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Extent of Environmental Contamination by Naturally Occurring Radioactive Material (NORM) and Technological Options for Mitigation

IAEA
International Atomic Energy Agency

EXTENT OF ENVIRONMENTAL CONTAMINATION BY NATURALLY OCCURRING RADIOACTIVE MATERIAL (NORM) AND TECHNOLOGICAL OPTIONS FOR MITIGATION

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The Agency's Statute was approved on 23 October 1956 by the Conference on the Statute of the IAEA held at United Nations Headquarters, New York; it entered into force on 29 July 1957. The Headquarters of the Agency are situated in Vienna. Its principal objective is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world".

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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, including the legacy of past practices and accidents.

In keeping with this, the IAEA has initiated a comprehensive programme of work covering all aspects of environmental remediation:

- Factors important for formulating strategies and selecting technologies for environmental remediation;
- Site characterization techniques and strategies;
- Assessment of remediation technologies;
- Assessment of technical options for cleanup of contaminated media;
- Post-restoration compliance monitoring;
- Remediation of low level dispersed radioactive contamination in the environment.

However, radioactive residues are found not only in fuel cycle activities, but also in a range of other industrial activities, namely:

- Mining and milling of metalliferous and non-metallic ores;
- Production of non-nuclear fuels, including coal, oil and gas;
- Extraction and purification of water, for example for the generation of geothermal energy, as drinking and industrial process water, and in paper and pulp manufacture;
- Production of industrial minerals, including phosphate, clay and building materials;
- Use of radionuclides, such as thorium, for properties other than their radioactivity.

Naturally occurring radioactive material (NORM) may be present in these activities and may lead to exposures at some stage of the processes and in the use or reuse of products, residues or wastes. The present publication attempts to give an international overview of the relevant processes, the types of residues and wastes generated, and the pertinent management strategies and technologies, with a view to identifying shortcomings that may lead to the need for remediation. An overview of applicable abatement and remediation technologies and strategies is given, together with a brief discussion of the possible socioeconomic impact.

The intention is to help Member States gain perspectives regarding the industrial processes that may lead to NORM generation and where, within those processes, NORM can accumulate and eventually may lead to increased potential for exposure of the general public to radioactivity. It is also hoped to make decision makers in radiation protection aware of the industrial and socioeconomic implications of their decisions. Furthermore, it is hoped that this report will help to identify those process steps that are amenable to improved radiation protection and the type of residue and waste and the associated disposal sites that may be of concern and in need of remediation.

The Scientific Secretary for this publication was W.E. Falck of the Waste Technology Section, Division of Nuclear Fuel Cycle and Waste Management.

The IAEA is grateful to all contributors and reviewers.

CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

Naturally occurring radionuclides are present in many natural resources. Elevated concentrations of these radionuclides are often found in certain geological materials, namely igneous rocks and ores. Human activities that exploit these resources may lead to enhanced concentrations of radionuclides (often referred to as technologically enhanced naturally occurring radioactive material (TE-NORM)) and (or) enhanced potential for exposure to naturally occurring radioactive materials in products, by-products, residues and wastes. Such activities may include, for instance, the mining and processing of ores, the combustion of fossil fuels, or the production of natural gas and oil. If these residues containing naturally occurring radionuclides are not managed properly and safely, contamination over large areas is possible given the large quantities of such residues.

Definition of NORM:
For brevity and clarity the acronym NORM is used in this report to encompass all naturally occurring radioactive materials where human activities have increased the potential for exposure in comparison with the unaltered situation. Concentrations of radionuclides may (i.e. TE-NORM) or may not have been increased.

These residues containing NORM as altered by human activities from outside the nuclear fuel cycle have developed over the past three decades from a little known issue to an issue that is receiving a considerable amount of global attention. Important reasons for the increasing levels of concern are the:

- Large amounts of such NORM containing wastes and other materials;
- Potential long term hazards resulting from the fact that NORM is comprised of long lived radionuclides with relatively high radiotoxicities;
- Higher likelihood for members of the public to be exposed to NORM contained in wastes and products than to most other sources of radiation.

A significant step that should promote the development of mechanisms to implement effective management of wastes and residues, including those containing NORM, is Agenda 21, the action plan prepared by the governments participating in the United Nations Conference on Environment and Development (UNCED) in 1992 in Rio de Janeiro [1]. Agenda 21 contains several sections related to waste management. Two general themes are the need to:

- Reduce the amount of all types of waste being generated;
- Manage all waste in a manner that protects human health and the environment.

Notwithstanding the ongoing discussion about whether and when NORM containing residues and wastes are or are becoming ‘radioactive waste’ [2], a number of the principles formulated for radioactive waste management would be directly applicable [3]:

- “Radioactive waste shall be managed in such a way as to provide an acceptable level of protection of the environment”;
- “Radioactive waste shall be managed in such a way that it will not impose undue burdens on future generations”;
- “Interdependencies among all steps in radioactive waste generation and management shall be appropriately taken into account”;
- “The safety of facilities for radioactive waste management shall be appropriately assured during their lifetime”.

Hence, an important first step for industry and regulatory bodies in a country is to understand when and where NORM can occur within a given process and also to identify the locations where concentrations of NORM can be greatest within a given process. This report is intended both to identify the industries which may involve NORM and to provide perspective on the amounts, characteristics and radionuclide concentrations which can be found in products, by-products and residues associated with a variety of different processes.

Determining at what concentration for a given exposure situation NORM becomes a potential radiological concern is a high priority in many countries. A number of countries have established concentration levels at which NORM is considered ‘radioactive’ and must be controlled; however, there is no international consensus on approaches to determine these concentration levels or on the specific concentration levels. This has proved to be problematic firstly due to the potential economic impacts associated with control of NORM containing residues in a manner consistent with management of other radioactive wastes and secondly due to the disagreement on appropriate exposure scenarios and models [4].

The task is further complicated by the fact that the relevance of exposure to NORM depends very much on local or regional circumstances and behavioural patterns. For example, an oil well or mine located in an unpopulated, remote area poses less of a risk for potential exposures to the general public than similar facilities located near a populated area. Likewise, rural populations in tropical developing countries will face a different potential for exposure than a rural family in Europe (different types of homes, amount of time spent at home, etc.). Thus, exposure scenarios to be considered when determining the concentration levels at which NORM related doses are considered significant will depend on living conditions and human behaviour in that region.

As a result of region specific considerations, the amenability of NORM to regulatory control and the mode for the implementation of such controls are subject to scientific and technical debate. Attempting to set up a system of clearance levels for NORM is further complicated by the fact that introducing new regulations or extending existing controls to residues containing TE-NORM is likely to have a significant technological and socioeconomic impact, given their wide occurrence and often large volumes.

1.2. