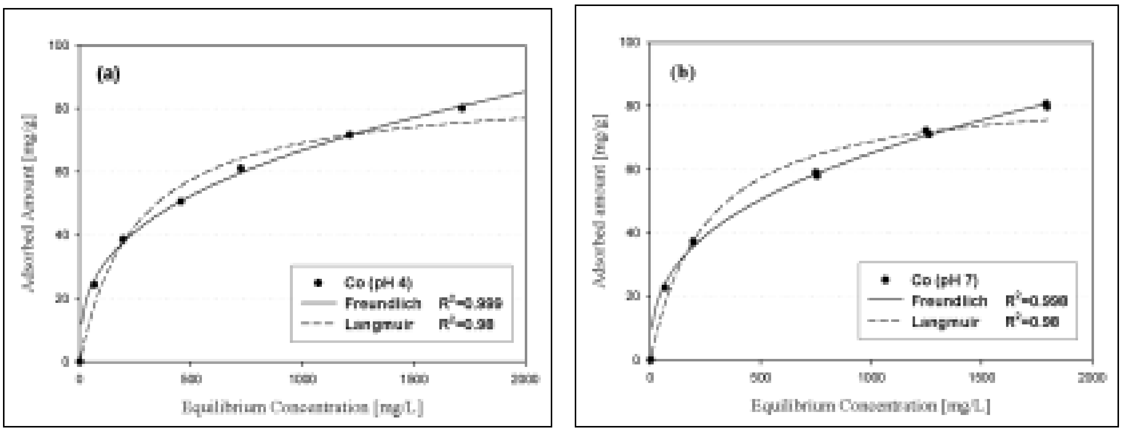
Fig. VIII-5. Sorption kinetic of Co on Undaria pinnatifida.

VIII-5.2.2. Sorption isotherm for Undaria pinnatifida

Figure VIII-2 shows the sorption isotherm for Co on *Undaria pinnatifida* at differnt pH-values. The results indicate that the sorption capacity of *Undaria pinnatifida* is rather high in comparison to other sorbents. Table VIII-I. shows the sorption capacity for three different sorbents. Figure VIII-2 also indicates that the sorption characteristics are independent of the solution pH. There is no significant difference for pH 4 and pH 7. This indicates that the sorption mechanism is not dependent on the proton concentration. Complexation between the metal and functional groups on the organic materials, such as –OH and –COOH was proposed in the literature as a fixation mechanism. However, the present results seem to indicate that the major fixation mechanism on *Undaria pinnatifida* is not complexation, but ion exchange.

Table VIII-1. Sorption amount (mg/g) in single solute system of *Undaria pinnatifida* for Co (C_0= 200 mg/l)

Sorbent	***Chlorella sp.***	***Spirulina sp.***	***Undaria Pinnatifida***
Co	3.2	7.5	25
Cs	2.4	1	under investigation



(a) pH 4, (b) pH 7.

Fig. VIII-6. Sorption isotherm of Co on Undaria pinnatifida for different pH values.

VIII-5.2.3. Effect of non-radioactive salts

Other metal constituents, such as sodium, calcium, magnesium, iron, in solutions to be treated compete for binding places on the substrate [VIII-6]. Figure VIII-7 shows the effect of sodium on the sorption of cobalt on *Undaria pinnatifida*. The effect of sodium in not significant. Usually the multivalent ion have higher competition effect. Currently the effect of calcium and magnesium on the cobalt sorption is under investigation.

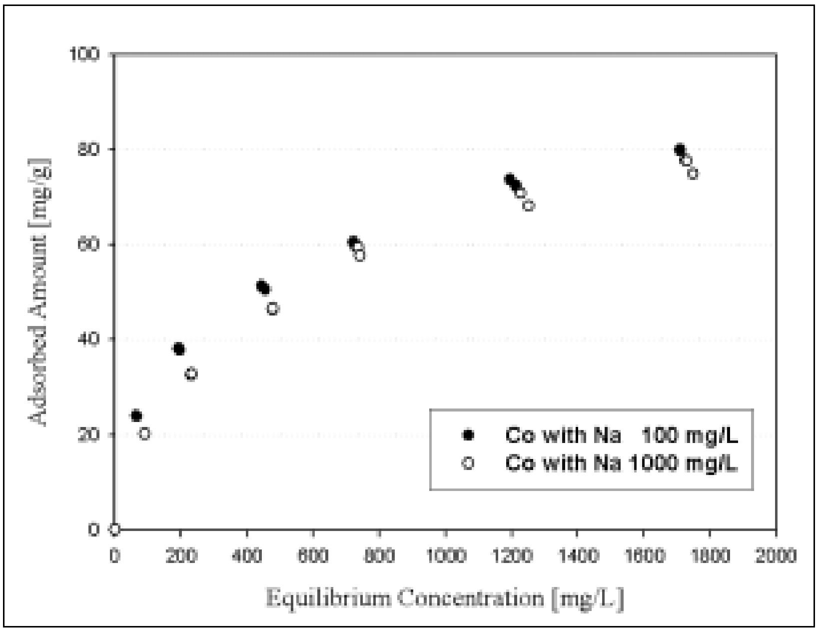


Fig. VIII-7. The effect of Na on the binding of Co on Undaria pinnatifida.

VIII-5.2.4. Biosorption for other radioactive species with seaweeds

The current result of biosorption showed that the sea biomaterial have a high sorption capacity and suggested further studies with the material targeting other radioactive materials such as Cs, Sr, and U. We keep working on the experimental study for removal of radioactives with biomaterials.

VIII-6. Summary and Conclusion

At first sorption study of uranium on natural clay and organo- clay. Generally, lower pH showed higher sorption amount and the sorption was decreased as the solution pH increased. It can be explain as UO_2^{2+} is the dominant species for the lower pH while $UO_2(OH)^+$, $UO_2(OH)_2^0(aq)$, $UO_2(OH)_3^-$ have low sorption characteristics to the clay and organo-clay. Organo-clay showed lower sorption capacity for uranium sorption in comparing with non-modified clay. This results imply the some sorption sites for uranium species are competing with HDTMA but not all of the sites are occupied by HDTMA indicating the organo-clay can be used for sites which contaminated with toxic organic compounds and uranium.

At second experimental study, biosorption for low level radioactive liquid waste.

Among the tested three biosorbents, *Undaria pinnatifida* has the highest sorption capacity for cobalt and implicate that biosorbents could be a good cadidate for concentrating and/or removal of ionic radioactive materials.

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ANNEX IX. POLAND I

IMPROVEMENT OF SOIL PROPERTIES APPLIED TO CAPPING AND MULTI-LAYER BARRIERS

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IX-1. ABSTRACT

Results are presented on the research on the selection of natural clayey materials for the remediation of mill tailings and the possible control of their properties. The following objectives were formulated: a) minimizing of the required soil mass, b) improvement of the material employed. Three methods for the modification of clayey soil were examined:

- application of an external electrical field,
- hydrolysis by applying strong acids and strong bases,
- a combinded method.

The research focused on two types of clays, on a kaolinite and on a illite/smectite. An electric field of E= 0.2 V/cm was used. The samples were also treated with 0.1 N chloric acid, strong base, peptizing agent and an industrial waste solution from an etching processes. Remaining heavy metal concentrations in the solutions were used as a measure for the effectiveness of modification to the clays.

Both, the chemical and the electrical method generated similar changes in the clay structures. The electrical method appeared to be simpler in field applications. However, the research showed that the chemical treatment of the clay material was the most efficient. Disappointing results where obtained from the electrical treatment of samples, i.e. the sorptive properties of clays did not improve substantially and the pH change was ambigeous.

IX-2. INTRODUCTION

The process of mining and milling of uranium ore results in tailings that are deposited in ponds. The tailings consist of ground-up minerals and of process water. From a geotechnical point of view these tailings have a liquid or semi-liquid consistency. The tailings pore waters may be acid, neutral, or alkaline, depending on the process technology. The pore water may also contain various radionuclides and heavy metals, as well as contaminants such as oils.

The properties of the tailings itself, as well as tailing ponds construction pose a significant threat to the environment. Remediation of tailing ponds and the surrounding environment can be very complex process [IX-1–IX-3]. The effectiveness of the technical solutions applied is a function of quantity and quality of data, as well as of suitable models [IX-1,IX-3].

The most significant technical problems are old, abandoned tailing ponds. Typically a combination of three independent barriers is present: geological barriers, technical barriers and waste body itself as barrier. Technical barriers include capping in order to limit water ingress and contaminants migration, radon release, as well as direct radiation exposure [IX-1, XI-3]. Simon and Müller [XI-3] divided the capping systems to: Standard capping systems and alternative capping systems. In the standard capping system there are used natural and conventional synthetic materials (e.g. clays, geomembranes) for their conventional properties (flexibility, impermeability, etc.). It sometimes happens that economical and technical conditions call for unconventional solutions, that is alternative capping system [IX- 3].

The goal of the present work is finding new alternative solutions for the capping and isolation of tailing ponds. Generally, such alternative solutions consists of physical or chemical modifications to conventional materials, allready built into the capping layers (activation or modification of properties) or applying a new, modified conventional material. The effectiveness of the new layers consists in active reaction with the tailing pond environment for example adsorption and immobilization of heavy metals.

As was shown by earlier research [IX-4–IX-6], as well as by research of others [IX-7–IX-12] that alternate application of strong acid and base to clayey soils causes hydrolyses of clayey minerals. Such modified clayey soils show improved sorbing properties for radionuclides and heavy metals. When added to the tailings, such modified clays, would counteract the migration of radionuclides and heavy metals. In the research presented here previously developed methods [IX-4,IX-6,IX-13,IX-14] were adapted and earlier results were put into perspective [IX-5,IX-6,IXI-14].

IX-3. THE SCOPE AND OBJECTIVES OF THE RESEARCH

The following goals and objectives have been used to conceptualise the research work:

Research goals

— Improvement of the retention capacities of clay layers;
— Elimination or decrease of migration of contamination from tailing pond;
— Immobilization of heavy metals/radionuclides and trapping them in the tailing pond.

Research Objectives

— Modification of clays that are conventionally used in tailing ponds to construct barriers;
— The agents and materials should be easily obtainable (preerably of local origin);
— The technology should not be complicated.

Earlier work [IX-4, IX-6,IX-8,IX-10,IX-17] has shown that the sorptive capacity of clays can be increased by modifying the structure through treatment with strong acids and bases. The modification of the structure facilitates the penetration of ions and the chemical treatment weakens the aluminosilicate bonds Si – O – Al.

Building on previous studies of electro-osmotic effects [XI-13,XI-14], it was found that electro-osmosis is capable of modifying the structure of clays [IX-4,IX-7,IX-15,IX-16,IX-18,, XI-19]. This forms the fundamental idea of the present research work, by which it was observed that an external electric field changes the properties of clays in a way similar to the treatment with acid and bases.

Emphasis was placed on:

a) improvement of the retention capacities of the clays by modification of their properties

b) creating new alternative layers on the basis of clay minerals

The property modifications to the clay minerals are brought about by applying an electric current (electrokinetic processes) or applying of strong acids and basics (clay mineral hydrolysis).

IX-4. EXPERIMENTS WITH AN EXTERNAL ELECTRIC FIELD

IX-4.1. Materials investigated

Kaolinite from the 'Maria' mine in Nowogrodziec and illite/smectite clay from the 'Kraniec' mine near Brzeg Dolny were used in the studies. Four samples of paste consistency (ca. 50% water content) were prepared form these materials. One samples served as a control and was untreated, while a second and third were subject to electro-osmosis, and the fourth was treated with the 0.1 N HCI for a time-period of one day. Sub-samples of these preparations were dried and subjected to microscopic examination in the air-dry state.

IX-4.2. Characteristics of external electric field

The experiments with an external electric field were carried out in rectangular plastic containers. The clay samples had a dimension of 15 × 15 × 50 cm.

The electrodes (three anodes and cathodes each) were made from 10 mm diameter perforated stainless steel tubes. The spacing between the anodes and the cathodes was L = 40 cm. A direct current (DC) of E = 0.2 V/cm electric field intensity was applied. The polarity was changed at 8 hour intervals in a second set of experiments.

IX-4.3. Microscopic examination

An STEREOSCAN-180 electron-microscope was used for the inspection of the gold-coated samples at the magnifications of 1000, 3000 and 10000 times. Black and white image of the structures were taken (see Figs. IX-1 to IX-6).

IX-4.4. Experimental results

The electron microscope examinations show that the clays in their natural, unaltered state consist predominantly of microaggregates with 'surface to surface' type contacts, and there are more 'interparticle' pores than 'interaggregate' ones. The unaltered clays are characterised by a laminar/domain-like structures (Figs. IX-1 and IX-2).

Treatment with 0.1 N hydrochloric acid (HCI) changes the morphology of the clay minerals as shown in in the electron microscope images (Figs. XI-3 and IX-4). The most important changes observed include:

- formation of aureoles of tiny matter around particles and aggregates, resulting in an obliteration of the sharpness of the primary shapes;
- exposure of tiny, single silt grains;
- formation of interaggregate pores of 0.003-0.005 mm average diameter;
- increase in the number of 'mixed' type contacts as opposed to 'surface to surface' type contacts;
- formation of a matrix/turbulent structure [IX-16].

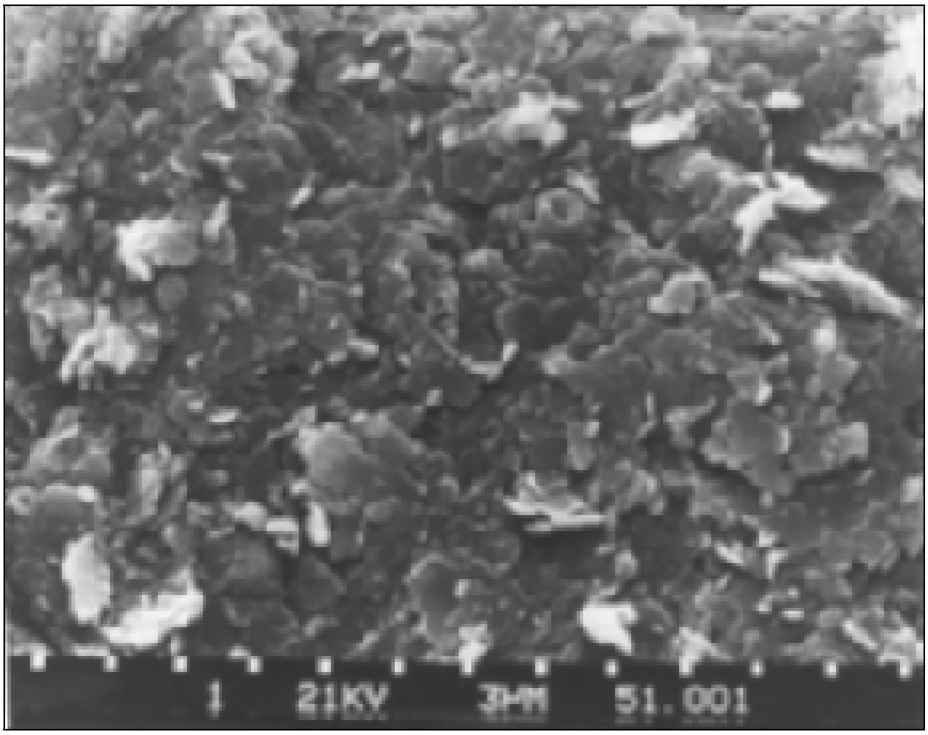

Fig. IX-1. Kaolinite. The structural construction (fabric) before the action of electric current and HCl.

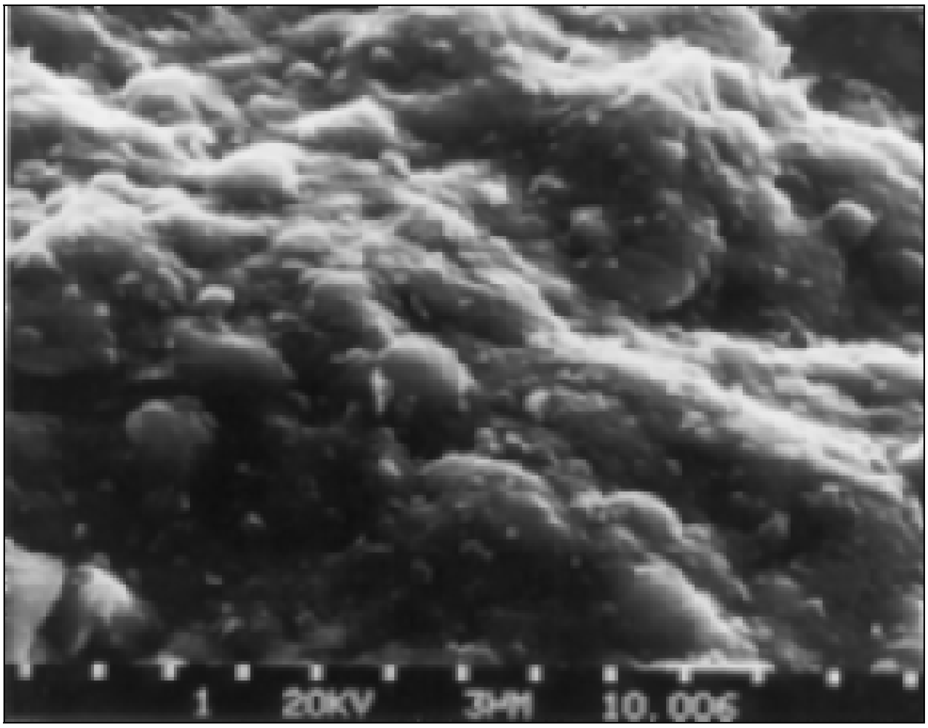

Fig. IX-2. Illite/smectite clay from the 'Kraniec' mine.

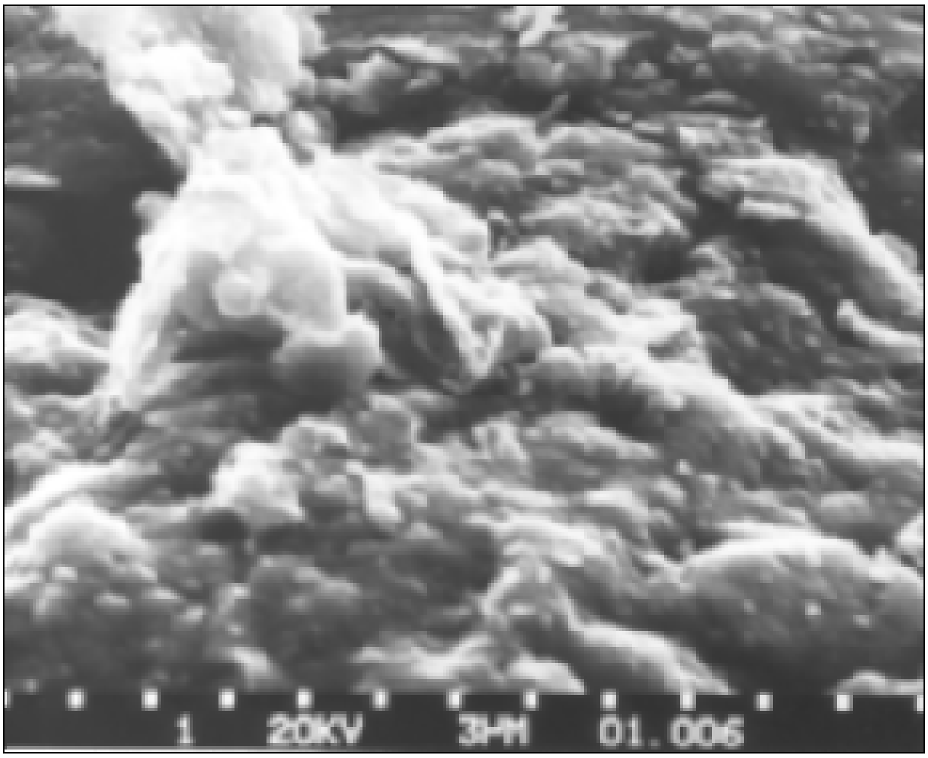

Fig. IX-3. Kaolinite after the activation with 0.1 N HCl.

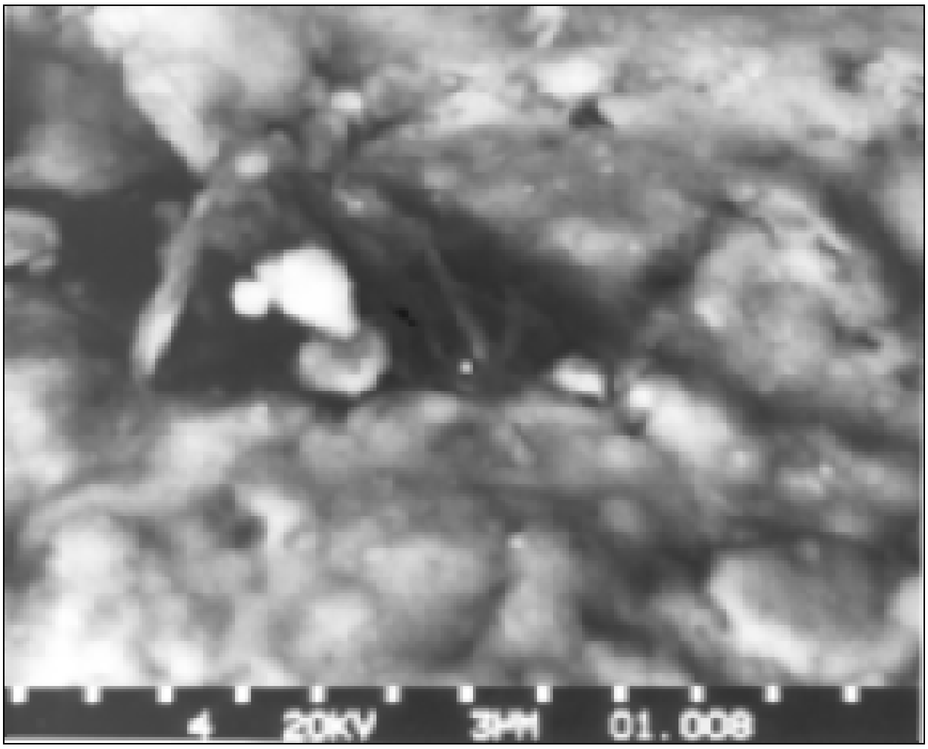

Fig. IX-4. Illite/smectite clay after the activation with 0.1 N HCl.

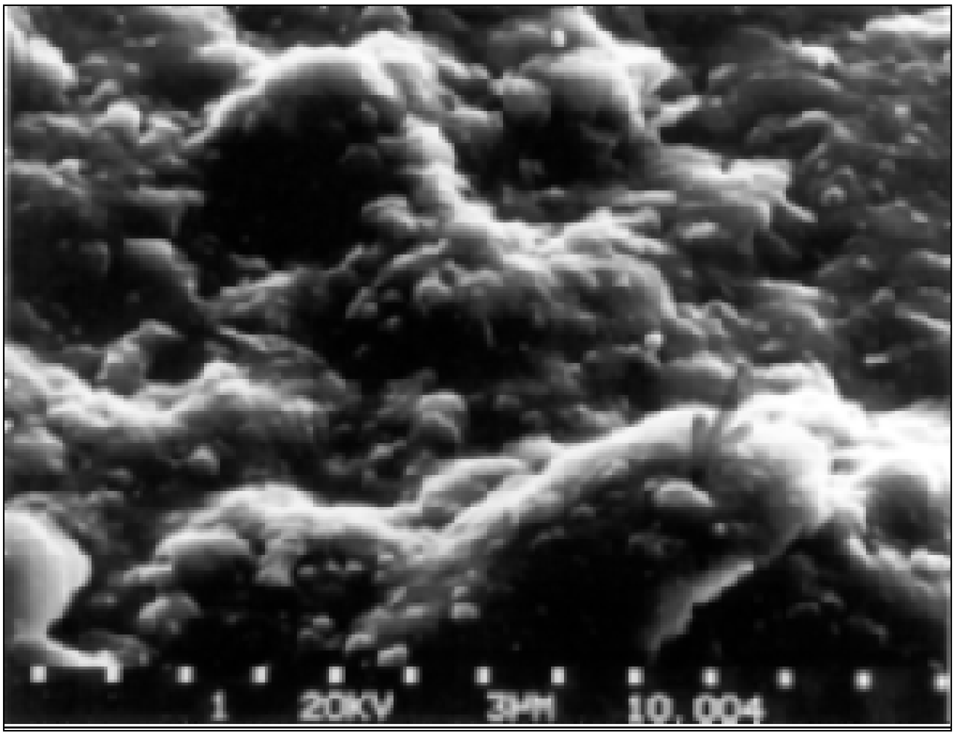

Fig. IX-5. Kaolinite after electro-osmosis – anodic zone.

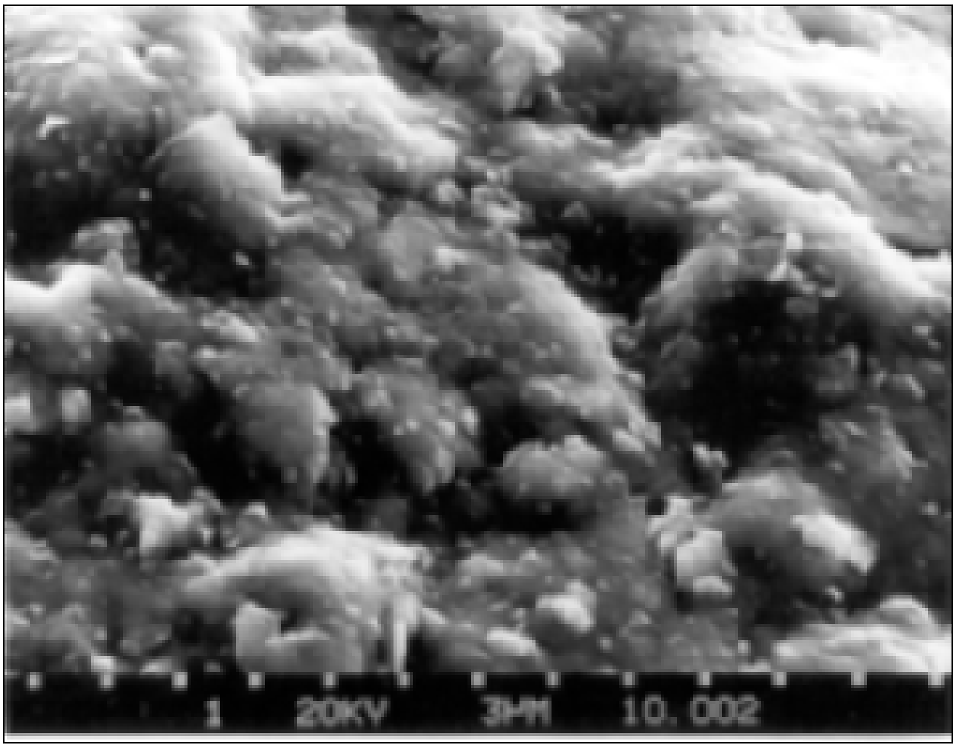

Fig. IX-6. Illite/smectite clay after electro-osmosis – anodic zone.

These changes are more evident in the illite/smectite (Fig. IX-4) than in the kaolinite samples (Fig. IX-3).

The application of external electric fields (electro-osmosis) with a constant polarity DC generates in the perianodic zone structural changes similar to those produced by the acid treatment. The changes having a similar character for both clay types examined (Figs. IX-5 and IX-6), namely:

- specific aureoles are formed on mineral aggregates and on single particles;
- the domain-type of the structural pattern are preserved, with an evident transformation in the matrix structure [IX-16];
- Numerous interaggregate pores of 0.001-0.003 mm diameter are formed.

The experiments during which the polarity the of electrodes was changed every eight hours did not induce an effect significantly different from that with constant polarity. The illite/smectite clay showed features almost identical to those produced with a constant polarity, while the kaolinite structure did not differ from the initial structure very much.

IX-5. CHEMICAL MODIFICATION OF CLAY SORPTION PROPERTIES

IX-5.1. Clays

The illite/smectite clays from the ‘Kraniec’ mine near Brzeg Dolny (Poland), used in these experiments, were in the dry-air state, while samples for the electro-osmosis experiments were made up from material at the natural water contents (21.5%).

IX-5.2. Industrial waste solutions

The metal removal capacity of the clays was investigated by bringing samples into contact with simulated tailings porewaters. As a subsitute an industrial waste solution from a metal pickling process at one of the metallurgical plants in Wrocław was used. The experiments had to give answers to the following:

- would consituents of the industrial waste solution be sorbed by the clays and to what extent ?
- what are the reagents proportions needed to achieve the highest effectiveness of the clays as alternative layers to prevent heavy metal migration ?

The respective metal concentrations in the industrial waste solution are given in Table IX-1.

Table IX-1. Heavy metals contents and pH in industrial wastes.

	Measured concentration	**Allowable max. value as per Polish standard**
Zinc	158 mg/dm^3	2.0 mg/dm^3
Copper	72 mg/dm^3	0.5 mg/dm^3
Lead	1.95 mg/dm^3	0.5 mg/dm^3
Chromium	< 0.1mg/dm^3	0.2 mg/dm^3
$pH_{reaction}$	2.75	6.5-9.0

IX-5.3. Clay modification method

The air-dry clay was activated in an acidic dispersion using 0.1N HCl of pH2-2.5, then in a CaOH solution of 12-14 pH. Adding 1% of 15M NaOH or sodium water-glass acted as peptising agent. The procedures were adapted from earlier experiments [IX-5,XI-6].

The treatment was carried out in a blender, adding first the acid, then the base, and finally the peptising agent. The added chemicals resulted in a paste-like consistency of the clays.

Untreated, in a natural state, clay was used for the electro-osmotic experiments. A DC of E = 0.2 V/cm field intensity was passed through the clays over a period four days The experiments were carried out in a 15×15×50 cm plastic container. The electrodes were made from brass pipes (Ø 10mm) wrapped with a brass net. They were arranged vertically so there were three cathodes at one end and three anodes at the other. The anodes respectively cathodes were spaced 3 cm apart and the distance between cathodes and anodes was 40 cm. Samples were taken from the peri-anodic and peri-cathodic zones.

Three series of tests were carried out:

Series I: waste solution + modified clay
Series II: waste solution + modified clay + water-glass
Series III: waste solution + clay after electro-osmosis

IX-5.4. Criteria for the effectiveness of the treatment

Two criteria were applied to evaluate the treatment effectiveness [IX-4–XI-6]:

(1) The final pH(f) achieved in the waste solution compared to the discharge requirements in the range of pH 6.5 to 8.5.
(2) The reduction of the contaminant concentrations in the effluents with respect to the maximum allowable value as per Polish discharge standard.

IX-5.5. Experimental results obtained

Series I: waste solution + clay

In this series various ratios of waste solution to clay were tested and the effect was expressed in terms of the final pH(f) reaction.

Test I/1	waste solution / clay =	1000 / 1
Test I/2	waste solution / clay =	500 / 1
Test I/3	waste solution / clay =	333 / 1
Test I/4	waste solution / clay =	250 / 1

Table IX-2. Change of contaminant concentrations in industrial waste solutions (Series I)

Variable	WA	Concentration [mg/dm^3]				Reduction [relative %]			
		I/1	I/2	I/3	I/4	I/1	I/2	I/3	I/4
Zn	158	150	156	152	4.2	5	1.3	3.8	97.3
Cu	72	70	63	63	< 0.05	2.8	12.5	12.5	> 99.9
$Cr_{gen.}$	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	nc	nc	nc	nc
pH(f)	2.75	5.8	2.9	2.72	2.68				

nc = no change

Series II: waste solution + clay + water-glass (WG)

The following combinations were prepared for the experiments:

Test II/1 waste solution / clay / WG = 1000 / 1 / 0
Test II/2 waste solution / clay / WG = 500 / 1 / 2
Test II/3 waste solution / clay / WG = 333 / 1 / 2
Test II/4 waste solution / clay / WG = 200 / 1 / 2
Test II/5 waste solution / clay / WG = 250 / 2 / 1
Test II/6 waste solution / clay / WG = 300 / 3 / 1
Test II/7 waste solution / clay / WG = 150 / 2 / 1

Note: Water-glass (WG) was added step-wise.

Table IX-3. Changes of contaminant concentrations in industrial waste solutions (Series lI)

	Concentration [mg/dm^3]								Reduction [relative %]						
	WA	II/1	II/2	II/3	II/4	II/5	II/6	II/7	II/1	II/2	II/3	II/4	II/5	II/6	II/7
Zn	158	130	135	133	150	<0.05	<0.05	<0.05	17.7	14.6	15.8	5.1	>99.9	>99.9	>99.9
Cu	72	65	58	56	70	0.08	<0.05	<0.05	9.7	19.4	22.2	2.8	>99.9	>99.9	>99.9
Cd	0.05	0.03	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	40.0	80.0	> 80	> 80	> 80	> 80	> 80
Cr_{gen}	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	nc	nc	nc	nc	nc	nc	nc
pH_k	2.75	2.8	8.5	8.5	6.6	8.3	8.2	8.3							

WA = waste solution; nc = no change

Series III: waste solution + clay after electro-osmosis

Test III/1 waste solution / clay from the cathodic zone = 200 / 1

Test III/2 waste solution / clay from the cathodic zone = 100 / 1

Test III/3 waste solution / clay from the anodic zone = 200 / 1

Test III/4 waste solution / clay from the anodic zone = 100 / 1

The experimental results obtained for the concentrations of individual constituent were scattered around the initial values. The reaction pH-values also did not show a significant trend. For this reason the experiment were not interpreted any further to date.

IX-6. SELECTED EXPERIMENTS WITH MODIFIED CLAYS

It was noticed earlier work that an electric current and strong acid and basics cause similar structural changes. However, it turned out that the electrokinetic method is difficult to apply in practice. For this reason, work with electrokinetic method was discontinued and the research programme on modification using chemical methods was enlarged. The results of two out of several modification experiments with clays are discussed below.

IX-6.1. Experiment A

IX-6.1.1. Petrological properties of the clay used

The material used was an illite-montmorillonite clay of tertiary age, containing 44.2% of clay particles with a diameter d<0.001 mm, 34.9% in the fraction 0.001 – 0.01 mm, and 20.9% in the fraction 0.01 – 1mm. The plasticity index is Ip = 34%.

IX-6.1.2. Chemical composition of soil

The chemical composition of the soil was determined as:

SiO_2	62.1%	Fe_2O_3	8.13%
Al_2O_3	18.51%	SO_3	0.38%
MgO	1.7%	CaO	6.32%
K_2O	0.3%	Na_2O	0.50%
Total carbonate	>10%	Loss on ignition	1.48%
pH	7.8		

IX-6.1.3. Preparation and characteristics of the suspension

After careful disintegration, a series of samples were prepared from the clay. The suspensions were prepared in distilled water, or 0.1 N of HCl or H_2SO_4. Some suspensions were neutralized by adding enough lime or cement together with an additional dosage of NaOH to achieve a pH 12 – 14 (see Tabs. IX-4 and IX-5). Finally the peptizing agent was gradually added. Some of the suspensions were prepared at room temperature and pressure, while others were heated to 100°C. The full set of suspensions prepared is listed in Table IX-4.

In order to measure their removal capacities, the suspensions then were mixed and stirred for one minute with a solution containing heavy metals (% by weight):

Ni – 0.123; Zn – 0.203; Se – 0.14; Pb – 0.010; Cd – 0.059, with a solution pH = 6.0.

The heavy metals removal from the solution was calculated in percent relative to the initial value and is presented in the Table IX-5. The results obtained indicate that the assumed modification of clay is adequate for release, and its effectiveness depends only on the used reagents and proportions.

Table IX-4. Characteristics of the various suspensions prepared for experiment A.

I	Suspension in water: 8 parts by weight of clay and 100 parts by weight of water.
II	Peptized clayey suspension in water: 8 parts by weight of clay, 0.05 parts by weight of tannin as a peptizing agent, 100 parts by weight of distilled water
III	Peptized clayey suspension in water: 8 parts by weight of clay, 0.1 parts by weight of clay of 15M NaOH as peptizing agent, 100 parts by weight of distilled water.
IV	Peptized clayey suspension in water: 8 parts by weight of clay, 0.05 parts by weight of 1 N sodium pyrophosphate ($Na_4P_2O_7$), 100 parts by weight of distilled water.
V	Clayey suspension in HCl solution (pH = 2): 8 parts by weight of clay, 100 parts by weight of 1 n HCl.
VI	Clayey suspension in HCl solution (pH = 2): 8 parts by weight of clay, 100 parts by weight of 1 n HCl, followed by a neutralization with calcium 5 parts by weight of CaO, i.e. to obtain pH = 12 and introduction of 0.1 parts by weight of tannin (peptizing agent) with heating to 100ºC.
VII	Calciferous-clayey suspension in water: 8 parts by weight of clay, 5 parts by weight of CaO, 100 parts by weight of distilled water.
VIII	Cement-clayey suspension in water: 8 parts by weight of clay, 5 parts by weight of cement, 100 parts by weight of distilled water.
IX	Clayey suspension in a solution of H_2SO_4 (pH = 2): 8 parts by weight of clay, 100 parts by weight of 1 n H_2SO_4, followed by neutralization utilizing additives: 8 parts by weight of cement, 0.15 parts by weight of tannin, and heating to 100ºC.

Table IX-5. Percentage of heavy metal removal for experiments A [relative to the initial value].

Suspension	Ni	Zn	Se	Pb	Cd
I	26.8	37.5	47.1	45.0	37.3
II	36.3	42.8	48.6	50.7	42.3
III	31.2	39.9	48.2	47.0	41.9
IV	39.1	43.4	49.2	51.4	44.3
V	29.8	39.2	48.1	49.9	43.5
VI	97.6	99.0	78.6	90.0	96.6
VII	84.3	87.1	68.9	83.5	82.4
VIII	76.7	81.2	74.5	77.9	83.1
IX	98.4	99.5	85.7	95.0	98.6

IX-6.2. Experiment B

IX-6.2.1. Petrological properties of the clay used

A cambrian clay of plasticity I_p = 22.9%, containing 33.5% of clayey fraction < 0.001mm was utilized for the modification. The following was determined in the chemical composition of soil:

SiO_2	64.3%	Al_2O_3	17.02%
Fe_2O_3	4.37%	CaO	1.85%
MgO	2.21%	SO_3	0.09%
Na_2O	0.71%	K_2O	3.6%
total carbonate	< 2%	loss on ignition	4.5%

IX-6.2.2. Sample preparations

In contrary to experiment A, the components were chosen so that the modified clay was in a form of a paste with a viscous-plastic consistency. Four types of modifications were selected to test removal effectiveness for heavy metals from an industrial waste solution. Different acid reagents were used for the preparation of the sorbent: 0.1 N HCl, H_2SO_4 or H_3PO_4, solutions of heavy metals at pH = 1-2, and acid industrial waste solutions of pH = 3. The pastes were neutralized by adding calcium oxide or Portland cement, in some cases calcium hydroxide sludge, and finally the peptizing agent of pH 12-14 was introduced. Some samples were prepared at ambient temperature, some samples at 100ºC, and other samples in an autoclave at 150ºC and a pressure of 5 atm (Table IX-6).

The clay samples prepared in such way were mixed with the previously prepared solutions of heavy metals at a ratio of 100 waste solution : 1 clay. The composition of the waste solution was as follows (% of mass):

Al – 1; Mg – 3; Fe – 5; Mn – 0.4; Ni – O.1; Co – 0.003; V – 0.0003; Cr – 0.5; Cu – 0.07; Pb – 0.04; Zn – 1; Cd – 1; and pH = 2.56.

Table IX-6. Characteristics of the various suspensions prepared for experiment B.

I	20 parts by weight of clay + 5 parts by weight of 0.1 N HCl solution (ratio 4:1) = hard plastic paste. The paste was heated up to 900ºC and left in that temperature for half an hour. Next, 1 part by weight of calcium was introduced to the viscous-plastic paste, in terms of CaO and the saturation humidity of the mixture the amount of distilled water was increased to 70 – 80% in relation to the weight of the clay. The mixture was then mixed and 0.1 parts by weight of NaOH was added. The obtained mixture was subjected to the temperature of 150oC and the pressure of 5 atm in the autoclave.
II	In this variant the components and proportions are the same as in variant I, but there was no temperature or pressure increase imposed.
III	20 parts by weight of clay + 5 parts by weight of industrial waste at pH = 2.56. The paste was heated as in variant 1, then the viscous- plastic paste was complemented with 6 parts by weight of calcium hydroxide slime of pH = 12 of CaO concentration of 10%. Whereas the mixture was complemented with a peptizing agent – 0.1 parts by weight of sodium pyrophosphate. Next, the obtained mixture was heated under the same temperature and pressure conditions as in variant I.
IV	This variant differs from variant III as after the introduction of alkaline reagent the temperature or pressure was not increased. The same industrial waste, that had to be purified utilizing the produced sorbing agents, was used to prepare the sorbing agent in accordance to variants 3 and 4.

After mixing, chemical analyses were carried out and the effectiveness of metal removal was determined (% relative to the initial concentration). The results are given in Table IX-7 and show a high removal effectiveness regardless of the preparation. It was also noted that the consumption of acid and alkaline reagents was several times lower than that for suspensions, and at the same time, a significant increase of acid and alkaline hydrolysis, followed by peptization was observed.

Table IX-7. Percentages of metal removal for experiments B [% relative to the initial concentration].

Susp.	Al	Fe	Mn	Ni	Cr	Cu	Pb	Zn	Cd	total
I	97.0	99.4	99.2	99.5	100	99.0	100	100	100	99.2
II	93.4	97.1	92.2	90.1	95.6	94.8	96.9	92.7	95.8	94.3
III	99.5	99.9	99.5	99.5	97.0	100	96.9	100	100	99.5
IV	95.2	97.8	96.3	94.1	96.5	98.2	97.1	93.8	97.6	96.3

IX-7. SUMMARY AND CONCLUSIONS

In this work the effect of inorganic chemicals and electrical field on the sorption properties of various clays. Activation of these sorption properties is key to creating new, alternative ways of constructing isolations and capping for tailing ponds or to revitalise old, ineffective clay layers.

Clays were mixed with 0.1 N hydrochloric acid and left to react for one day to induce hydrolysis. Hydrolysis occurs more intensively in illite/smectite clays than it does in kaolinite. A perceptible tendency to break the domain-type structure, the formation of aureoles on the surfaces of particles and aggregates, the formation of oval interaggregate pores of 0.003-0.005 mm diameter are evident effects of structural transformations.

An external DC electric field, of E = 0.2 V/cm, both with static and periodically changing polarity induced changes similar to those observed with acid treatment. These changes are, however, less intensive, manifesting themselves mainly by the formation of aureoles and interaggregate pores. An appropriately selected electric field could be used to induce a controlled changes in the clay properties.

The cyclical changes of the polarity induced similar changes as the continuous current flow. Investigations showed that electric current induces substantial structural changes in the first flow phase, that is up to eight hours.

The properties of illite/smectite clay were modified with mineral acids and a strong base with appropriate pH adjustment (NaOH and water-glass).

In preliminary experiments the most effective dosages of modifying agents were selected with a view to achieve an optimum sorption activity. The clays thus modified were used as a sorbent for the treatment of industrial waste solutions containing heavy metals. The effectiveness of metal removal and neutralisation at various ratios of clay to waste solution was determined.

The most effective combination appeared to be the ones in which waste solution was mixed with modified clay and with an additive of water-glass at the ratios of 250 / 2 / 1; 300 / 3 / 1, and 150 / 2 / 1. In addition, the expected buffering of pH was obtained for these mixtures with a considerably lower addition of modified clay.

The modification of clay properties using electric current did not yield results similar to those by chemical modification. The changes in sorptive capacities, as expressed in terms of the final heavy metals concentration in the suspensions, were erratic. The final pH values are difficult to interpret. For this reason, this avenue was not followed further.

The experimental results indicate that modified clays may be a useful component in barriers that are intended to prevent the migration of radionuclides and heavy metals in seepage from tailing ponds. Chemically modified clay may also be added to the tailings themselves in order to increase their retention capacity for radionuclides and heavy metals.

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ANNEX X. POLAND II

ROOM TEMPERATURE CERAMICS, THE BREAKTHROUGH MATERIAL FOR LONG TERM STABILISATION AND ISOLATION OF LOW LEVEL URANIUM RESIDUES ?

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X-1. ABSTRACT

While some 100 hundred locations with uranium mineralisations are found in Poland, only three contained economic reserves and where mined. The tailings impoundment at Kowary Podgórze required attention due to degradation. This paper examines the design and materials for improving the retention of radionuclides within the tailings body. Certain uranium and radium sulphate minerals exhibit low solubilities and fostering their formation appears to be a good way to improve the long-term stability of the tailings. Their formation conditions and wheathering resistance were investigated.

X-2. INTRODUCTION

Some 100 localities with uranium mineralisation are found in the Lower Silesia district in Poland. Only three of them (Kowary Podgórze, Radoniów and Kletno) contained economic reserves and were mined. The Kopaniec deposit contained subeconomic uranium ores only [X-1]. Uranium exploration and exploitation took place in the early 1950s and caused local pollution around the four active mining areas (Kowary Podgórze, Radoniów, Kletno and Kopaniec). Mining of uranium in Poland ended in 1963, but hydrometallurgical processing of low-grade ores was continued at Kowary until 1972 [X-2]. These processing activities resulted in a significant amount of solid residues that were stored in a tailings pond. It may release contaminants to the environment.

A survey of all contaminated sites within Lower Silesia has been carried out in 1971 and in 1991-1994, 1997 and 1998 [X-3]. These investigations revealed local-scale impact of uranium mining and milling on the environment in Lower Silesia. Uranium contamination is limited to the mining milling residue impoundments and their close vicinity at Kowary Podgórze, Radoniów, Kopaniec and Kletno. All of the impoundments are unprotected and are readily accessible for unauthorized trespassers, and it is still possible to find fragments of uranium ore in the deposited material that contain 0.7% of U. Dispersal of uranium can take place due to erosion and transport by river waters, chemical dissolution by rain waters and migration into groundwaters, and antropogenically, when mining wastes containing uranium are utilized for construction. Floods may acerbate uranium dispersion.

Both past and recent field γ-measurements showed the presence of high radiation levels on the surfaces of the mine dumps at Kowary Podgórze, Radoniów, and Kletno. The Kowary Podgórze and Kletno dumps contain ore fragments with up to 1,500 ppm and 9,370 ppm U (11,650 Bq/kg of radium), respectively.

X-3. THE KOWARY PODGÓRZE TAILINGS POND

In the Kowary Podgórze area two mines were in operation. The older, iron-ore mine has been active since the 11th century. The second mine was opened shortly after World War II. At this mine, uranium ores were extracted during the period 1951-1963. The tailing pond is located within the facilities of the old iron mine, on a slope (10 to 25°) of Mt. Rudnik. There is no on-going remediation work apart from the Kowary tailing. At Kowary Podgórze the dump at adits 19 and 19a is eroded and particles containing U-minerals are transported downstream of the Jedlica river [X-3]. A catastrophic flood that took place in Summer 1997 caused rapid erosion and dispersion of the uranium-bearing materials into the Jedlica river valley and stream sediments. The local situation of the tailing pond near Kowary Podgórze village is shown in the Fig. X.1.

The tailing pond contains sediments of sand and clay grain size fractions, that are composed of quartz, feldspar, various micas, amphibolite, clay minerals (as decomposition products of primary material), fluorite, pyrite and some other unidentified mineral phases, as well as some solid metallurgical residues with high content of Al, Ni, Zn and Na-sulfates.

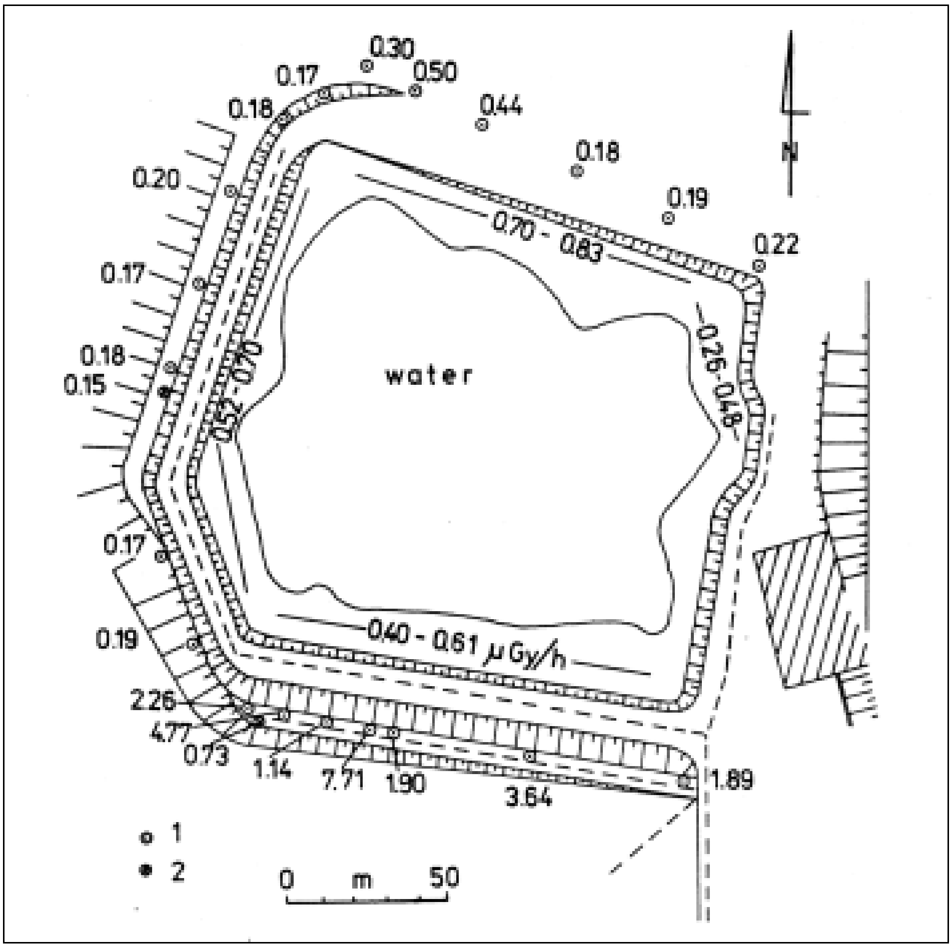

Fig. X-1. Tailing pond near the Kowary Podgórze village , situation as in 1999.
1- measurements points, 2- background measurements.

Table X-1. Key parameters of the tailings facility.

Period of U-mining operation	1951-1963
Period of milling and chemical treatment	1966-1972
Period of chemical plant	1972- present
Volume of extracted uranium	unknown
Total volume of the material (processing)	90 000 m^3
Total volume of the tailings	113800 m^3 = 250000 t
Surface of the tailings pond	13000 m^2
Length of the tailing dump	300 m (dams on three sides only, the fourth one is a natural slope)
Maximum height of the tailing dump	12 m
Total activity	7400 $Bq \cdot kg^{-1}$
Surface Radon activity	1000-1200 $Bq \cdot m^3$
γ-dose: tailings	up to 1 $\mu Gy \cdot h^{-1}$
γ-dose: dump (S-slope)	up to 8.87 $\mu Gy \cdot h^{-1}$
Uranium content	about 30 ppm, max.- 240 ppm
Radium content	$3\text{-}8 \cdot 10^{-5}$ ppm Ra Kg^{-1}
Other metals content	unknown

X-4. CONCEPTUAL MODEL FOR THE LONG-TERM STABILIZATION OF URANIUM MILL TAILINGS

In order to minimise the impact of uranium on the environment, covering of the impoundments with non-radioactive material was suggested for sites located away from residential areas. Complex designs for capping, and bottom and surface drainage systems have been developed world-wide in order to prevent the dispersion of radionuclides and other contaminants from (mining) residues into soils and groundwaters, and to prevent the emanation of radon into the ambient atmosphere.

The complexity of performance criteria leads to the exploration of new materials, including the room temperature ceramics for the immobilizing of various contaminants. If the tests of such ceramics would prove satisfactory, they will open the way for new technologies in the long-term stabilization of wastes. The proposed approach is based on an understanding of diagenetic processes and their chemical reactions, including redox reactions.

In the case of the Kowary tailing pond a proposal to built a new disposal facility nearby appears to be the best solution. The existing tailing is located on the high, right bank of the Jedlica river. This bank is severely eroded by the river. The bottom of the tailing bed is protected against seepage of the contaminated water. One edge of the tailing is a natural slope. Such mode of tailings disposal gives free access for groundwaters. During the planning of the disposal facility, classification of radioactive material is required. Thus, actually radioactive materials can be separated from barren rocks. This helps to reduce the volume of material to be relocated and, hence, to minimize the required capacity of the new facility. The following investigations are aimed at reducing the volume of the new constructed waste disposal:

- dewatering
- backfilling to the exisitng mine works (which helps to reduce volume of the residues, and reduces the accessibility of still dangerous mine works, e.g. shafts and adits)
- alternative use of residues for constructing motorways (sandy fraction mixed with sand)

— remediation of the existing tailing pond (with a view to separate radioactive from non-active residues)
— construction of a new disposal facility using natural minerals as an active membranes in order to encourage dewatering

The general objectives and design elements are outlined in Table X-2.

Employing room-temperature ceramics for the construction of active membranes within a part of the newly constructed disposal facility is expected to result in reduced leakage of contaminants to the groundwater and soil. In the proposed project, natural materials (anhydrite, gypsum, and calcium hydroxide) would be used for stabilization and fixation of contaminants. The internal structure of the disposal facility is composed of intercalations of waste and the active barrier is a key factor of the long-term stabilization. The construction is shown in Figure X-2. Design details of the disposal facility will be proposed after the final development of the low temperature ceramics based on natural materials has been completed.

tailing profile - existing		tailing profile - proposed	thickness	
top: waste + water		top soil of the new tailing	0.3-0.4 m	
waste		2-50 mm j fresh rock fraction (top dewatering system)	0.2-0.3 m	
		soil (clays)	0.2 m	
		material from the dam	0.2 m	
		anhydrite mixed with waste (**active membrane**)	0.3 m	
		waste from the tailing	0.7-1.0 m	
		anhydrite + calcium hydrooxide (**active membrane**)	0.1-0.2 m	
		waste	0.7-1.0 m	
		anhydrite + calcium hydrooxide (**active membrane**)	0.1-0.2 m	
		waste from the tailing and from decommissioning	0.7-1.0 m	
		anhydrite + calcium hydrooxide (**active membrane**)	0.1-0.2 m	
		waste from the tailing	0.7-1.0 m	
		anhydrite + calcium hydrooxide (**active membrane**)	0.1-0.2 m	
		peat (**active membrane**)	0.1-0.2 m	
		soil from the dam + clays (**active membrane**)	0.2-0.3 m	
wast		20-200 mm j fresh rock fraction (dewatering system)	0.3-0.4 m	
top soil ?		top soil (natural)		
bed rocks		bed rocks		
total thickness: up to 12 m			**3.5** - 5.5 - 7.0 m	

Fig. X-2. Proposed depth-profile for a reconstruction of the uranium waste disposal facility

Table X-2. Objective and design elements.

Main Risk	General Design	Design Elements
Direct radiation	Shielding	Cover and membrane system
Tailing dispersion	Long-term stability Long-term erosion protection	Erosion protection barrier against extreme events: *minimize the grade of the slope* *vegetate cover*
Water contamination: *surface water* *groundwater*	Isolation of tailing: *seepage barrier* *erosion protection* *radionuclides traps* *heavy metals traps*	Infiltration barrier: *top* *internal* *bottom*
Radon emanation	Radon attenuation	Radon barrier

X-5. RATIONALE FOR THE USE OF ROOM-TEMPERATURE CERAMICS

Various diagenetic processes have been observed in (uranium) mill tailings and it appears advantageous to exploit and stimulate these processes for the long-term stabilization of the tailings. During microscopic studies, the following minerals have been recognized: quartz, K-feldspar, plagioclase, biotite, muscovite, chlorite, amphibolite group and clay minerals, calcite, dolomite, epidote, fluorite, garnet, gypsum, magnetite, haematite, goethite, pyrite, chalcopyrite, pyrrhotite, barytes, rutile, anatase, sphen, and zirconium. Clay minerals are evidence for decomposition of plagioclase and K-feldspar in an acidic environment, and are responsible for cementation of terrigenous materials. The presence of goethite and haematite suggest a quick oxidation of various minerals. These types of natural processes are well known as limonitization, which is also responsible for cementation. Presence of sulfides within the residues trigger powerful oxidation reactions. During such reaction, S^- is converted into S^{6+}. This ion can very easily form acidic environment [X-3]. This is the beginning of numerous diagenetic processes such as kaolinitization, illitization, anhydritiztion and silicification. All these processes are responsible for the creation of local geochemical environments. The reactions proceed until an equilibrium between stable phases is reached.

These were the basis for the selection of natural cements. The most common natural cements, such as anhydrite, gypsum, lime (or calcium hydroxide) have been included. All these materials have no or little environment impact. Their physical properties, such as bulk permeability, physical strength, and their low price are important factors considered in their selection. These materials can be named low-temperature natural ceramics.

Finely dispersed uranium minerals are oxidized quickly with the release of uranyl ions that can migrate away from the tailings. Reduction processes, to the contrary, typically lead to the fixation of uranium in solid phases with comparatively low solubility. Uranium precipitates typically within the pH-range between 6.5 and 8.5 pH and the Eh-range between 0 and -0.4 V. According to Eh-pH diagrams (system U-C-O-H, 25°C, 1 bar), $UO_{2(s)}$ is stable over the whole range of pH and depends only on Eh, that is –0.4 to +0.1 V for acidic environments and below –0.4 V for alkaline environments [X-4] $BaSO_{4(s)}$ and $RaSO_{4(s)}$ are stable under similar conditions (usually 3-12 pH and –0.8 up to +1.2 Eh). The redox boundary (between $FeS_{2(s)}$ and $Fe_2O_{3(s)}$) runs through the middle of the stability field of $UO_{2(s)}$ [X-4].

Precipitation of uranium within the tailing can fix free uranyl ion in new, stable phases that would be in equilibrium with existing mineral phases and newly precipitated sulphates, iron hydroxides and clay minerals. Freshly precipitated iron hydroxides have highly active surfaces that can scavenge heavy metal and other cations. This type of reaction, which is often observed in the natural environments, is the basis of the project, and a model for stabilization of uranium mining and milling residues. In the proposed model for the remediation of tailings, attention is paid to the sulfate concentrations. Sulfates are a stimulator of dissolution processes. Sulfides are common constituent of residues originating from natural uranium ores. According to Wisotzky [X-5] a two step oxidation process is proposed. During the first step, sulfide-sulfur is oxidized, followed by the oxidation of ferrous iron. The final results of oxidation are similar to those presented in the equations listed below (after [X-6]).

$$2\ FeS_2 + 7\ O_2 + 2\ H_2O \leftrightarrow 2\ FeSO_4 + 2\ H_2SO_4 \tag{1}$$

Ferrous sulfate is instable in the oxidized zone and is converted to:

$$12\ FeSO_4 + 6\ H_2O + 3O_2 \leftrightarrow 4\ Fe_2(SO_4)_3 + 4\ Fe(OH)_3 \tag{2}$$

Ferric sulfate is usually unstable under oxidising conditions and is subject to hydrolysis according to equation:

$$Fe_2(SO_4)_3 + 6H_2O \leftrightarrow 2Fe(OH)_3 + 3H_2SO_4 \tag{3}$$

During the oxidation, an active iron-hydroxide gel is produced. Then, this gel is dehydrated and converted into limonite (mixed compound), monohydrate (goethite = FeOOH), hydro-hematite ($Fe_2O_3 \cdot ½H_2O$), and haematite (Fe_2O_3). Such reactions occur in a neutral or weakly acidic environment.

The effects of redox reaction have been observed at the Kowary tailing oxidation of iron that consumes an extra charge. In the course of these reactions, the solid phases of iron hydroxide (goethite) and haematite, as well as gypsum are formed.

The oxidation of iron, consumes an extra charge, will be creating an environment conducive to the reduction of UO_2^{2+} to UO_2. The thus obtained pitchblende is quite stable in normal (surface and subsurface) oxidising and locally anoxic environments. In order to minimize dissolution of UO_2 by sulfuric acid formed according to reactions (1) or (3), the construction of an active membranes containing calcium oxide or hydroxide is recommended. These compounds will react with any free acid by forming gypsum.

As a result of the oxidation, various elements can be mobilised from the mining residues, but radium remains associated with sparsely soluble phases such as Ca-, Ba- or Sr-sulfates. Radium solubility is very low under normal conditions. Hence, radium is usually precipitated together with the earth-alkaline elements, forming insoluble sulfates as a first solid precipitate. Barium-radium sulfates are well known from mine waters of some hard coal mines, as well as uranium mines. To ensure that the radium remains in the residues, it is proposed for the construction of the new disposal facility to use modified anhydrite and calcium hydroxide as components of the internal active membranes.

A key problem for the proposed remediation project is the stabilisation of pH in order to maintain a suitable environment for the oxidation and cementation (fixation) reactions. To achieve this proper environment and the stabilizing reactions, a special design for the disposal facility is proposed. It consits of intermixed layers of residues and barrier components, composed of natural materials. The presence of CaO or $Ca(OH)_2$ and anhydrite in the internal membrane will reduce vertical migration of sulfates. Redox reactions will be responsible for secondary precipitation (reduction) of uranyl anions. Similar reactions are responsible for the formation of uranium deposits.

The addition of calcium hydroxide as proposed for construction of the active membrane, should buffer pH in the near-neutral area. Active solidification of waste pile will be running successively with redox reaction due to natural cementation of sandy-clay waste fraction with natural precipitated phases [X-7]. These is a good probability for long-term stabilization of radio nuclides with the proposed construction method for the tailings.

Sorption can also be an important factor for reducing migration of some radionuclides and other pollutants. Sorption of uranium and other contaminants occurs on iron and silica gels thus reducing the contamination of groundwaters.

X-6. MINERALOGICAL ANALYSES

The mineralogical composition of the mining residues was determined using an ore microscope and XRD. Under the microscope, pan concentrates obtained from two visually distinctive types of residues have been studied. The XRD analyses were carried out on fine-grained material (clay fraction) separated from the mining residues. The fine fraction of the residue was obtained by suspending the material in distillated water. After four hours of sedimentation, the supernate containing the clays in suspension was collected and then dried. Radium activities were measured at the Faculty of Physics and Nuclear Techniques of the University of Mining and Metallurgy in Kraków using gamma spectroscopy.

X-7. ASSESSMENT OF RADON EMANATION

X-7.1. Conceptual appraoch

Radon is a natural radioactive element in gaseous form. It is a decay product of radium, which is present in many rocks and building materials. Radon escapes from these via diffusion and convection. The radon release from solid materials to the air-filled pore space is known as emanation. The parameters that characterise quantitatively this effect are the emanation coefficient and/or exhalation rate [X-8]. The **emanation coefficient** expressed as percentage, is defined as a ratio of the number of Rn atoms released from the solid phase into the pore space to the total number of Rn atoms that produced by the Ra-decay. The **exhalation rate** is defined as an exhaled radon activity per mass or surface unit of a sample per unit of time.

The radon activity N_1 [Bq] produced by the radium activity in the investigated material sample is calculated by the following equation:

$$N_1 = Ra \times m(1 - e^{-\lambda_{Rn} t_1}) \tag{4}$$

where: Ra is an activity concentration of ^{226}Ra in the investigated material in [Bq/kg], λ_{Rn} – decay constant of Rn, m – the mass of the material sample [kg] and t_1 – a time of the build-up of radon activity inside the emanation chamber from a material sample.

A relative exhalation rate R in percentage is defined as the ratio of the measured radon activity exhaled from a sample to the total radon activity emanating from it.

$$R = \frac{N}{N_1} 100\% \tag{5}$$

The mass exhalation rate E_m [$Bq \cdot kg^{-1} \cdot h^{-1}$] may be calculated by equation 6:

$$E_m = \frac{N \cdot \lambda_1}{m \cdot (1 - \exp(-\lambda_1 \cdot t_1))} \tag{6}$$

And the surface exhalation rate E_s [$Bq \cdot m^{-2} \cdot h^{-1}$] is expressed as followes:

$$E_s = \frac{N \cdot \lambda_1}{s \cdot (1 - \exp(-\lambda_1 \cdot t_1))} \tag{7}$$

where *m* and *s* are the mass [kg] and the area [m^2] of the sample and the rest of symbols are the same as above.

X-7.2. Analytical methods

For the determination of the emanation rates, a chamber with the dimensions of 20 × 20 × 25 cm (10 dm^3) was used. A sample with a volume of about 0.5 dm^3 is hermetically enclosed in the chamber. Depending on the kind of a investigation, the time the sampling time ranges from a few days to 30 days. This time period is selected so as to ensure enough Rn being emanated. Then a gas sample of of 100 cm^3 is extracted from the emanation chamber through a valve into a syringe. The gas sample is mixed with 40 cm^3 of a liquid scintillator in a glass tube of 2 cm diameter and 25 cm length. Next, the scintillator mixture is divided into counting vials of 22 cm^3 volume and the count rates are determined by α/β liquid scintillation counter3 hours after the extraction of the gas sample. The contents of ^{226}Ra in the samples is determined by a gamma-ray detection system with a NaI(Tl) scintillation crystal or a semi-conductor detector. The radioactive equilibrium between ^{226}Ra and ^{214}Bi was maintained by tight sealing of the samples and keeping them for 32 days so that radon could not escape.

X-8. POTENTIAL LEACHIBILITY OF RADIUM ISOTOPES

X-8.1. The problem

Under ambient environmental conditions, generally radioactive residues associated with uranium deposits appear to be highly stable and very insoluble. However, radium co-precipitated in a sulfate matrix is not thermodynamically stable under reducing conditions and it could be released to the environment.

The question of leachability of radium isotopes from contaminated materials exposed to acid-rain and other ageing processes is generally unknown. The leachability of radium from contaminated soil was investigated using fluids having wide range of pH, reflecting different and more extreme chemical parameters.

X-8.2. Leach tests

An aliquot of approximately 50 g of contaminated soil was mixed with an extraction fluid at a 1: 20 solid-to-liquid ratio for approximately 72 h according to the ASTM D5284-93 standard method for sequential batch extraction [X-9]. The soil and extraction fluid were mixed together in 1 lpolypropylene bottles that were tumbled end-over-end throughout the extraction period using a tumble mixer. After mixing, the extraction fluid was filtered through a 0.45 μm pore size membrane filter. Then the radium contained the extracted fluid was precipitated and measure by liquid scintillation counter. The procedure of sample preparation procedure and measurement of radium isotopes is described in more detail elsewhere [X-10].

The leaching potential is defined as the percentage of the original ^{226}Ra content of the sample measured in the leaching solution after contact with the soil for a period of 72 h.

A procedure to simulate natural leaching conditions was devised: the Ra-content of prepared samples were measured , then these samples were buried at a depth of 0.5 m in a natural environment. To this end, all samples to be tested were transported back to the Kowary Podgórze village, to a bury place close to the uranium mine and tailings deposit. This place was located on the slope of the existing dumps, containing uranium minerals. The samples were left for a period of 10 months, i.e. through winter-spring- summer. After this period of time, the samples were dug up, cleaned, dried and measured again for their Ra-content.

X-9. ANALYTICAL RESULTS

X-9.1. Mineralogical composition of the residues

Macroscopically, two different types of waste have been identified. The first type is a sandy fraction (below 2 mm) located within an internal part the waste disposal site. The second type of waste has been found on the southern slope of the tailing dam. It is coarse grained fraction, probably obtained at mineral plant of the former iron mine. In comparison to the other type of waste, this waste is characterized by higher amounts of magnetite and haematite. A full list of the minerals identified is given in Table X-3.

Table X-3. Mineralogical composition of the residues (minerals in bold are responsible for cementation)

Major Minerals	**Minor Minerals**
quartz	magnetite
K-feldspar	**haematite**
plagioclase	**goethite**
biotite	**pyrite**
muscovite	other sulphides
chlorite	baryte
amphibolite group	rutile
clay minerals	anatase
calcite	sphene
dolomite	zircon
epidote	
fluorite	
garnet	artificial phases, glass and slag
gypsum	

During microscopic observation evidence for pyrite and magnetite oxidation has been found. It is clear that the more recently deposited tailings can be treated as a classic sediment and that the well-established concepts of diagenesis can be applied to them. The role of diagenetic processes is very important for planning of long-term stabilization of waste disposal facilities. In the course diagenetic processes, free uranium can be easily incorporated into iron hydroxides and adsorbed on their active surface. Decomposition of feldspars will play also an important role. Free oxygen is responsible for an initiation of pyrite oxidation in the residues. The free sulphuric acid produced during this reaction may lead to the decomposition of the feldspars. As a result of feldspars decomposition, new clay minerals will be precipitated, which then can adsorb free uranyl anions.

Two types of materials deposited in the tailing pond can be distinguished by their radionuclide composition (Tab. X-4). The first one is characterized by a lower radium activity, ranging between 3,200 – 3,820 $Bq \cdot kg^{-1}$. The total radium activity of the second type residue is by about 2,000 $Bq \cdot kg^{-1}$ higher (compare Tab. X-3).

Sample SP6 was obtained mixing sample SP1+SP2+SP3+SP4+SP5 (see Tab. X-4).

Table X-4. Radionuclide contents of different types of residues from the uranium tailing at Kowary Podgórze (by gamma spectroscopy).

Sample	^{40}K [$Bq \cdot Kg^{-1}$]	^{226}Ra [$Bq \cdot Kg^{-1}$]	^{232}Th [$Bq \cdot Kg^{-1}$]
SP1	1,330±200	3,240±100	10±10
SP2	1,420±200	3,820±100	10±10
SP3	1,290±300	5,780±200	20±10
SP4	8,70±300	5,670±200	30±10
SP5	7,70±200	3,260±100	90±10
SP6	1,030±500	4,210±300	6±6

X-9.2. The mineralogy of 'room temperature ceramics'

Natural materials have limited application to the physical stabilization of waste because of poor knowledge of in-site reaction. Study and selection of materials is required before application. Reaction with a natural substances are running relatively slowly, however when the processes are completed achievements in litification are much better than other artificial relevant. Some natural cements and their characteristic are presented in the Table X-5.

X-9.3. Physical parameters of some samples used in the experiment

Some geotechnical and hydraulic parameters relevant for the stabilisation and fixation of the sandy fraction of the residueshave been determined (Tabs. X-5 and X-6). All measurements were carried out by Dr. J. Mazurek from the Dept. of Geomechanics, Civil Engineering and Geotechnics, AGH Kraków, Poland.

The experimental sample (SP6) mixed with modified anhydrite showed low compressive strength in comparison to materials normally used for construction. After water absorption capacity measurements, all these samples were damaged. However, they cannot be neglected as a cements for the waste fixation. Field test showed that these samples hold well their properties after 10 month of storage in the local environment. The local environment at the chosen testing place is characterized by a wide temperature range (from +35°C to-35°C), high annual precipitation rate (1500 mm/y), and slope angle steeper than 10°. Physical parameter measurements showed also that the way of preparaing the samples plays an important role.

Parameters such as porosity, compressive strength, and Young modulus were basis of sample selection, however further experiments with bigger sample sizes are needed.

Table X-5. Characteristics of different natural materials that could be used for the long-term stabilization and isolation of low-level uranium residues.

Material	Advantages	Disadvantages
clays: kaolinite, illite, bentonite montmorillonite, mixed phases (smectites)	• very good sorption capacity • good water retention • simple technology of application. • very often occur in the big quantity in the nature • low price	• mono-mineralic clays are rare in nature, • possible desorption • changes during diagenesis • -chemical modification may be required
carbonates: calcite, dolomite, magnesite, siderite	• pH buffering, • very common in nature • reasonable price • cementation effects • useful for ARD treatment	• not useful in the alkaline treatment processes • grinding is required
sulfates: gypsum, anhydrite, barytes	• γ-shielding (barytes) • Rn-shilding (gypsum, $CaSO_4 \times \boldsymbol{0.5\ H_2O}$) • conversion of anhydrite to gypsum • occur in good quality and quantity in nature • reasonable price	• reconversion of gypsum to anhydrite in the presence of NaCl-waters • grinding is required
bauxite	• high sorption parameters • occurs in a few areas only • reasonable price	• limited occurence
diatomites	• very high sorption capacity • low density • reasonable price	• very rare in nature
Fe-hydroxides	• very high sorption capacity • occur in a few areas only • reasonable price	• modification may be required
red soil	• high sorption capacity • occurs in some areas	
zeolites	• very high sorption capacity	• grinding is required • rare in nature
hydroxyl apatite	• very high sorption capacity • expensive	• grinding is required • very rare occur in nature in high quantity
quartz: gravel and sand	• good drainage • occurs only at some places • reasonable price	• sorting/grading is required
carbonate gravel	• drainage • pH buffer	• useful against ARD

X-9.4. Radon exhalation rates from conditioning mixtures

Calculated values of the relative, mass, and surface exhalation rates of the samples taken from the uranium tailing piles are presented in Table X-8. These results show that samples have relatively small exhalation rate E. Among the tested materials, mixtures of SP6 with natural cement (anhydrite, gypsum) have the lowest exhalation rate.

Table X-6. Water absorption capacity mesurements according to polish standard pn-66/b-04101.

Sample	Mass of dry sample m [g]	Mass of saturated sample m_1 [g]	Sample volume v [cm^3]	Dry density C_o [$g \cdot cm^{-3}$]	Mass of adsorbed water n_w [%]	Volume of adsorbed water n_o [%]	Remarks
0.9 SP6 + 0.1 lime	169.60	208.78	120.19	1.41	23.10	32.60	After 72 h some disintegration was observed
0.7 SP6 + 0.3 gypsum	159.49	218.47	129.81	1.23	36.98	45.44	No change
0.8 SP6 + 0.2 gypsum	162.85	215.08	123.63	1.32	32.07	42.25	sample broke at ½ of its height. The depth of fissure was ½ of sample diameter. Some additional disintegration was observed.
0.9 SP6 + 0.1 cement	190.67	228.21	121.29	1.57	19.69	30.95	No change
0.8 SP6 + 0.2 cement	197.59	237.18	124.25	1.59	20.04	31.86	No change
0.7 SP6 + 0.3 cement	200.22	243.75	125.96	1.59	21.74	34.56	No change

Table X-7. Geotechnical parameters of some samples used in the experiment

Sample	Height h [mm]	Everage surface S [mm^2]	Compressive strength Rc [Mpa]	Young modulus E [Mpa]	Remarks
0.9 SP6 + 0.1 lime	49.20	2397.87	0.60	30	Irregular shape, some grains were crumbling away
0.7 SP6 + 0.3 gypsum	50.50	2643.85	0.47	30	Regular shape, some grains were crumbling away
0.8 SP6 + 0.2 gypsum	51.70	2488.69	0.28	17	Irregular shape, some fissures were observed, and some grains were crumbling away
0.9 SP6 + 0.1 cement	49.35	2457.65	1.55	102	Regular shape
0.8 SP6 + 0.2 cement	49.75	2487.52	4.87	450	Regular shape
0.7 SP6 + 0.3 cement	49.85	2535.08	11.41	1320	Regular shape

Table X-8. Relative, mass and surface exhalation rates measured for sample SP6 and various admixtures of binders to this material.

Sample	**Mass of sample (σ)** [g]	**^{226}Ra (σ)** [$Bq\cdot kg^{-1}$]	**R (σ)** [%]	**E_m (σ)** [$Bq\cdot kg^{-1}\cdot h^{-1}$]	**E_s (σ)** [$Bq\cdot m^{-2}\cdot h^{-1}$]
SP6	904.22 (5)	4210 (80)	10.95 (6)	3.48 (18)	147 (15)
0.9 SP6 + 0.1 cement	998.39 (5)	3800 (70)	15.3 (8)	4.39 (22)	243 (12)
0.7 SP6 + 0.3 cement	1046.05 (5)	2950 (80)	17.1 (9)	3.81 (17)	211 (10)
0.9 SP6 + 0.1 anhydrite	959.45 (5)	3790 (70)	11.76 (8)	3.36 (11)	186 (25)
0.7 SP6 + 0.3 anhydrite	975.9 (5)	2950 (90)	4.29 (27)	0.83 (5)	46.1 (22)
0.9 SP6 + 0.1 gypsum	885.80 (5)	2966(100)	9.00 (6)	2.01 (11)	112 (6)
0.7 SP6 + 0.3 gypsum	861.41 (5)	2840 (80)	1.04 (4)	0.223 (2)	12.3 (7)

σ – standard deviation

X-9.5. Results of leach tests

The quantity of ^{226}Ra leached from the SP6 sample depends on chemical composition of solvent used in the experiment. In the Table VI are shown results of leaching test. Leaching potential of normal water is very low, but increasing of NaCl content in the water can rapidly change condition of the leaching (Tab. X-6).

Table X-9. Results of ^{226}Ra leaching from samples of low-waste uranium using different extraction fluids.

Sample	**Extraction fluid**	**Leaching potential** [%]	**Specific activity** [$Bq\cdot kg^{-1}$]
SP6	water	0.16	4100
SP6	Water + NaCl (100 $g\cdot dm^{-3}$)	4.8	
SP6	Water + Na_2S (1 $g\cdot dm^{-3}$)	0.07	

X-9.6. Results of experiments simulating natural leaching conditions

The ^{226}Ra activity of samples after 10 month burial in the Kowary Podgórze environment (close to the uranium residues dump) is reduced by 0.6% to 6.2% compared to activities before burying (Tab. X-10). The data suggest a small loss of radium over the period of 10 months, but the measured differences are only slightly above the statistical errors. It suggests that in the testing environment with a pH similar to the one measured in the tailing pond, radium can be trapped within the residues cemented by selected natural cements.

The experiment showed that the natural cements (calcium hydroxide can be replaced by calcite or limestone) could play a significant role in the stabilisation of radionuclides in the system proposed for a new milling residue storage facility (see Table X-11).

Table X-10. ^{226}Ra, ^{232}Th specific activity of selected materials prepared on the base of materials from Kowary tailing pond.

Sample	Specific Activity (σ) [Bq/kg]	
	^{226}Ra	**^{223}Th**
SP6	4170 (80)	6 (6)
0.70 SP6 + 0.30 cement	2581 (80)	21 (8)
0.70 SP6 + 0.30 gypsum	2840 (90)	13 (10)
0.70 SP6 + 0.30 anhydrite	2950 (90)	12 (10)
Anhydrite	9 (2)	2 (2)
Hydrated lime	19 (3)	2 (2)
Gypsum	8 (2)	2 (2)
Cement pure	73 (6)	30 (3)
0.60 SP6 + 0.40 anhydrite	2440 (50)	15.5 (10)
0.90 SP6 + 0.10 cement	3750 (70)	25 (8)

σ- standard deviation

Table X-11. Change of specific ^{226}Ra activities of the selected materials after 10 month deposition underground.

Sample	Specific ^{226}Ra activity (σ) [Bq/kg]		Activity ratio
	before deposition	**after deposition**	**(σ)**
0.7 SP6 + 0.3 cement	2581 (80)[a]	2461 (50)	0.953 (36)
0.7 SP6 + 0.3 gypsum	2840 (90)[a]	2799 (56)	0.979 (35)
0.7 SP6 + 0.3 anhydrite	2950 (90)[a]	2932 (58)	0.993 (34)
0.9 SP6 + 0.1 cement	3750 (70)[a]	3566 (70)	0.942 (37)
0.9 SP6 + 0.1 calcium hydroxide	3750 (70)	3610 (72)	0.962 (35)
0.9 SP6 + 0.1 anhydrite	3750 (70)	3750 (73)	0.981 (35)
0.8 SP6 + 0.2 gypsum	3340 (65)	3277 (65)	0.997 (34)
0.8 SP6 + 0.2 anhydrite	3340 (65)	3240 (63)	0.970 (34)
0.9 SP6 + 0.1 cement	3810 (70)	3570 (72)	0.942 (37)
0.8 SP6 + 0.2 cement	3350 (65)	3240 (65)	0.967 (34)

[a]calculated on the base of material content; σ – standard uncertainty

X-10. DISCUSSION

Investigation of the mechanical properties of some natural 'ceramics' used in the experiments showed that they are good materials for fixation and solidification of residues. They can also play an important role in the chemical stabilisation. The advantage of the natural materials is that they can be easily assimilated in the environment, and their porosity is lower compared to those samples treated with Portland cement. A decrease in of the porosity of the waste pile reduces the radon flux. Moreover, all residues contain various clays, and such material cannot be cemented with Portland cement. Natural cements should be selected by their mineralogical composition and considering the geochemical properties of the waste. The active membranes proposed, especially those containing lime or calcite, would stabilize the pH-values within the proposed new waste disposal facility. Natural zeolites, which have not been investigated experimentally because of lack of time, could also be considered for the construction of bottom and top isolation membranes (see also table X-5).

Leaching and field tests showed that natural cements can be used for trapping radionuclides within the the disposal facility. A mixture of modified anhydrite powder with gypsum and hydrated lime can be proposed for the construction of the internal, active membranes (cf. Fig. X-2). However, the final composition of the active membrane should be selected to fit the mineralogical composition of the residues. The proposed mineralogical membrane could actively react with both fluids and residues. A chemical fixation would only be successful, if the system of proposed mineral membranes can create a geochemical equilibrium within the waste pile.

X-11. SUMMARY AND CONCLUSION

Room temperature ceramics (natural cements) have been tested in the laboratory for both cementation capabilities of milling residues and effectiveness in reducing the radon emanation. Positive results of litification obtained using mixtures of modified anhydrite powder (typical used in construction), anhydrite powder mixed with Portland cement, anhydrite powder mixed with calcium hydroxide with residues (SP6) from the Kowary tailing pond. Some physical measurements, ^{226}Ra activity, Radon flux and leaching test have been undertaken with such prepared testing samples. Measurements of physical parameters showed that small admixtures of natural cements (ceramics) could easily fix the sandy and clay fraction of the residues.

The new environment in the place of the new storage for the waste is also a critical stage of the experiment. After litification of active membranes within the new waste disposal, the control and modification of pH and Eh will be very limited.

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ANNEX XI. RUSSIAN FEDERATION

POLYMERIC COATS FOR THE STABILISATION OF CONTAMINATED SURFACES

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XI-1. ABSTRACT

VNIINM has been developing remediation techniques for contaminated sites as a result of nuclear accidents since 1986. An approach, based on basic science approach (radiochemistry, radionuclide migration, soil science, synthesis of polymers, etc.), was used to develop specialised technologies for the remediation of contaminated sites. This approach uses synthetic polymers to sequester radionuclides. Ecologically safe polymers and procedures for soil decontaminations were developed and tested. This technology also fosters the formation of a protective grass cover by providing structure formers.

The new polymeric compositions prevent soil erosion by water and wind with an efficiency over 95 % and a 1-3 year stability to atmospheric influences. Techniques for laboratory and field testing were developed; water and wind erosion test of soil with various polymeric structures are carried out.

XI-2. INTRODUCTION

Dispersion of radioactive substances from the surface of tailings ponds and waste rock piles due to water and wind erosion is a significant problem faced by the relevant industries.

Disposal, long-term stabilisation, and isolation of uranium mill tailings requires the development of adequate technologies. The effect of atmospheric factors (rainfall, snow-melt, and wind) on contaminated areas can cause secondary contamination of the environment. Currently, with a view to long-term stabilisation of such territories, multibarrier concepts have been developed that genrally consist of covers made up of crushed rock, sand, and soil.

The project suggests applying supplementary protection against water and wind erosion, namely polymeric structure formers based on interpolyelectrolite complexes (IPEC). These polymers have been successfully tried and applied on an industrially scale to suppress radioactive dust generation after the Chernobyl accident.

Tailing pulps usually consists of solid particles in the 0.225-0.1 mm size range, while particles with weighted mean size of 0.08 mm are typical tailings beach areas. The finely divided materials (<10 μm) are readily swept away great distances as aerosols at an initial lift rate of 3-5 m/s to cause airborne contamination, with air particle concentrations exceeding standards by tens of times. This presents a serious inhalation hazard. The polymers are able to resist the action of natural factors on top soils, thus providing a supplementary protective barrier.

XI-3. ACTIVITIES UNDER THE PROJECT

The following main activities were undertaken in this project:

(1) Analysis of existing means of protection and remediation, polymeric preparations used, depending on natural factors, soil types and types of contamination;
(1) Selection of a method of protecting contaminated surfaces with polymeric coatings as a function of the given conditions;
(2) Development of laboratory testing procedures;
(3) Testing of polymers with real and surrogate waste materials;
(4) Research on the effect of polymer coatings on to development of grass vegetation;
(5) Recommendations for applying polymer covers to uranium mill tailings.

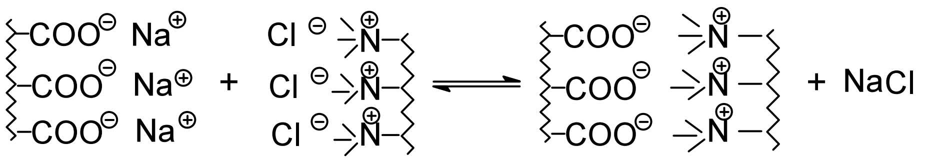
XI-4. POLYMERS AND THEIR APPLICATION

XI-4.1. Synthesis of polymers

It is well known that the bulk of radioactivity is accumulated on the fine particles and the organic fraction of geological materials. This size fraction is found to be the most susceptible to dust generation and erosion

One possible way of stabilisation/fixation is to create additional bonds based on water-insoluble high molecular weight compounds. Similar compounds (trade marks MM-1, MJ-1) based on interpoly-electrolyte complexes (IPEC) have been developed by VNIINM and Moscow State University (MSU) to immobilize radioactivity captured by soils following the Chernobyl accident (1986-1993), and for another contaminated site remediation purposes.

IPECs are the products of interactions between oppositely charged macromolecules [XI-1]. The interpolyelectrolyte reaction proceeds by the following mechanism:



(XI-1)

IPECs can be considered as intelligent (smart) materials, due to their ability to adapt themselves to complex structures of dispersed systems via rapid exchange processes and to attain the optimal set of bonds with different colloidal particles and surfaces. A new generation of IPEC structure formers with microgels (MLA-1, MLA-2) was developed in VNIINM and MSU. They have not only a high efficiency in fine particle stabilisation, but also high ability to support grass vegetation during remediation activities.

XI-4.2. The soil-polymer layer formation

The technology for preparing and applying IPECs is quite simple. It involves (i) the preparation of dilute aqueous polyelectrolyte mixtures where ionic interactions between oppositely charged polyions are completely suppressed. (ii) the application of these aqueous solutions as dispersed systems usually by spraying; (iii) washing of the dispersed systems by water in order to remove mineral salts. The commercially made synthetic and natural constituents are non-toxic [XI-2-XI-9].

The main objective of IPEC application is a good interaction between the small negative charges on mineral surfaces (resulting from the dissociation of silanol Si-OH groups) and the polycation forming part of IPEC. The joint application of polications and polyanions result in formation of long-term stable surface-polymer complex (Fig. XI-1).

The IPEC treatment of soils produces a polymer coating 3-6 mm thick, which tightly binds the fine grains to each other by way of aggregation of small particles and by coagulation of soil colloids.

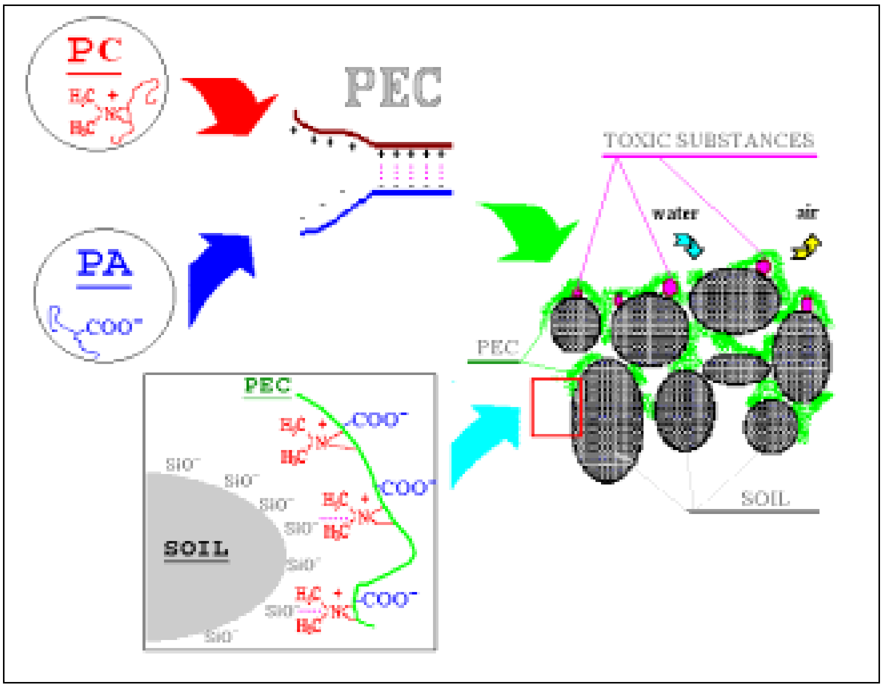


Fig. XI-59. Formation of protective polymer coatings.

XI-5. LABORATORY TESTS

XI-5.1. Samples

During the period of 2001 – 2003 the polymeric structure formers were successfully tested under laboratory conditions and in the field with material from a mill tailings beach of the Ulba Metallurgical Plant (Kazakhstan, see Annex VII), and in the laboratory with surrogate tailing materials from Zovtny Vody (Ukraine, see Annex XII).

XI-5.2. Characterisation of Ulba samples.

For the tests we used actual samples from Ulba. It was loose material of 5.0 kg in mass. The initial moisture content was 18%. Prior to the investigations the material was dried to the air-dry conditions. The results of the sieve analysis are summarized in Table XI-1 and an. external view of the samples is given in Fig. XI-2.

The chemical composition and and the grain-size distribution of actual samples from Zovtny Vody and the surrogate material are given in Tables XI-2 and XI-3.

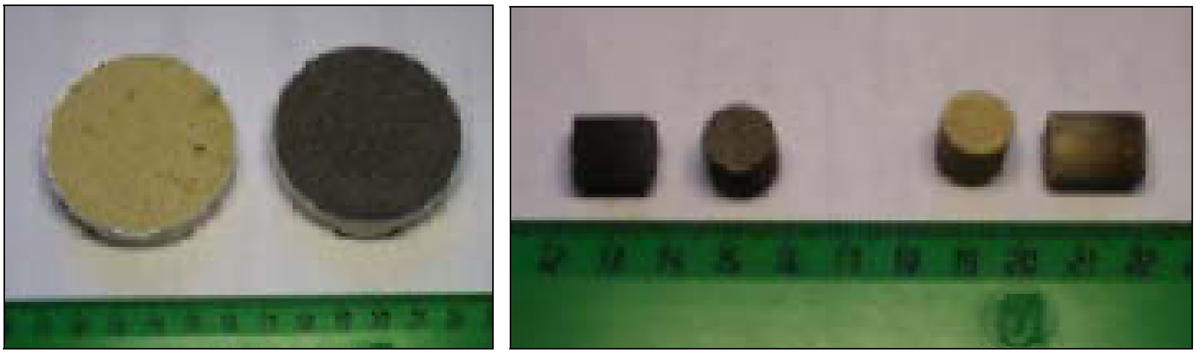


Fig. XI-60. View of Ulba tailings preparing for laboratory tests.

Table XI-20. Grain size distribution of tailings from Ulba Metallurgical Plant.

Sizes class [mm]	Content [%]
>1	1.9
1 >0.63	9.4
0.63 >0.4	10.9
0.4 >0.315	9
0.315 >0.2	19.1
0.2 >0.1	38.2
0.1 > 0	11.5
Total:	100.0

Table XI-21. Chemical composition of Zovtny Vody model samples [solid wt %].

Composition	Actual	Model	Composition	Actual	Model
SiO_2	59,3÷64,0	62.0	ZrO	0,10÷9,13	5.8
Fe_2O_3	2,4÷3,84	2.7	PbO	0,010÷0,012	0.0
FeO	0,58÷1,67	0.9	CaO	2,0÷3,26	2.6
MgO	0,87÷1,82	0.0	TiO_2	0,29÷0,37	0.3
Na_2O	7,87÷11,6	10.1	Al_2O_3	15,38÷16,8	15.6
K_2O	0,48÷0,8	0.0	P_2O_5	0,17÷0,40	0.0
S_{total}	0,04÷0,09		pH	8÷9	

Table XI-22. Grain size distribution of actual Zovtny Vody and model samples.

	Size class [mm]					
Content [wt %]	**>0.25**	**0.25 >0.14**	**0.14 >0.10**	**0.10 >0.074**	**0.074 >0.04**	**>0.04**
actual	1.0	5.4	8.7	9.8	15.3	59.8
model	0.95	5.6	8.5	10.1	14.1	60.75

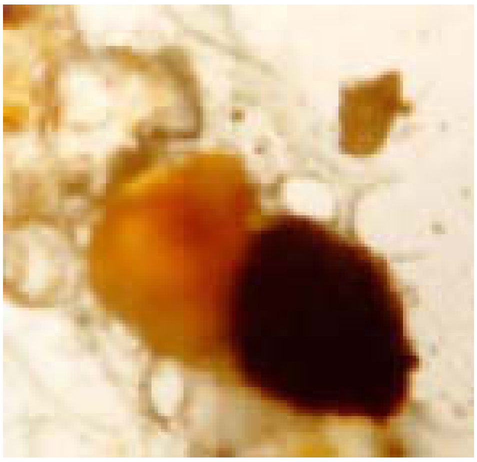


Fig. XI-61. Soil particles plus polymeric film.

Fig. XI-62. Particles conglutination by polymer

Fig. XI-63. Long polymeric bridge with particles.

Fig. XI-64. Glueing of small particles onto big ones by polymer.

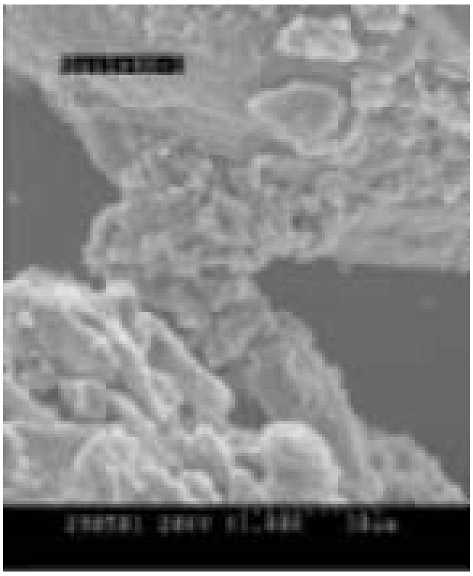


Fig. XI-65. Electron microphotograph.

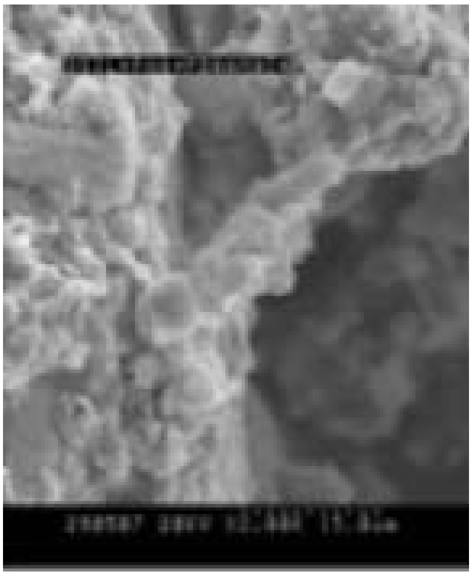


Fig. XI-66. Electron microphotograph.

XI-5.3. Structure of Polymeric crusts

The microstructure of protective soil-polymer layers has been studied by light microscopy. Fine particles are attached to each other as a function of the polymer nature and amount:

(1) by a solid polymeric film (Fig. XI-3). Whatever its size, each sandy particle is enveloped with the polymeric film that also fills the spaces between them to form a solid thick waterproof layer (this would require a 5-10 fold greater amount of polymer compared to the 1 l/m^2 dosage of IPEC needed otherwise);

(2) at points of particle contact (Fig. XI-4). Spaces between particles are free, allowing the movement of water and air;

(3) by thin bridges (Figs. XI-5 and XI-7), when particles of different size are separated by a considerable distance (from 1 mm to 1-2 mm);

(4) due to small-size particles (5-20 mm) sticking to larger ones(over 700 mm) (Figs. XI-6 and XI-8). This occurs during the evaporation of the polymeric solution covering the particles. The fine particles are picked up by the solution from interspaces and attached and attracted by large particles.

The soil particles are attached to each other only at intergranular contacts or by polymeric bridges. Owing to this, the IPEC-treated soil remains penetrable to moisture and air. The bonding decreases the wind and water erosion of small particles on which the bulk of the radioactivity is located. In addition, because the IPEC have free functional groups, it has been found to form strong complexes with aqueous radionuclides species.

XI-5.4. Wind erosion of contaminated surfaces

XI-5.4.1. Wind tunnel tests

Treated samples were subject to erosion test in a wind tunnel. The facility (Figs. XI-9 and XI-10) consists of a stainless steel (1Kh18N10T) body which encloses removable containers of different size that are to be filled with the samples to be tested, an air filter to trap particles in the air supply, a dust collector for the particles eroded from the sample surface, and a vacuum pump (NT 700 KARCHER with a maximum output of 94 l/s at 218 mbar).

Samples were placed in the rectangular container, rolled smooth and slightly compressed. The density of all specimens was a constant 1.50 g/cm^3. The samples then were exposed to an air stream over a velocity range from 10.0 to 40.0 m/s in the wind tunnel.

XI-5.4.2. Experiment № 0 - control sample.

An untreated sample was exposed for 10 min in the wind-tunnel facility to an air flow with various velocities between 1.74 and 10.2 m/s. The loss of mass in the course of the experiment was 108.5 g (Figs. XI-11 and XI-12).

Intensive erosion already began at a velocity of 6.38 m/s and after increasing the velocities in increments up to 10.2 m/s the material was almost completely carried off from the cell. This is an evidence for the low wind erosion resistance of untreated samples.

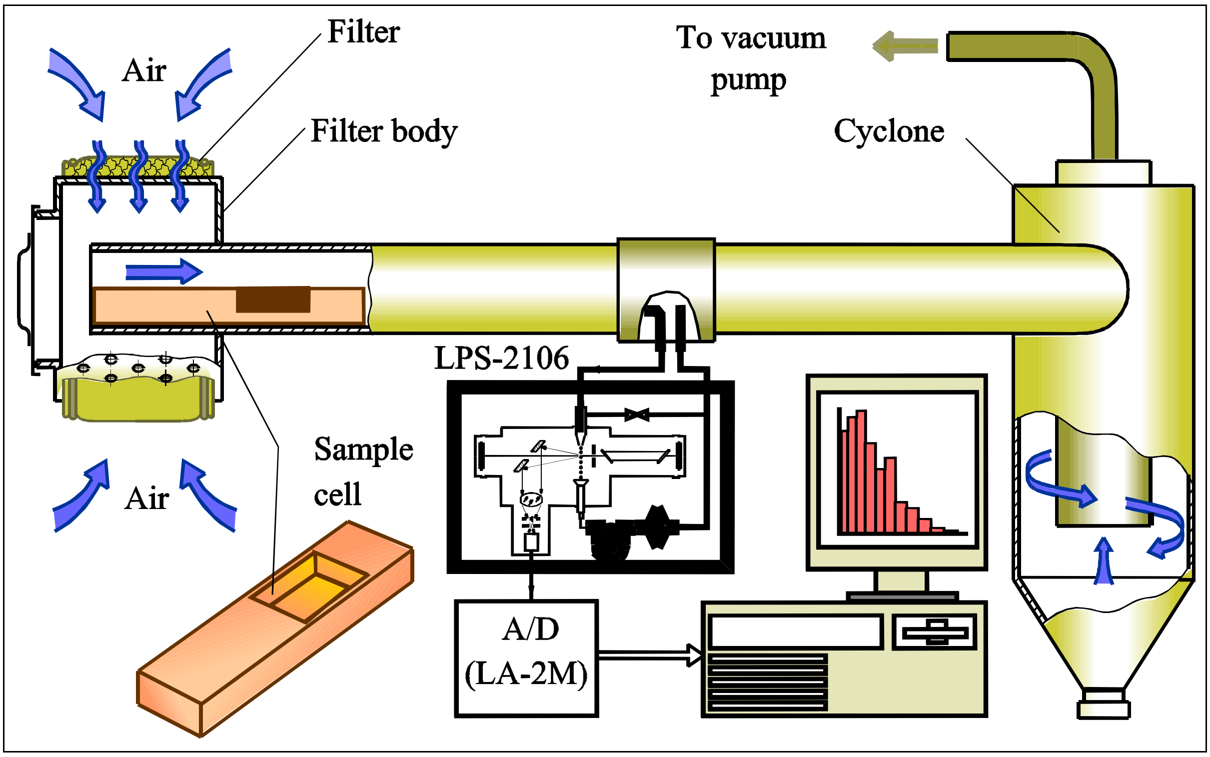


Fig. XI-67. Schematic diagram of the wind tunnel.

Fig. XI-68. View of the wind tunnel.

Fig. XI-69. View of untreated sample before experiment

Fig. XI-70. Appearance of untreated sample after experiment.

Fig. XI-71. View of sample moistened with distilled water and treated with MJ-1, before experiment

Fig. XI-72. View of sample moistened with distilled water and treated with MJ-,1 after experiment

XI-5.4.3. Results from treated samples

The generation of aerosols in the 0.3-10 μm size range was studied in the wind tunnel for a velocity range between 7.5 – 40.0 m/s. The results suggest that within the limits of experimental accuracy and instrument range no erosion occurs at the wind speeds tested. The maximum erosopm is not more than 0.05 g/cm^2, i.e. 60-100 times less than that for untreated samples.

The results indicate that essentially no aerosols are produced over an intact protective coating, but a sharp increase in their amount when the protective cover is demaged. On the strength of the tests, we can conclude that the dust suppression technique developed is effective for wind speeds as high as 40.0 m/s.

XI-5.4.4. Experiment 4 - simulation of treatment condition for real tailings.

A cell is filled with a sample dried to air-dry condition and impregnated with distilled water from the bottom up by immersion into a vessel (simulated conditions of moist tailings beaches). After that the moistened surface was treated with MJ-1 at a dosage of of 2.0 l/m^2. The sample was dried at 30ºC to an air-dry state. The cell then was exposed in the wind-tunnel to air flows at a velocity of up to 45.5 and up to 70.5 m/s (Figs. XI-13 and XI-14).

The experiment did not result in a destruction of the sample. The aerosol concentrations in the air flow did not exceed 1-8 mg/m^3. The loss of the mass in the course of the experiment was 1.5 g. The experiment has demonstrated the high erosion resistance of the protective coating that is formed by the polymer MJ-1.

XI-5.5. Water Erosion tests

XI-5.5.1. Theoretical considerations

Water erosion is a combination of fine particles detachment, transfer and deposition by surface run-off and interflow. For mill tailings two sources of run-off are coneivable: rain water or snow-melt.

The water erosion can be quantified in terms of a material removal rate, expressed as its weight removed from a unit area (typically ton/ha) or as a thickness of the layer lost (typically mm/year). Erosion resistance is quantitatively expressed in terms of the scouring erosion rate ($V\Delta_{pw}$):

$$V\Delta_{pw} = 1{,}55 \cdot \sqrt{\frac{g}{\rho_o n'}\left[\left(1-\frac{P}{100}-\right)\overline{d}_w(\rho-\rho_o\ )\right.} \qquad \text{(XI-2)}$$

where g acceleration of gravity [m/s];
n' coefficient, taking into consideration the pulsating character of velocities. This coefficient is equal to 2.28 for flow in rill channel and to 1.46 for flow in a laboratory channel;
ρ, ρ_0 specific gravity of the soil and water respectively [g/cm^3];

It follows from the equation that the erosion rate depends on $\overline{d_w}$ the weighted mean size of water-resistant aggregates.

XI-5.5.2. Hydrodynamic test channel

The stainless steel (1Kh18N10T) hydrodynamic test channel (Figs. XI-15 and X-16) consists of the plenum chamber, a sample container, shutter, water gauge, flow meter, an effluent collector, and a collector for eroded soil. Tests were conducted on soil samples of 0.02 m in depth, 0.7 × 0.1 m^2 in area and weighing 0.78 kg. The water flow rate within the channel was Q = 0.3 l/s at H (the water depth over a sample) = 20 mm and W (free cross sectional area of water over the tray)= 0.3 m.

The facilities were subjected to comprehensive acceptance tests.

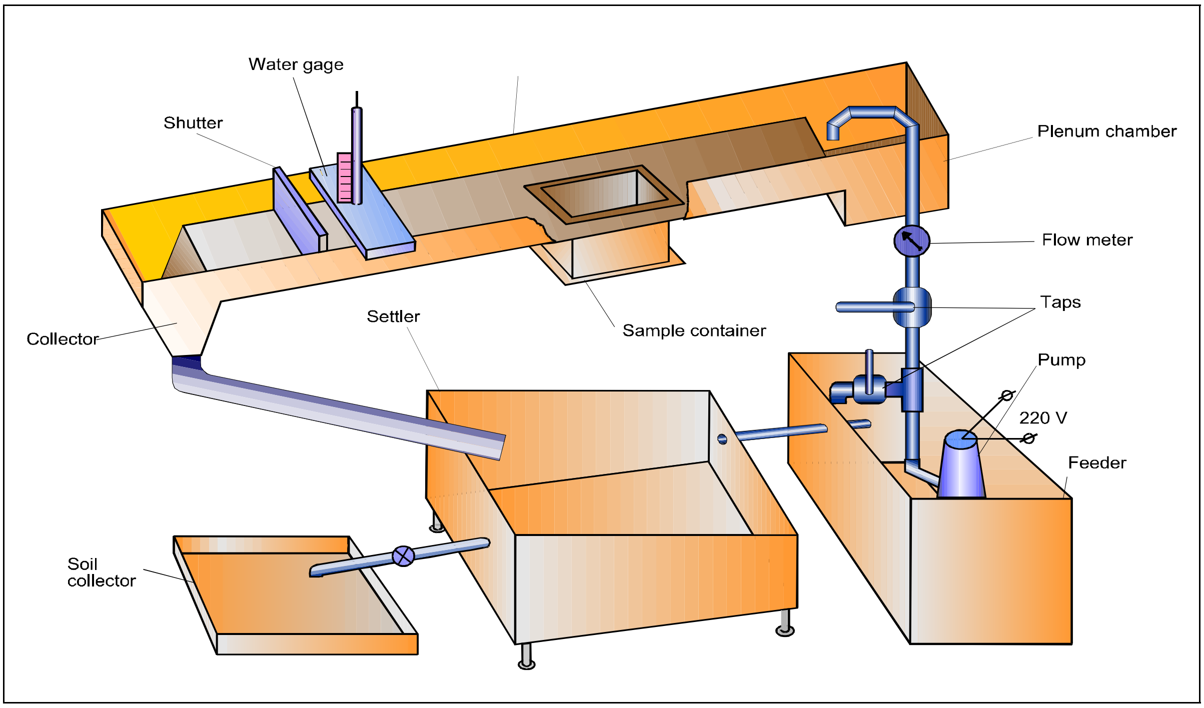


Fig. XI-73. Schematic of the hydrodynamic test channel.

Fig. XI-74. View of hydrodynamic test channel.

XI-5.6. Sample preparation and experimental conditions

The fine materials were treated with 1.0, 2.0 and 4.0 l/m^2 IPEC water solution. The other experimental conditions are summarised in Table XI-4.

Table XI-23. Experimental conditions for water erosion tests

Duration	6 hours
Velocities of water flow in hydrodynamic facility	0.10-1.50 m/s
Water flow height in hydrodynamic testing facility	0.01 m
Surface area of sample loaded into cell	0,02 m^2
Mass of sample loaded into the cell	130 g
Dosage of agent	4,0 l/m^2

XI-5.6.1. Results

Tables XI-5 and XI-6 summarise the experimental results with MM-1 IPEC formulation. It is evident from these results that the erosion resistance of Ulba tailings samples is substantially improved by the surface treatment with polymers. The erosion threshold for untreated samples is at a flow velocity of around 0.50-0.60 m/s. After treating samples with IPEC, the erosion threshold increases to 0.80 – 0.90 m/s flow velocity and more. This is explained by larger size aggregates formed after IPEC treatment; they are mechanically more resistant to water erosion. It is noted, that both the polymeric agents shown almost identical effectiveness.

Table XI-24. Erosion resistance of samples treated with MM-1.

Water flow velocity [m/s]	**Amount of material eroded P [g]**		**Effectivenes of polymeric coating $K=P_c/P_{IPEC}$**
	Control (without treatment) P_c	**MM-1 P_{IPEC}**	
0.10	0.7	<0.5	1.4
0.28	1.8	<0.5	3.6
0.40	1.5	1.6	0.94
0.50	2.6	1.4	1.86
0.67	8.7	2.3	3.78
0.83	>40	3.6	>11
1.20	-	15.7	-
1.45	-	>40	-

XI-6. Tests for mechanical strength

A minipenetrometer MECMESIN Compact GAUGE 200 N was used for measuring shear strength. The coatings applied to actual and surrogate samples show a high mechanical strength (25 kN/m^2 for a coating 2 mm thick, Fig. XI-17). The mechanical strength of IPEC-based coats is 1.5-2 times that of those based on individual IPEC components.

The mechanical strength of coatings exposed to rainfall of normal acidity (simulation of 20 mm of rain followed b drying) is found to be within 20.0 kN/m^2 (for an application of 1.0 l/m^2 of the polymer formulation).

Fig. XI-75. Measuring the mechanical strength.

Table XI-25. Mechanical strength of protective coatings based on IPEC formulations after irrigation with water

№	Formulation	Formulation application [l/m^2]	H_2O application [l/m^2]	Conditons	Mechanical strength [N/cm^2]
1	Control	–	50	Irrigation with water for 3 h	67.5
2	Control	–	50	Irrigation with water for 7 h	77.8
3	MLA-1	1	50	Irrigation with water for 2 h after formulation applied	37.1
4	MLA-1	1	50	Irrigation with water for 7 h after formulation applied	53.9
5	MLA-2	1	50	Irrigation with water for 2 h after formulation applied	23.4
6	MLA-2	1	50	Irrigation with water for 7 h after formulation applied	23.4
7	MJ-1	1	50	Irrigation with water for 2 h after formulation applied	103.4
8	MM-1	1	50	Irrigation with water for 2 h after formulation applied	85.2

XI-7. Revegetation with grasses

A stable perennial vegetation cover will further improve the erosion resistance of tailings impoundments. The use of IPEC to promote seeding and rooting on tailings and other contaminated soils may be contemplated.

XI-7.1. Greenhouse Experiments

To study the influence produced of IPEC components and their concentration on the growth and evolution of perennial plants used for pastures, a greenhouse experiment was carried out. Clover (*Trifolium repens*), timothy (*Phleum protenze*), Sudan grass (*Sorghum sudanense*) and mixed pasture grasses for arid conditions were used in the greenhouse experiment. IPEC components were applied to soil surfaces after sowing. The application of seeds was 16.0 g/m^2. During the whole vegetation period plastic containers with plants stayed in a greenhouse at a temperature of +23ºC. The green mass of the shoots was gathered after 68 days (Fig. XI-18).

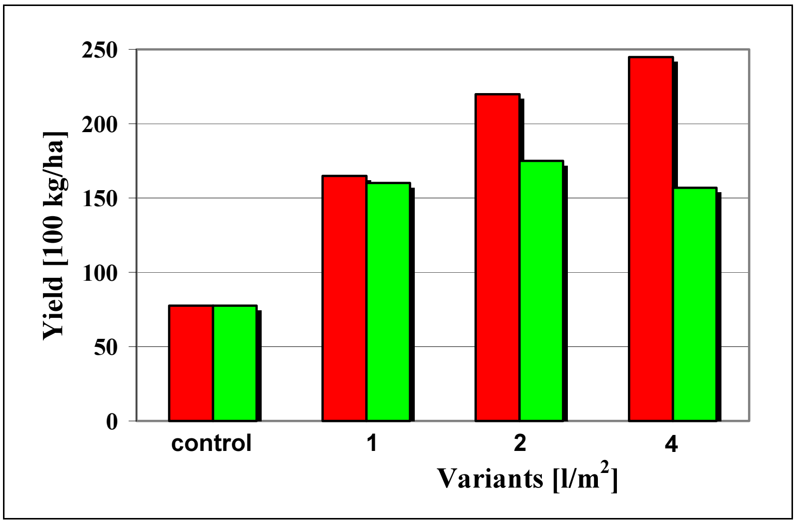


Fig. XI-76. Growing dosages of IPEC , applied to soil after seeds sowing, have rendered positive influence on plant growth and development (red column – MJ-1, green column – KNO_3)

1 2 3 4 5

Fig. XI-77. Effect of IPEC and IPEC components on the Sudan grass growth (1 – control, 2-4 – IPEC components, 5- IPEC).

When the soil was placed into containers and humidified up to 70 % from a field moisture capacity, clover, timothy, mixed lawn grass crop of 150 seeds each and 50 seeds of Sudan grass were introduced. The seeds were covered with a soil layer 5 mm thick and treated with 100 ml of solutions containing IPEC or IPEC components per container. After addition of the polymers each tank was filled with 200 ml water to promote copolymerisation of IPEC components.

XI-8. Results

The introduction of IPEC and KNO_3 substantially influenced the development of the vegetation. With the introduction of increased quantities of IPEC the biomass yield of clover and weed increased from 0.8 g/m^2 in the control to 2.45 g/m^2 (this is a 306 % increase in comparison to the control) for the highest dosage of IPEC, i.e. 4.0 l/m^2 (Fig. XI-18).

It was found that when polycations and polyanions were successively introduced in quantities of 0.5 l/m^2 of the mixture, the highest yield of plants was achieved in the experiment with dosage of 157 % in relation to the control (Fig. XI-19).

XI-9. Recommendations for application of polymeric covers for uranium mill tailings localisation

Results obtained during the project show that polymers developed on the bassis of IPECs have a high efficiency as anti-erosion agents and may be recommended for application in remediation. Application of 1.0-2.0 l/m^2 of a dilute water-polymeric compositions with polymer concentration only 2.0 wt% results a high stability of the upper layer of tailings.

The polymers may be used in the construction of multibarrier systems as efficient composition for aiding vegetation. In this case joint application of polymers and seeding grasses may be recommended following to tailings pile top cover design – erosion barrier (fig. 20). Clover (*Trifolium repens*), timothy (*Phleum protenze*), Sudan grass (*Sorghum sudanense*) and mixed grasses adapted for local conditions showed good results. The addition of nutrients may be needed for an improved vegetation process.

XI-10. SUMMARY AND CONCLUSIONS

(1) A review of the scientific and technical information on the remediation/stabilisation of uranium mill tailings indicated that erosion of the surface covers could be a significant problem. In this case the application of supplementary protection in the form of polymeric structure formers may reduce wind or water induced destruction of a surface cover.

(2) The erosion resistance of polymer-based protective coatings was assed under laboratory conditions in a wind tunnel and a hydrodynamic test channel. Samples from a mill tailings beach at Ulba Metallurgical Plant (Kazakhstan) and of simulated wastes from the Zovtny Vody Plant (Ukraine) were tested. It was shown that the application of polymeric structure formers significantly improves the wind and water erosion resistance of unconsolidated materials powder materials. The erosion threshold of wind erosion increased from 6,8 m/s for a dry untreated sample to 70.5 m/s for a sample treated with MJ -1. The water erosion resistance of the mill tailings increased by a factor of 1/5-2 (K_{eff}> 11).

(3) As result of IPEC treatment the erosion resistance even for soils unprotected by a vegetation layer increased in 1.4 times at a water flow velocity of 0.10 m/s and in 3.78 times at water flow velocity 0.67 m/s. The greatest effect estimated as increase of erosion resistance on aggregates is noted in an interval of IPEC dosages of 1.5-2.5 l/m^2. The increase of the of polymer dosage of up to 4.0 l/m^2 is possible if necessary, thus allowing to maintain high mechanical strength.

(4) It was observed that when polycation and polyanion were successively introduced at a quantity 0,5 l/m^2 of mixture the highest crop of plants was achieved in the experiment with made up 157 % in relation to the control.

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ANNEX XII. UKRAINE

RESEARCH AND DEVELOPMENT OF MEASURES TO BE TAKEN FOR LONG TERM STABILIZATION AND ISOLATION OF URANIUM MILL TAILINGS

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XII-1. Abstract

This paper contains results of research on the environmental impact of uranium mill tailings used for backfilling mined-out voids from uranium ore mining at Zhovty Vody. The general objective of the research was a comparison of qualitative and quantitative characteristics of the performance of using mill tailings as backfilling instead of the customary sand. Specifically the backfill massif strengths, the radiation impact on the mine staff, possible groundwater contamination with radionuclides from the backfill massif, and air and soil contamination with dust and gas emissions from the mine and the backfill mixing plant were investigated.

It was concluded that the use of tailings as a backfill material would be feasible, both from a technical as well as an environmental impact point of view.

XII-2. Introduction

Production of U_3O_8 in the Ukraine involves uranium ore mining and milling processes and results in various radioactive residues. The residues are stored in piles at the mine site (mining waste) and in tailings ponds (milling wastes and tailings) located in natural geomorphological depressions blocked by dams. Such residue management technology was acceptable 40 years ago, but does not meet current radiation safety requirements.

In the Ukraine the more appropriate and preferable way of waste management is the disposal in worked-out mine voids. This option significantly reduces negative impacts on the ambient atmosphere and terrestrial environments, which is particularly important for the Ukraine with its developed agriculture and fertile soils located over territories of prospective uranium mining. However, practical use of this option is not yet permissible due to the non-availability of relevant regulations and standards and a feasibility study.

Taking into account these problems, the only real option may be use of uranium mill tailings as backfill material for the uranium ore mine [XII-1][XII-2].

Production of one tonne of uranium ore entails excavation of 0.37 m^3 mine void to be backfilled depending on the choosen mining technology. For backfilling, a special mixture is used that consists of a binder, sand and water. After being placed into the mine it solidifies into a hard and dense material [XII-3]. The materials balance for the current technology is given in Figure XII.1

If the sand is replaced by mill tailings, it may be possible to utilize up to 35% of the total waste quantity arising (Fig. XII.2). In this case, the waste disposal becomes part of the mining technology, which in turn allows to undertake a feasibility study and to assess ecological concequences of the facility for the surrounding area in accordance with procedures described

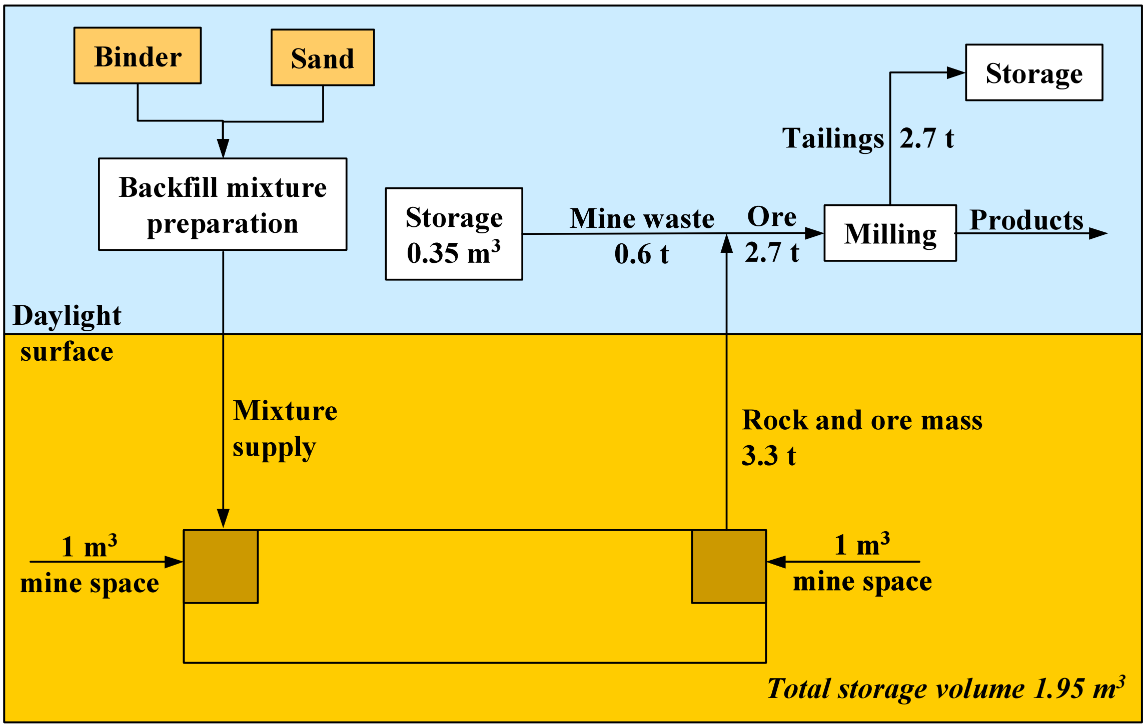


Fig. XII-1. Materials quantities for the existing mine backfilling technology.

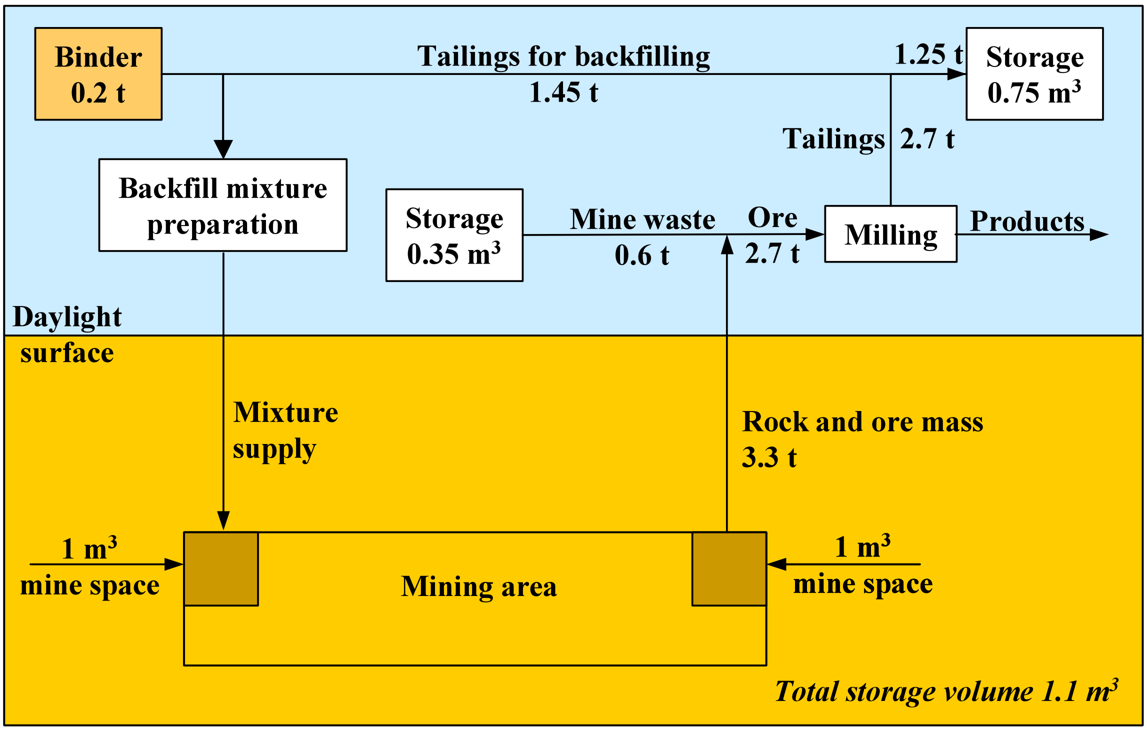


Fig. XII-2. Materials quantities for using mill tailings in the mine backfill materials.

in relevant national regulations and standards [XII-1][XII-2][XII-4][XII-5]. The material balance for such application is given in Figure XII.2.

The consequences of utilising uranium mill tailings as backfilling in mined-out voids are discussed in the report.

XII-3. Objectives

The engineering decision to replace the sand by mill tailings in the backfill material provides yet no complete isolation and long-term stabilization of uranium mining and milling wastes. However, it improves disposal conditions and completely changes the picture of environmental impact. While reducing the direct impact on surface, unfortunately at the same time it worsens the radiation situation within the mine. In addition, the permeability of the backfill materials and the leachability of chemicals and radionuclides from them may cause contamination of groundwaters [XII-4][XII-6][XII-7].

A peculiarity of the existing ore reserves is that uranium deposits with a good prospect to be developed are usually located in highly populated regions with a highly developed agriculture. The general objective of the project was to comprehensively assess the ecological consequences of a uranium mining technology utilising mill tailings as a backfill material.

The general objective of the research hence is a comparison of qualitative and quantitative characteristics of the performance of customary backfill mixtures, in particular of mill tailings instead of sand, during the mine operation.

The purpose of the research is to ensure that radionuclides released from the tailings will not reach the environment in quantities and concentrations that may cause irreversible harm to both nature and humans.

The complex ecological assessment is performed taking into account the requirements established by appropriate Ukrainian standards [XII-1][XII-2].

XII-4. Research topics

Research topics and obtained results are given in Table XII-1.

Table XII-1. Research objectives and results.

Research topics	Research results
Assurance of backfill massif strengths	The backfill composition ensuring massif strength has been determined
Radiation impact on the mine staff	Mine air contamination has been predicted; dose loading on the personnel has been calculated
Groundwater contamination with radionuclides from the backfill massif	Uranium and radium migration along groundwater flow has been predicted
Air and soil contamination with dust and gas emissions out of mine	Radon and dust volume activity variance in the surface air has been predicted

XII-5. Assessment of the geotechnical properties of backfill mixtures

When a mineral deposit is developed by heading-and-stall method, the main parameter determining the backfill massif strength is compression strength that is to be from 3 to 7 MPa after six months of solidification. According to regular technology, back-fill mixtures are made of sand with regulated grain composition. The tailings have completely different grain composition; comparison is given in Table XII.2.

Table XII-2. Grain size distribution of sand to be used in backfill mixtures and uranium mill tailings.

Grain size [mm]	Percentage	
	sand	mill tailings
5-2.5	0-20	–
2.5-1.25	5-25	–
1.25-0.63	15-25	–
0.63-0.315	35-10	–
0.315-0.16	35-20	6.4
0.16-0.1	10-0	8.7
0.1-0.074	–	9.8
0.074-0.04	–	15.3
< 0.04	–	59.8
Total	**100**	**100**

Table XII-3. Backfill mixture compositions that provides adequate compressive strength.

No. of composition	Amount [kg] per m^3					Compressive strength limit [MPa]
	Cement	Slag	Tailings	Crushed rock	Water	
4	200		1450	–	400	3.1
6б	350		1380	–	400	4.9
6в	400		1360	–	400	7.0
16	200		–	1550	400	3.0
17	250		–	1500	400	4.6
18	300		–	1450	400	6.2
25	350		950	400	400	4.4
26	400		950	350	400	5.2
38	400		950	350	380	4.5
44	–	600	950	250	400	3.0

Different grain size distributions require the evaluating the geomechanical performance of the massif after its having solidified. The procedure for the determination of compressive strength used for controls at Ukrainian mines is based on testing of backfill material standard samples after curing them for 7 days, 29 days, 3 months and 6 months respectively. The composition of the backfill mixture was selected by experiment, considering different ratios of binder and mill tailings. In total 49 samples were prepared and tested. The test results allowed to select a backfill mixture composition that is close to the standard values of compressive strength. A reasonable performance is achieved when Portland cement is used as a binding agent at a rate of about 200 to 400 kg per m^3 of the mixture.

The backfill composition is selected on the basis of experiments with different ratios of binding agents and tailings as a filling agent.

XII-6. Radiation impact on staff

Radiation conditions in the mine workings have been predicted. Mine air contamination at 40 workplaces was considered together with the air removed from the mine by the ventilation system by modelling the distribution of contaminated air along the mine workings. The model was developed on the basis of the assumption that collector ventilation system would be applied to ventilate the mine workings, where the contaminated air is collected into a special vent header arranged near the main level. The parameters of the model are given in Table XII.4.

Table XII-4. Main parameters of the Rn exposure model.

Parameters	Value	Unit
1. Number of mine workings (equivalent to calculational branches) in model	410	no.
2. Number of units in model (branch joints)	195	no.
3. Radon flux into mine sections:		
— being worked	59.52	$kBq \cdot s^{-1}$
— backfilled with solidified material	67.3	$kBq \cdot s^{-1}$
4. Radon flux from water exuded from solidified backfill materials in the mine	0.233	$kBq \cdot s^{-1}$
5. Pressure in mine ventilation network	4507	Pa
6. Air consumption of the mine	476	$m^3 \cdot s^{-1}$
7. Number of work places considered:		
— mine roadheading	2	no.
— bore drilling in blocks in operation	12	no.
— ore loading in car- truck	12	no.
— ore extraction from mine block	12	no.
— mine void abandoning and backfilling	2	no.

Table XII-5. Modelled mine air contamination.

Mining activities	Weighted-average concentration in air at working places [Bq/m^3] for each use of backfill material			
	sand-and-slag mixture (SSM)		cement-and-tailings mixture (CTM)	
	Radon	Rn progeny	Radon	Rn progeny
1. Mine roadheading	408	48.84	408	50
2. Drilling in operational blocks	2119	559.4	2132	567
3. Ore loading in car truck	2104	550.6	2104	550.6
4. Ore extraction from mine block	1624	385.17	1629	285.17
5. Mine void abandoning and backfilling				
— mounting and maintenance of backfilling pipe-line	3607	1670	4475	2006
— backfill materials supply	2069	520	2069	520

Mine air contamination parameters (see Table XII.5) were used to calculate the dose to staff at the main workplaces in the mine, both when conventional backfill technology is applied and when mill tailings are utilised for backfilling.

The inhalation component of effective dose for the staff depending on the workplaces location (Table XII.6) 1.03-1.2 times increases when tailings used for mined-out space backfilling.

Table XII-6. Calculated mining personnel exposures.

Mining activities	**Personnel exposure [mSv/year]**		**Ratio SSM/CTM**
	sand-and-slag (SSM)	**cement-and-tailings (CTM)**	
1. Mine roadheading	0.64	0.66	1.03
2. Drilling in operational blocks	7.4	7.5	1.01
3. Ore loading in car truck	7.28	7.2	1.0
4. Ore extraction from mine block	5.1	5.1	1.0
5. Mine void abandoning and backfilling			
— mounting and maintenance of backfilling pipe-line	9.64	11.55	1.2
— backfill material supply	6.88	6.88	1.0

XII-7. Contamination of groundwaters with radionuclides from the backfill massif

After the deposits are exhausted, the mine is closed, and the groundwater table has recovered to its pre-mining level, the backfill massif is expected to become a source of uranium series radionuclides. Release to the groundwater is followed by migration along the water flow paths to the areas of natural discharge.

Predictions of contaminant transport were made using the "Ground Water Flow Simulation (GWFS)" software produced by Geosoft-Eastlink Company (Russia/Sweden) and the FLOW utility designed to model migration processes in a vertical cross-section. The geometry of the hydrogeological model domain is determined by the distribution of the backfill material in the geological section and also by the groundwater flow patterns. A (hydro)geological cross-section is shown in Figure XII.3.

The list of groundwater contaminants to be investigated were determined according to the radiochemistry of the backfill material. This list includes ^{238}U and ^{226}Ra, for which the concentrations in the backfill material are 1,722 $Bq \cdot kg^{-1}$ and 5,922 $Bq \cdot kg^{-1}$ respectively in accordance with previous research and estimation results.

The radionuclides will migrate in an aquifer that largely consists of fractured granites. U and Ra migration distances depend on the porewater chemistry of the granites and their sorption properties. The source term depends on the chemistry of the backfill material and the water flux through the backfill. The latter in turn depends inter alia on the permeability of the backfill material.

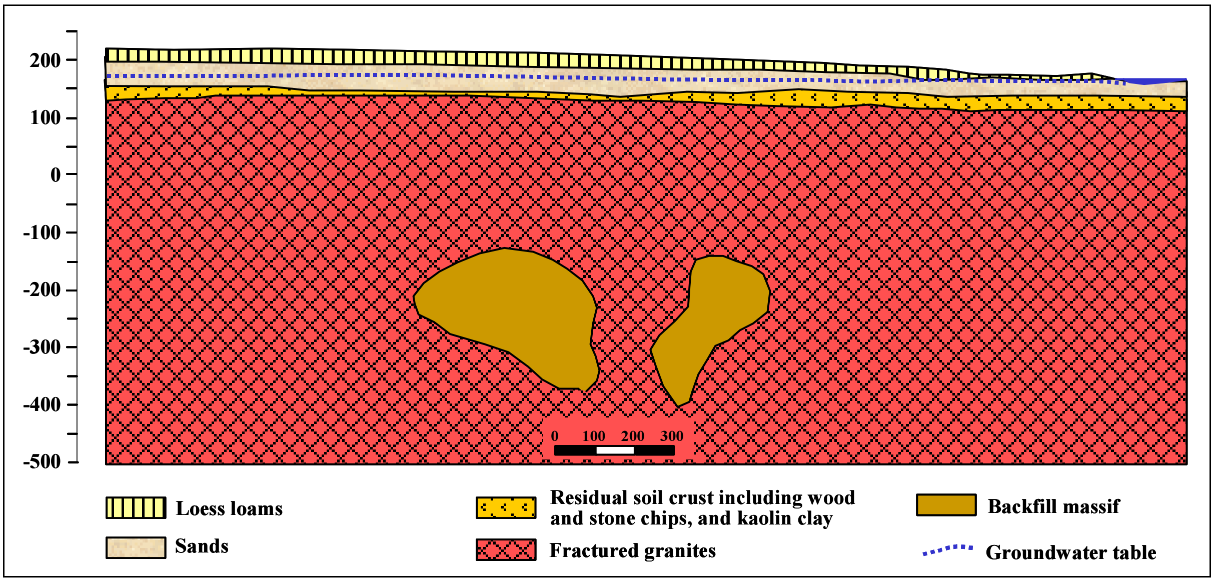


Fig. XII-3. Geological cross-section of the mine site.

Two cases for the state of the backfill material were considered for the prediction of the long-term contaminant behaviour:

- backfill material has the same permeability as the enclosing rock, i.e. permeability coefficient is 0.06 $m \cdot d^{-1}$;
- the backfill material has degraded and its permeability coefficient amounts now to 1.4 $m \cdot d^{-1}$.

The permissible and average concentrations of the radionuclides in the vicinity of ore deposit are listed in Table XII-7.

Table XII-7. Groundwater contamination criteria.

Radionuclide	**Permissible concentrations according to NRBU-97**		**Average concentrations around the deposit**	
	$Bq \cdot l^{-1}$	**$mg \cdot l^{-1}$**	**$Bq \cdot l^{-1}$**	**$mg \cdot l^{-1}$**
^{238}U	10	0.4	0.55	0.02
^{226}Ra	1		0.05	

The prediction was made for time points corresponding to a 100 and 1000 year period after the mine has stopped to be worked, the mine voids have been backfilled, and the hydrogeological regime of the groundwater has been reconstituted.

After 100 years, the boundary of permissible uranium concentration (see Table XII-7) is predicted to occur at distance of 129 m from the backfill massif for a maximum permeability coefficient 1.04 $m \cdot d^{-1}$, while the average concentration (0.02 mg/l) will be observed at a distance of 755 m (Fig. XII-4).

In 1000 years the boundary of average uranium concentrations, will move northwards and be out of the model domain (Fig. XII-5).

After 100 years the boundary of average radium concentration will be at 663 m distance from the backfill massif for a permeability coefficient of 1.04 m/day (Fig. XII-6).

After 1000 years the boundary will have moved back due to radioactive decay and would be observed at 465 m distance (Fig. XII-7).

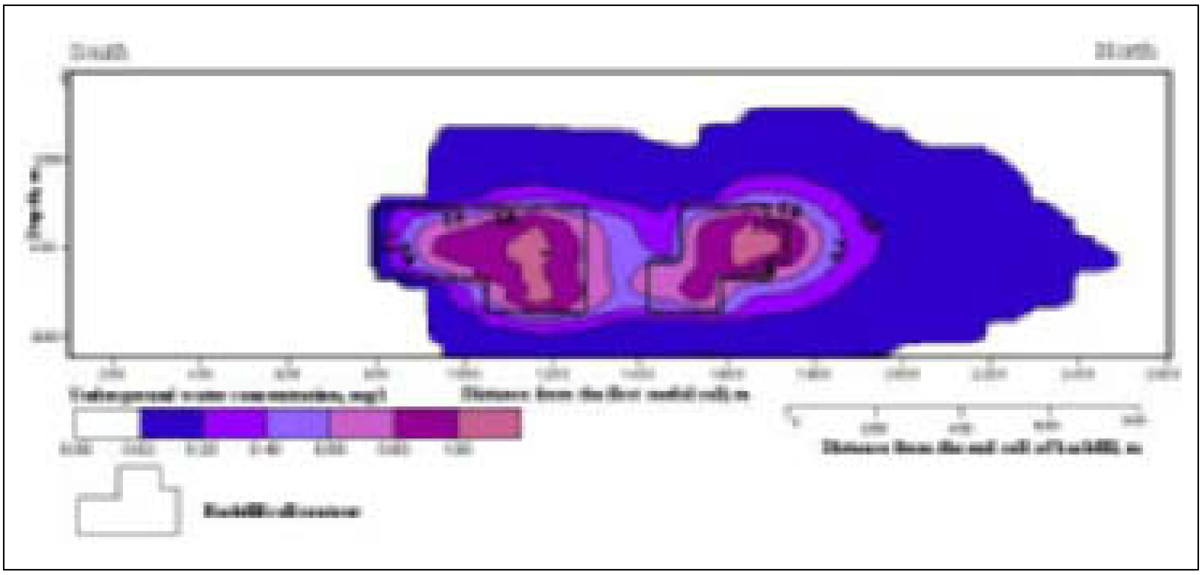


Fig. XII-4. Model outputs for uranium transport by groundwater in the aquifer in prediction for 100-year time period and permeability coefficient 1.04 m·d^{-1}.

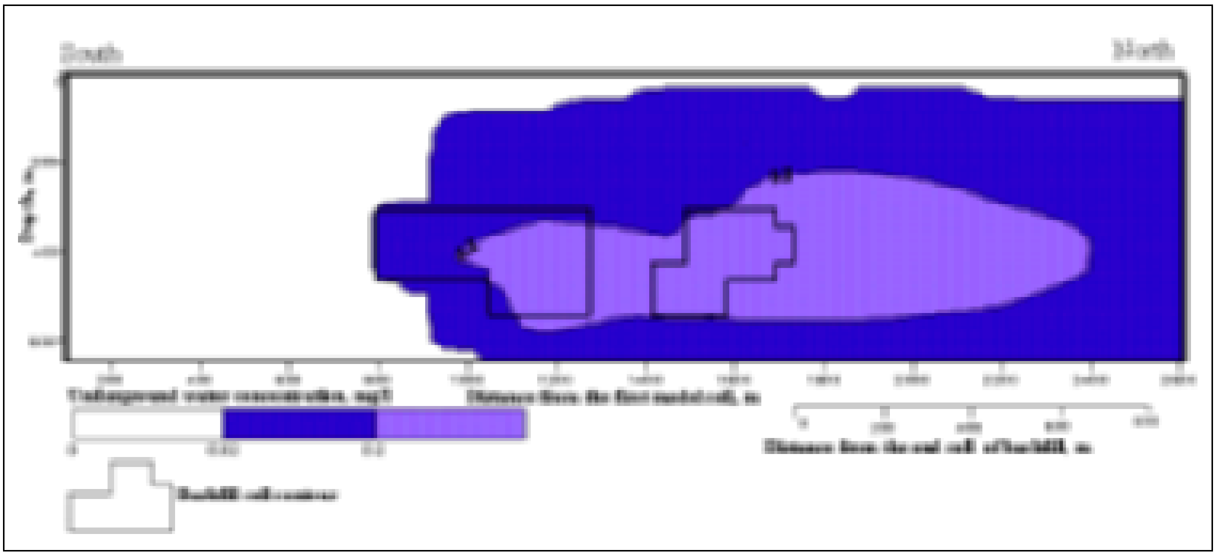


Fig. XII-5. Model outputs for uranium transport by groundwater in the aquifer in prediction for 1000-year time period and permeability coefficient 1.04 m·d^{-1}.

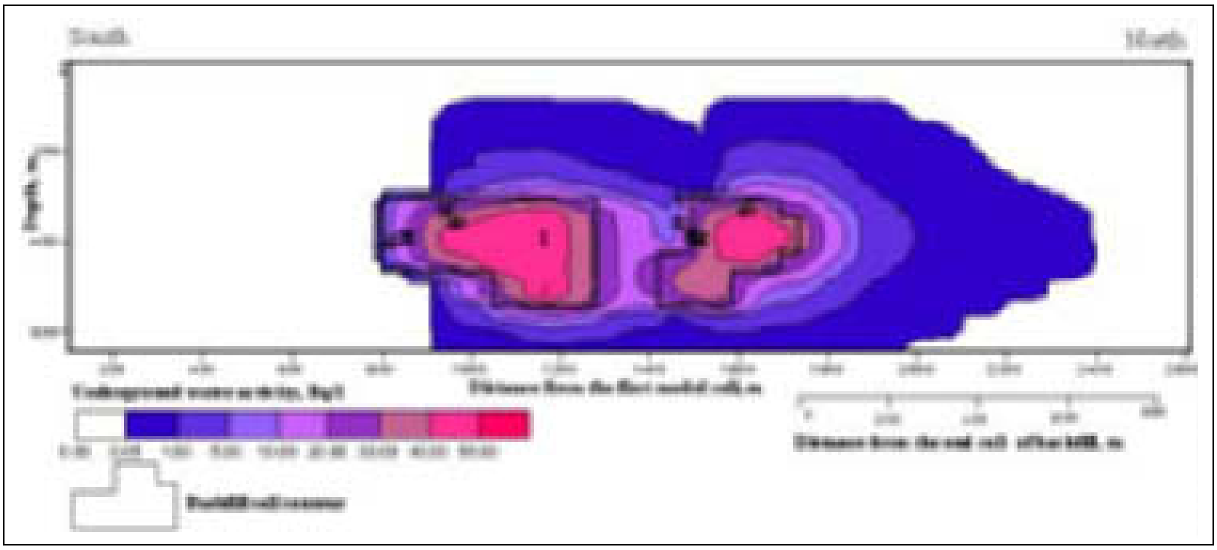


Fig. XII-6. Model outputs for radium transport by groundwater in the aquifer in prediction for 100-year time period and permeability coefficient 1.04 m·d⁻¹.

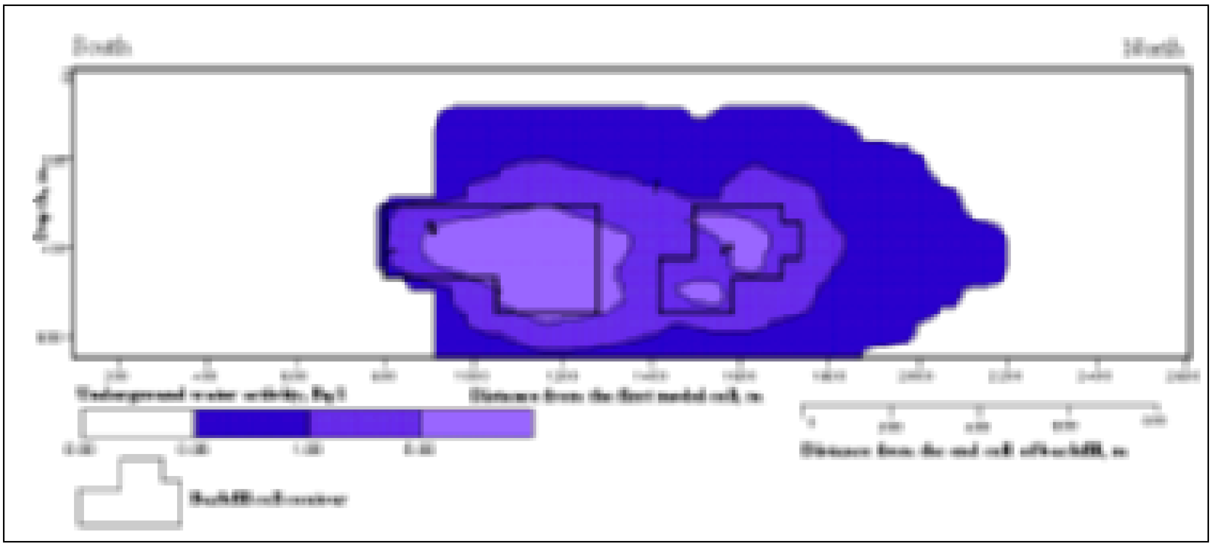


Fig. XII-7. Model outputs for radium transport by groundwater in the aquifer in prediction for 1000-year time period and permeability coefficient 1.04 m·d⁻¹.

XII-8. Air and soil contamination

Emissions are released into the ambient air from the mine ventilation exhausts, by which contaminated mine air is brought to the surface from underground workings and from the ventilation systems of the backfill mixing plants. Radon is emitted from the backfill massif, while the backfill mixing plants emit radon and dust.

The mine air is contaminated as a result of radon emanation from the backfill massif prepared with use of uranium mill tailings. Generation of dust is due to the tailings being handled, transported, and mixed. Atmospheric emission point geometric parameters and measured emission values are presented in Table XII-8.

Table XII-8. Characteristics of atmospheric emission points.

Emission point	**Emission point parameters**						
	Operating time [h·y^{-1}]	**Emission speed** [m·s^{-1}]	**Geometric parameters** [m]	**Radon emission** [kBq·s^{-1}]	[kBq·y^{-1}]	**Dust emission** [g·s^{-1}]	[t·y^{-1}]
Mine ventilation exhaust	6094	10.7	Ø= 5.6 H=20	1892.9	$4.15 \cdot 10^{10}$	–	
Backfill mixing plant ventilation systems:							
- dewatering plant	8160	10.0	Ø= 0.8 H=24	0.23	$6.76 \cdot 10^{6}$	0.031	0.687
- storage area, mixing area	6094	6.0 16.0	Ø= 1.0 H=20	338.76	$5.267 \cdot 10^{9}$	0.06	1.764

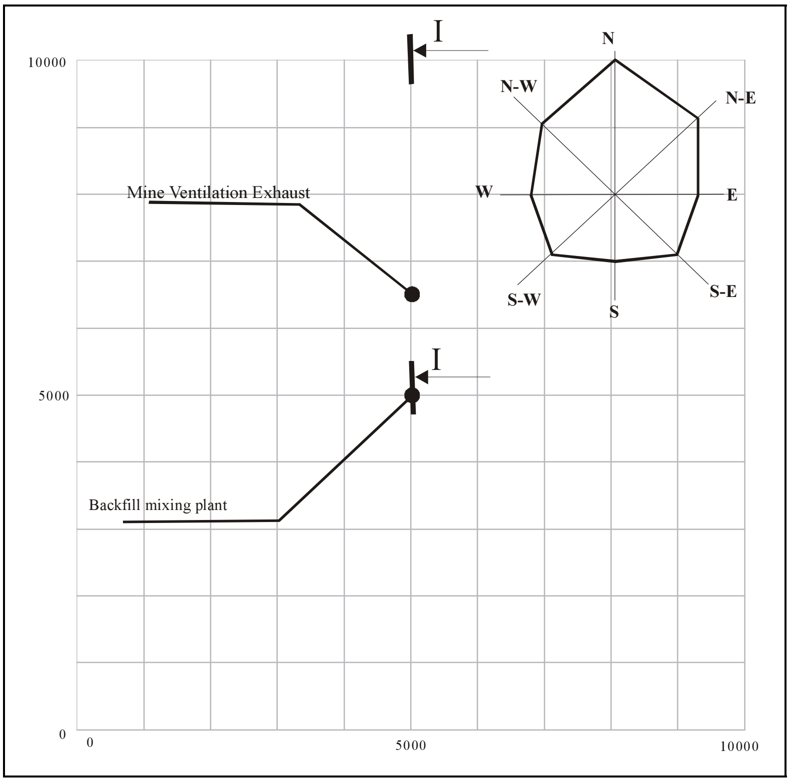


Fig. XII-8. Model grid, emission point locations, and average wind direction distribution.

The impact of atmospheric emissions is evaluated on the basis of surface air concentrations. Modelling has been carried out for an area of 10,000 m ×10,000 m, with a grid space of 250 m. The emission point locations are presented in Fig. XII-8.

Radon and dust volume activities in the surface air (on section I-I) resulting from calculations for emissions from the mine ventilation exhaust and the backfill mixing plant under annual average wind speed conditions (cf. Fig. XII-8) and the 'unfavourable' weather conditions respectively are given in Fig. XII-9.

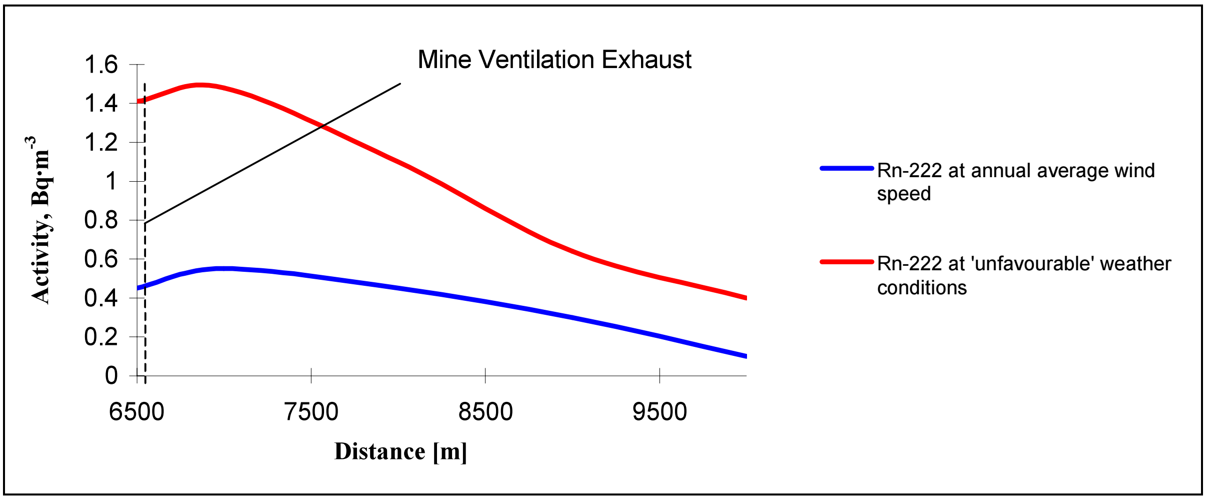


Fig. XII-9. Volume activity variation for radon emitted from the backfill massif through the mine ventilation exhaust.

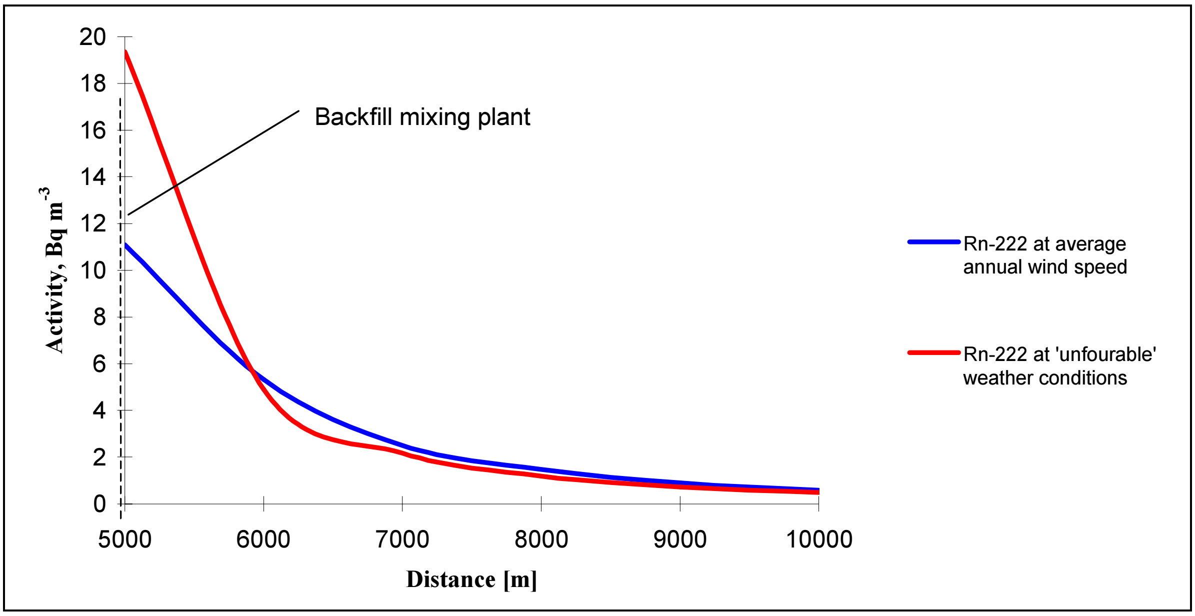


Fig. XII-10. Volume activity variation for radon emanating from the backfill mixing plant.

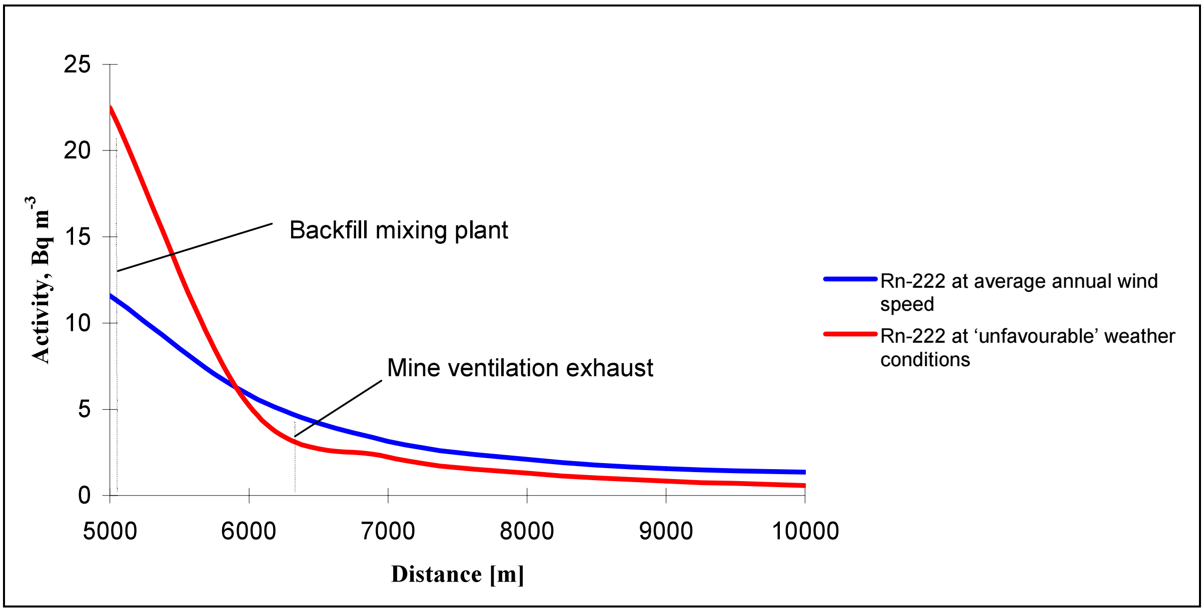


Fig. XII-11. Volume activity variation for radon in the surrounding air due to the combined effect of mine ventilation and backfill mixing plant exhaust.

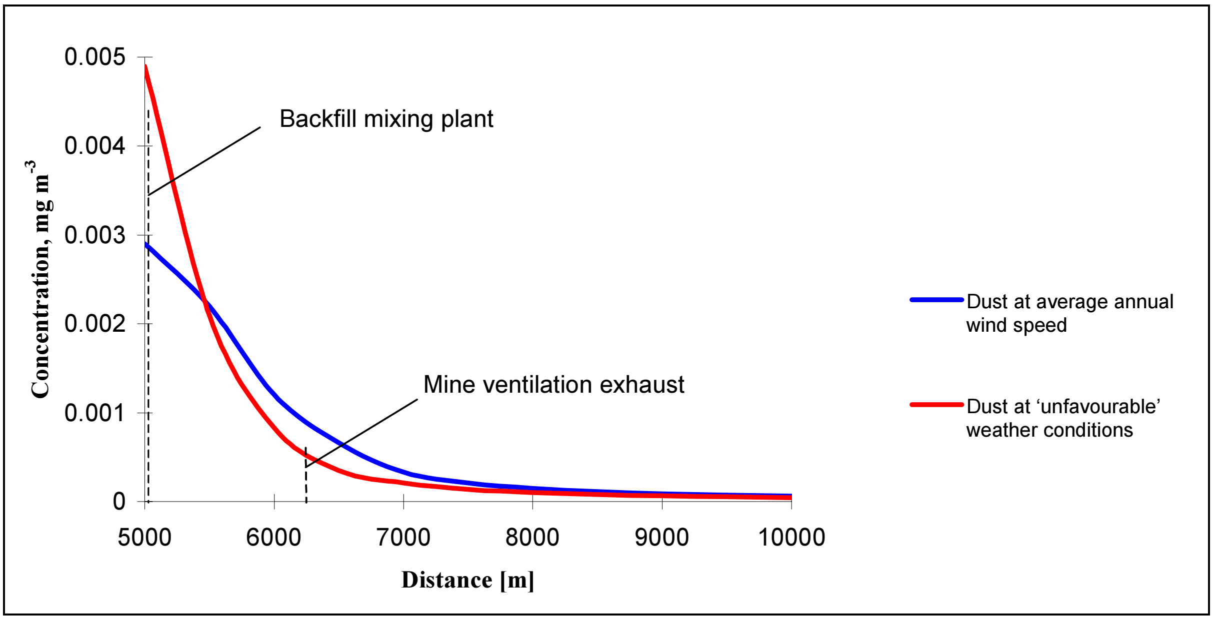


Fig. XII-12. Dust dispersion from the backfill mixing plant.

Maximum radon activities over the site are calculated as 11.6 Bq·m^{-3} for the average annual wind speed and as 22.5 Bq·m^{-3} for the 'unfavourable' weather conditions.

Average measured concentrations of the basic radionuclides in the dust are as follows: 2,516 Bq·kg^{-1} for uranium; 2,789 Bq·kg^{-1} for ^{226}Ra; and 9,768 Bq·kg^{-1} for ^{230}Th.

The distribution of volume activities for radionuclides in the dust emitted to the surface air from the backfill mixing plant for the average annual wind speed and the 'unfavourable' weather conditions respectively are shown in Fig. XII-13. For the ‘unfavourable’ weather conditions, the activity at the site increases by 4.6-18%.

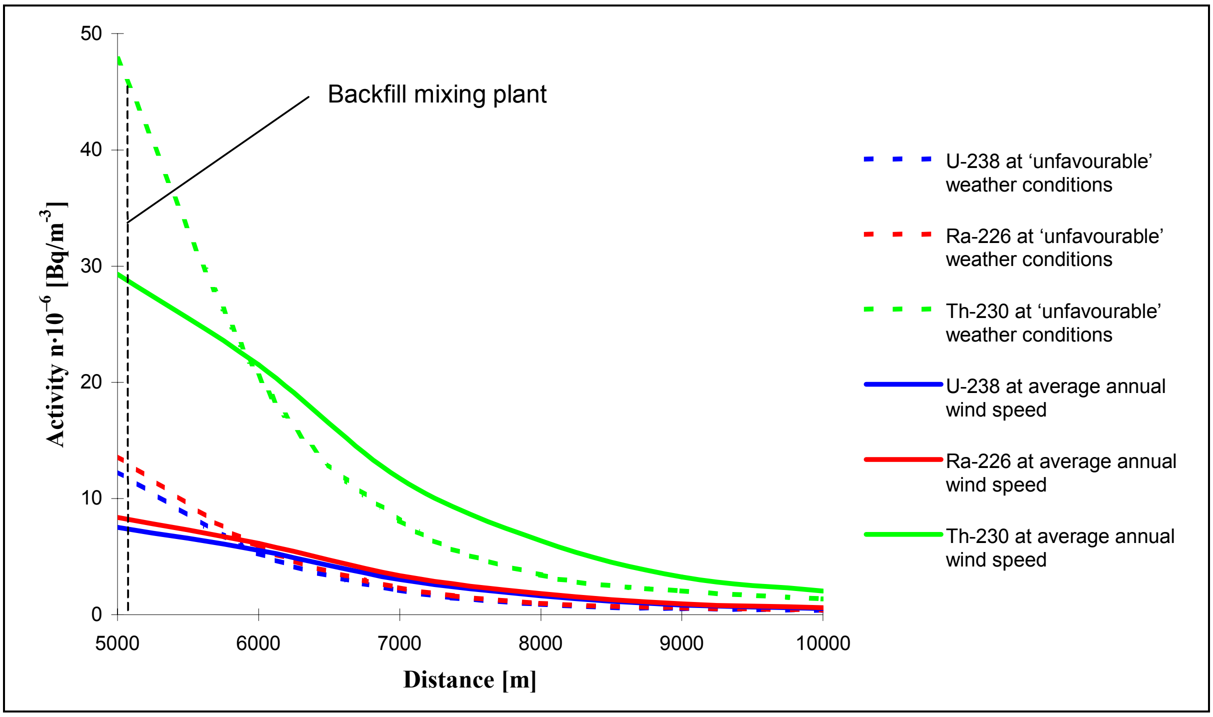


Fig. XII-13. Volume activity distributions for radionuclides in dust from the backfill mixing plant.

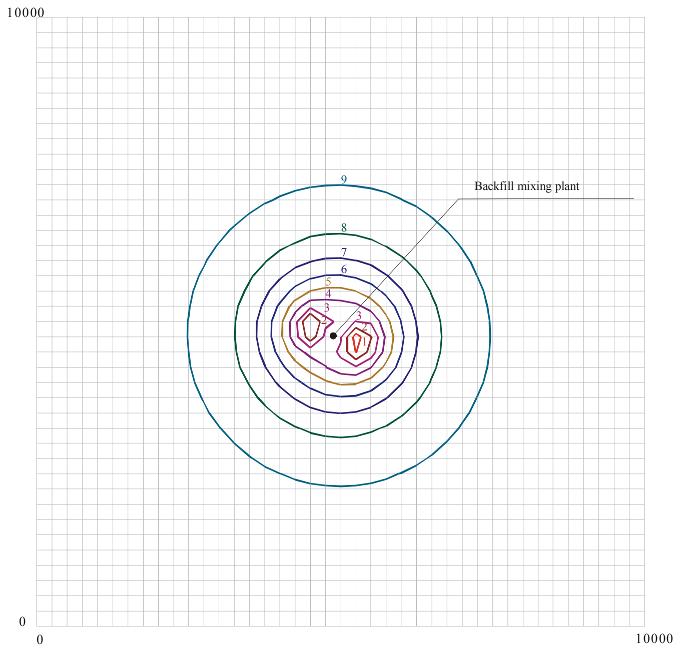
The radioactive contamination soils in the vicinity of the uranium mine is de-termined by wind dispersion and the settling of dust generated during the backfill mixture preparation involving uranium mill tailings.

The total quantity of dust being emitted to the surrounding air and settling onto the soils amounts to 0.002 $mg \cdot m^{-3}$ for an assumed operational period of 75 years. The total input of basic radionuclides into the soil per year and over the assumed operational period is given in Table XII-9.

Table XII-9. Calculated dust-borne input of radionuclides into the soils.

Radionuclide	**Annual input [$kBq \cdot y^{-1}$]**	**Input over 75 years [kBq]**
^{238}U	6167	$4.625 \cdot 10^5$
^{226}Ra	6835	$5.126 \cdot 10^5$
^{230}Th	23941	$1.796 \cdot 10^6$

The specific activity for radionuclides deposited onto the soils is calculated based on the radionuclides concentration in the ambient air. For the purpose of the calculations, the thickness of soil layer over which the radionuclides are dispersed is assumed to be 5 cm. The soil density is assumed to be1.4-1.5 $g \cdot cm^{-3}$. The calculated distribution of total radionuclide activities in the soil for the annual input of dust is presented in Fig. XII-14.



Iso-line	Specific activity [Bq/kg]		
	^{238}U	^{226}Ra	^{230}Th
1	0.0043	0.0047	0.017
2	0.0032	0.004	0.014
3	0.0028	0.003	0.012
4	0.0023	0.0025	0.0089
5	0.0018	0.002	0.0068
6	0.0013	0.0014	0.0051
7	0.00077	0.00084	0.003
8	0.00033	0.0004	0.0013
9	0.00011	0.00012	0.00043

Fig. XI-14. Calculated radionuclide specific activity distribution in the soil as a result of annual sedimentation of dust from the uranium mill tailings.

Table XII-10. Background activity concentrations of radionuclides in the soil.

Sample No.	**Activity concentration [Bq·kg^{-1}]**			
	^{238}U	**^{226}Ra**	**^{232}Th**	**^{40}K**
1	50	40	80	200
2	130	60	70	400
3	60	40	80	300
4	50	40	60	400
Average	72.5	45	72.5	325

Table XII-11. Calculated dust-borne contamination of soils at the border of the industrial zone.

Radionuclide	**Dust-borne activity concentration [Bq·kg^{-1}]**	**Natural background [Bq·kg^{-1}]**	**Dust-borne contribution [%]**
^{238}U	0.165	72.5	0.22
^{226}Ra	0.183	45	0.4
^{230}Th	0.64	-	-

The calculated mining derived activity concentrations are compared with the background activity concentrations of natural radionuclides in the soil. Background concentrations have been found by an experimental approach using four samples taken at places unaffected by mines (Table XII-10).

Maximum specific activities of the radionuclides in the soils are observed within the industrial zone of the object. Table XII-11 shows the expected radionuclide activity concentrations at the border of the industrial zone resulting from an input into the soil over an operational period of 75 years.

XII-9. Conclusions

The use of mill tailings as a component in the backfill mix for a uranium mine results in changes in the radiation impact on the staff and the population living in the surrounding area that is affected by dust and gas emission from the mine.

The additional contribution to the ambient air contamination in the vicinity of the uranium ore mine is estimated to be 10-18%. However. the radiation impact does not exceed the limiting values established by the state regulations.

The contamination of groundwaters due to the leaching and migration of radionuclides from the backfill massif is estimated to be limited to the natural halo of radionuclides in the host rock around the uranium deposit.

The soil contamination from the preparation of backfilling mixtures using uranium mill tailings does not significantly increase in comparison with the impact from conventional backfilling technology.

REFERENCES TO ANNEX XII

[XII-1] DBN A.2.2.1.-95, "Structure and content of matters for assessment of environmental impact..."
[XII-2] NRBU-97, "Radiation Safety Standard of Ukraine".
[XII-3] REGULATORY MATERIAL, "Temporary manual for determination of outcrop spans and standard strength of solidifying backfill materials for uranium mine of Ukraine, RM 95.8-684-82 (1982) (in Russian).
[XII-4] SPORO-85, "Sanitary rules of radioactive waste management", (1996) (in Russian).
[XII-5] OST 95 592-86, "Radioactive waste. General requirements to radionuclide content and activity measuring methods", (1987).

ANNEX XIII. USA

RESEARCH AND DEVELOPMENT OF MEASURES TO BE TAKEN FOR LONG TERM STABILIZATION AND ISOLATION OF URANIUM MILL TAILINGS

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US DOE, Grand Junction Office

XIII-1.Abstract

The U.S. Department of Energy has taken a holistic approach to the remediation of uranium mill tailings and contaminated groundwater at the Title I sites designated in the Uranium Mill Tailings Radiation Control Act. This approach is suggested as guidance for the characterization and remediation of other sites with contamination, depending on site-specific or country-specific conditions.

This paper describes the process using certain specific examples.

XIII-2.Introduction

XIII-2.1. Initial scoping

The first step in the environmental restoration of a uranium mill tailings site (Fig. XIII-1) is to collect all available existing information to become familiar with the site. The primary goals of this initial scoping are to determine whether any risks and hazards are present that require immediate attention and to define the project scope (problem definition). The results of this scoping will be a summary report describing the physical features of the site, a summary of historical information, and an initial assessment of risks and immediate corrective measures.

Fig. XIII-1. During the early years of processing uranium ore in the United States, liquid waste was piped to ponds where the liquid component evaporated, leaving fine sandlike tailings.

XIII-2.2. Define problem and potential risks

Problem definition includes determining the physical parameters of the site, examining historical records, and identifying potential risks. Physical parameters include the major physiographic features (man-made and natural), climate, site ownership and access, and current and former land use. Historical photographs, maps, and reports are useful in identifying physiologic features, especially maps for utilities (e.g., electricity, gas, and water) and as-built drawings. Interviews with current and former employees are helpful in identifying former practices that may have contributed to environmental releases. Site records should include procedures, environmental reporting requirements, process information, chemical usage, waste disposal, permits, and maintenance activities. A literature search may provide additional information, such as public documents, process information, and technical studies.

An inspection of the site should include a walk-through of all on-site facilities. Specific items to look for associated with the milling process include raffinate ponds, tailings ponds, processing facilities, evidence of underground tanks and piping, utilities, landfills, and waste pits. In addition, potential exposure pathways should be investigated, such as windblown contamination, surface water bodies, wells, and wetlands (Fig. XIII-2).

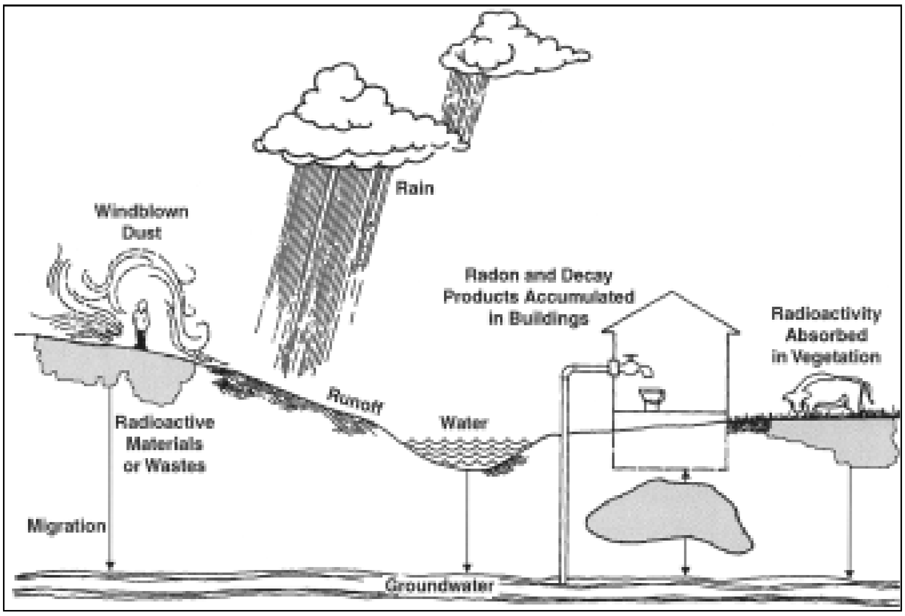

Fig. XIII-2. Environmentally dispersed radionuclides and wastes from uranium mill tailings use various pathways to affect living organisms, soil, surface water, and groundwater.

Information gathered during the review of historical information and the site inspection should be compiled in a list of contaminants of potential concern with potential pathways and receptors. Contaminants of potential concern may be radioactive constituents of the tailings and leftover ore; chemicals used in the processing, such as acids or caustics; and substances used in the operations, such as fuels and solvents. This initial phase of the risk assessment determines whether immediate corrective actions should be implemented.

All the information gathered in the initial scoping should be documented in a summary report that includes the collected data, an analysis of the data, immediate corrective actions as warranted, and a summary of data gaps needed to complete the characterization of the site. The data analysis should include a site conceptual model and the results of the initial risk assessment. An evaluation of the need for an emergency or interim action should be made based on the initial evaluation of types of contamination and exposure. An emergency action may be to remove people from an area or to provide an alternate drinking water source.

XIII-2.3. Initiate emergency and interim actions

If an emergency or interim action was deemed necessary in the initial scoping phase, the action should be completed as soon as possible. A minimum amount of review and public input should be done so the action can be initiated as soon as possible.

XIII-3.Planning

A clear definition of short- and long-term objectives and goals is necessary for efficiency and focus of the project. Objectives may include meeting regulatory standards or reducing risk to acceptable levels. Goals may be established to complete specific phases of the work within a certain time period or budget.

Stakeholders include owners, regulatory agencies, nearby residents, state and local governments, and other interested parties. Stakeholders are identified early in the project to establish communication (Fig. XIII-3).

Fig. XIII-3. Communication with stakeholders is an integral component of remedial action.

Applicable regulations are identified to determine cleanup standards and goals. The cleanup standards and goals will guide the level of characterization to be performed and will narrow the list of potential remedies. Focusing on potential remedies early in the process helps to move the project toward the end goal of site cleanup. It also helps limit data collection to that which is necessary to implement the potential remedies.

Management plans are prepared to ensure consistency within the various project tasks, to gain consensus from the various stakeholders, to document the progress of the project. The Project Plan describes the goals and objectives, scope, schedule, budget, project organization, roles and responsibilities, procedures, and milestones. The Public Involvement Plan describes actions to encourage stakeholder involvement and understanding to support the decision-making process. The Quality Assurance Plan describes the quality requirements including data collection, laboratory analysis, and technical calculations. The Health and Safety Plan describes the health and safety concerns of the project, roles and responsibilities, emergency procedures, job safety analysis, and training requirements. The Analytical Laboratory Procedures Plan describes the analytical procedures to be used, including quality assurance requirements. A Procurement Plan describes the procurement processes and major subcontract requirements. A Records Management Plan describes the control, retention, index, filing, and storage requirements for project records (Fig. XIII-4). A Database Management Plan describes how the data will be managed, including how it will be stored, what types of data will be stored, who will have read/write access, data formats, and electronic transfers.

Fig. XIII-4. Records Management Plans provide the requirements for project records, both historical records and records generated during the cleanup process.

Technical plans are prepared to document the evaluation of existing data, to define what additional tasks need to be accomplished, and to provide a means of gaining consensus among stakeholders on future work. Technical plans generally fall into two categories: work plans and procedures.

Work Plans are prepared for performing fieldwork, such as installation of monitor wells, pilot tests, and performance monitoring. Work Plans will typically include an evaluation of existing data, a discussion of data deficiencies, data quality objectives, and procedures. Data collection procedures are included to ensure that methods are understood and approved by stakeholders. Sampling and Analysis Plans contain procedures for all types of media sampling and analysis and for location and frequency of sampling. Analytical methods include field methods as well as laboratory methods.

XIII-4.Site characterization

XIII-4.1. Field investigations

Field investigations include performing land surveys and develop base maps, including geographical, topographical, and man-made features. Accurate coordinates and elevations need to be established for existing wells, surface water features, geologic contacts, historical source areas, etc.

Collection of field data consists of conducting field sampling (Fig. XIII-5a) in accordance with Work Plans. Schedule activities to maximize resources, to enhance data quality, and to minimize costs. A phased approach should be used to sequence the activities to achieve a

(a) (b)

Fig. XIII-5. (a) Collection of soil samples is necessary to determine the extent of contamination and define the types of contaminants; (b)

Fig. XIII-6. A field laboratory offers quick turnaround times for some types of sample analyses and allows investigators to determine extent of sample collection.

more logical sampling approach; for example, start with nonintrusive methods to obtain a more complete and comprehensive understanding of the problem before more direct characterization methods are employed. Integrate existing information with new data to revise the site conceptual model and to refine data collection needs concurrent with or before proceeding to the next activity. Lead time for procurement activities, resource availability, remoteness of site, weather conditions, and other issues should be considered. Work readiness reviews can be used to minimize the possibility of delays and problems because of incomplete planning and preparation.

Coordinate field sampling with the analytical laboratory(s) with respect to holding times, analyte list, sample matrix, preservatives, and shipping and transportation regulations for hazardous material. Consider the lead time for laboratory audits and qualifications, procurement, etc. Focus the investigation as much as possible on using field laboratories and to reduce the volume of samples submitted to fixed-base laboratories(Fig. XIII-6).

XIII-4.2. Geographical information system database development

Location, sample, and base map information for geographical information system (GIS) database development involves establishing a database structure with location information such as wells, boreholes, surface (Fig. XIII-5b). Sample information may include surface water, groundwater, soil, sediment, tailings, fauna, biota, analytical results, inorganic, organic, radiologic, etc. Base map features include roads, fences, buildings, ponds, lakes, streams, rivers, boundaries, geological, ecological, etc.

XIII-4.3. Data evaluation and reports

XIII-3.3.1 Determine nature and extent of contamination

Prepare maps, diagrams, and figures to assist in the evaluation of the geological, hydrological, geochemical, and ecological data. Geologic evaluation considers regional and local geology, bedrock structure and topography, and unconsolidated units. Hydrologic characterization evaluates surface hydrology, groundwater hydrology, and aquifer interaction with surface features. Physical features, coordinate survey information, geologic, and hydrologic data are input into a groundwater flow model to develop the hydrostratigraphic model. Geochemical characterization delineates the area where contamination exists, documents the individual chemical constituents that contribute to the contamination, and evaluates the fate and transport of the site-related contaminants. This evaluation includes an analysis of natural background, source areas and contaminants, water quality, soil, and sediments. The geochemical processes need to be identified for each contaminant that cause contaminant retardation, such as adsorption, absorption, precipitation, diffusion into immobile porosity, transfer to vapor phases, etc.

The geochemical characterization can be combined with the groundwater flow model to predict the concentrations and distributions of contaminants. Ecological evaluations are performed to assess ecological risks associated with site-related contaminants and to assist in developing the site conceptual model. Vegetation at the site and surrounding areas should be mapped to support the water balance model. Vegetation can influence both recharge and discharge from groundwater sources, depending on the health of the plant communities, soil properties, and depth to water.

XIII-3.3.2 Finalize site conceptual model

Integration of the site characterization data provides the most current understanding of the extent and magnitude of contamination (Fig. XIII-7), exposure pathways, and risks to public health and the environment. This integrated model of the site can be used to support the proposed groundwater remediation objectives.

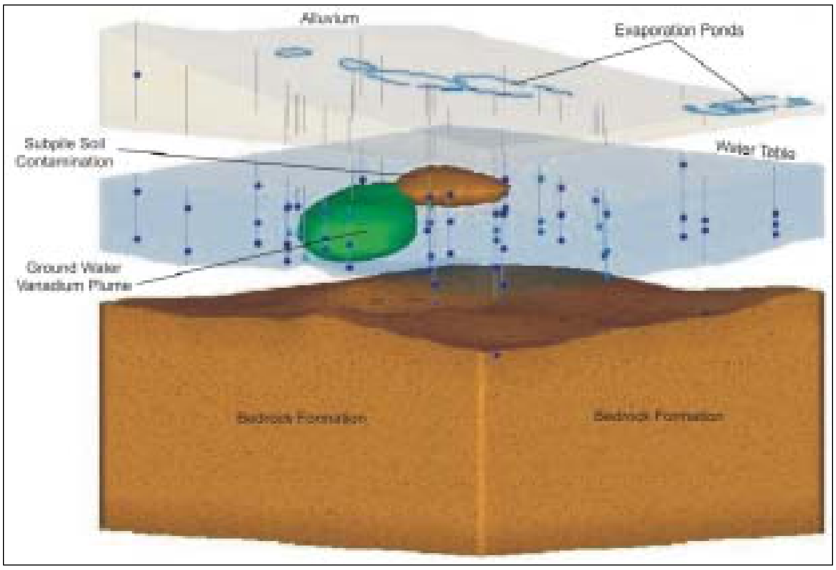

Fig. XIII-7. Selected geochemical and hydrological information is displayed in a three-dimensional model.

XIII-4.3.1. Prepare environmental and human health risk assessment

The environmental and human health risk assessment will determine ecological and human receptors for each potential exposure pathway, identify contaminants for each pathway, and compare contaminant concentrations in media to appropriate benchmarks and to toxicity data to estimate risk.

XIII-5.Remedial alternatives

Finalizing the objectives includes determining a final list of applicable rules and regulations and a detailed evaluation of the risks. The applicable rules and regulations and the risks will determine the cleanup levels, time required for cleanup, and a list of contaminants. The holistic approach develops surface remediation objectives that consider the effects of surface and vadose zone contamination on groundwater contamination.

Surface remediation objectives should address specific regulations that are in effect, the risks from exposure to radon and radiation from the tailings, and vadose zone contamination that may affect groundwater contamination. In the United States, regulations dictate acceptable levels for radon and radioactive emissions, allowable levels for radioactive contaminants left in the soil, and design standards for construction of disposal cells.

Groundwater objectives should address the allowable concentrations of contaminants in the groundwater. The allowable concentrations of contaminants should be based on the current and potential uses of the groundwater and the desired groundwater quality for those uses. The objectives should also address the cleanup time to restore the groundwater to the desired groundwater quality. Clean up time may be dictated by a regulation or be based on a policy decision.

XIII-5.1. Development and evaluation of surface remediation alternatives

Surface remediation alternatives should be developed that address the remediation objectives for both the surface and the groundwater. Groundwater cleanup standards may determine the amount of tailings and contaminated soil to remove. If regulations dictate that a disposal cell be built and the standards for design, alternatives may only consider whether to leave the tailings in place or to relocate the tailings and how much contaminated material to remove (Fig. XIII-8). Alternatives also may consider institutional controls, such as deed restrictions, as a way to limit exposure or a variation in design standards for a disposal cell. In the United States, a no action alternative (i.e., no remediation) is developed for comparison purposes.

Fig. XIII-8. Windblown tailings increase both the areal extent of contamination and the area that requires characterization.

Surface remediation alternatives should be evaluated for effectiveness, implementability, and cost. Effectiveness evaluates how well the alternative meets the objectives. Implementability evaluates whether it is technically feasible to construct and operate the alternative and whether there are administrative issues that may affect construction and operation. Cost may evaluate the overall cost of the alternative or just the initial construction cost. The overall cost includes the construction cost, operation and maintenance cost, and the long-term monitoring cost. If the alternative uses a new technology or technique, treatability or pilot tests may be needed to verify performance.

The selected alternative should provide the best balance of effectiveness, implementability, and cost, depending on the importance assigned to each evaluation criteria. For example, the alternative that provides the highest measure of effectiveness will probably have the highest initial cost.

A report should be prepared that documents the remediation alternative that was selected and the rationale for that selection. The report should be made available for all interested parties to review.

XIII-5.2. Development and evaluation of groundwater remediation alternatives

As with surface remediation alternatives, groundwater alternatives should be developed that address the remediation objectives. Alternatives should consider a range of actions that would meet the objectives during different time periods. Typically, groundwater remediation alternatives involve natural attenuation with monitoring, in situ remediation, extraction of contaminated groundwater and treatment, or a combination of approaches.

Evaluation of groundwater alternatives is similar to evaluation of surface alternatives. Alternatives are evaluated for effectiveness, implementability, and cost. The time to restore the groundwater to the desired quality may vary widely between alternatives. For example, it may take 100 years or more for natural attenuation to restore the groundwater while extraction and treatment may restore groundwater quality much sooner.

The selected alternative should provide the best balance of effectiveness, implementability, and cost. The maximum allowable time for groundwater quality to be restored to acceptable levels is often a key aspect of the decision. For example, if groundwater quality must be restored within 100 years, natural attenuation may not meet that objective. A report should be prepared that documents the alternative that was selected and the rationale for that selection. The report should be made available for all interested parties to review.

XIII-6.Remedial design/action

XIII-6.1. Surface remedial design/action

The design of a disposal cell and other surface remediation components should consider factors such as the design life of the disposal cell, the cover, the radon barrier, surface water runoff, erosion protection, geotechnical considerations, nonradiological contaminants, and transient drainage. Design of the cover to eliminate or minimize infiltration of water during the life of the disposal cell is a key component (Fig. XIII-9). Construction of the disposal cell or stabilization measures should consider worker safety, safety to the community during construction, control of spread of contamination by wind, and control of runoff through contaminated areas.

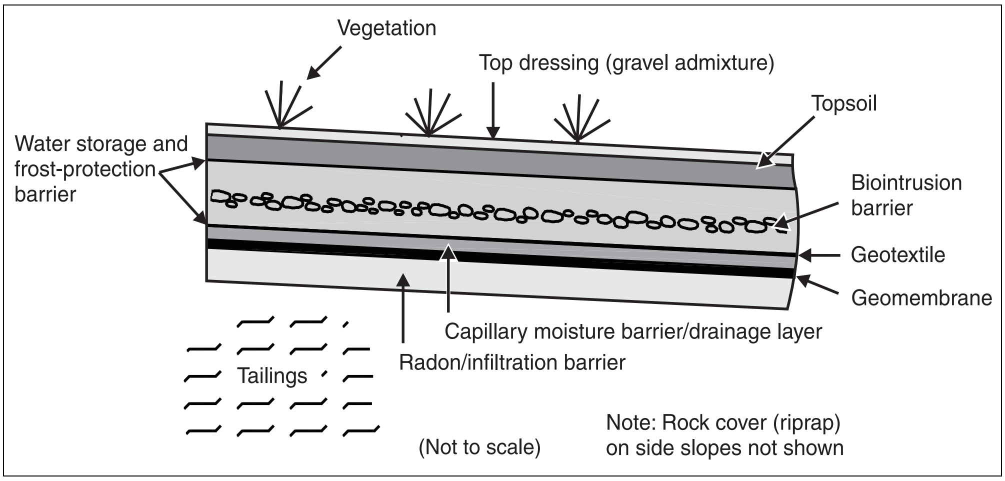

Fig. XIII-9. Cross section of one type of cover emplaced on a uranium mill tailings disposal cell in the United States.

A disposal cell designed to last for several hundred years to 1,000 years requires periodic monitoring and maintenance. Monitoring involves obtaining samples from groundwater wells to determine whether the cover is allowing water to infiltrate and site visits to inspect the erosion control measures and the integrity of the cell.

XIII-6.2. Groundwater remedial design/action

Depending on the selected remedy, the groundwater remediation design will vary considerably. For natural attenuation, design activities will involve establishing a monitoring plan and designing additional monitor wells. For an extraction and treatment remedy, activities will involve designing the extraction wells, determining placement of the wells, designing injection wells or infiltration trenches (if used), and designing the treatment system.

The remedial action phase may be limited to installing additional monitor wells required to monitor natural attenuation (Fig. XIII-10). If the selected remedy is extraction and treatment, remedial action will involve installation of the extraction and injection wells and construction of the treatment system.

Fig. XIII-10. A drill rig is positioned prior to drilling a monitor well at a remediation site.

All groundwater remediations require a significant amount of monitoring constituents and their concentrations in the groundwater. For natural attenuation and in situ remediations, post-construction activities are limited primarily to monitoring. For extraction and treatment remedies, operation of the extraction well field and the treatment system are the major activities.

XIII-7.Performance evaluation

Monitoring activities are designed to determine the progress of the remediation approach in meeting the goals and standards established for the site. Physical boundaries are monitored to ensure integrity of fences, changes in land use, etc. Sampling and analysis is performed to document increases and decreases in contaminant concentrations during remedial action, to track progress of natural attenuation sites, and to provide early warning of plume migration into uncontaminated areas. An institutional control program should be developed to prevent future use of potentially harmful contaminated groundwater. Compliance of any property restrictions needs to be monitored on use of drinking water, livestock watering, irrigation of edible vegetation, and uses of ponds for ornamental or landscaping purposes.

Fig. XIII-11. This mill, located near Grand Junction, Colorado, processed uranium ore from 1951 until 1970; three tailings ponds are visible in the foreground.

Fig. XIII-12. Plans for a portion of the remediated Grand Junction, Colorado, millsite, adjacent to the Colorado River, include development of a riverfront park and trail system. Only one building (in circle) remains after cleanup of the site.

Verification involves continued monitoring of contaminant concentrations for a defined period of time after completion of remediation to determine the effectiveness of the remediation. Results are statistically compared against established cleanup benchmarks for the site. Trend analysis should be applied to evaluate the monitoring data and to demonstrate model predications for natural attenuation sites. Flow-and-transport models are updated as new information is obtained and new model predictions are compared to cleanup objectives.

Closeout reports should be prepared to document that remediation actions have been completed and goals have been achieved (Figs. XIII-11 and XIII-12).

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Technical Meetings

1st Research Co-ordination Meeting, 25–29 September 2000, Vienna, Austria

2nd Research Co-ordination Meeting, 6–10 May 2002, Rio de Janeiro, Brazil

Consultants Meeting, 9–13 December 2002,Vienna, Austria

3rd Research Co-ordination Meeting, 29 September–3 October 2003,
Bystrice nad Pernstejnem, Czech Republic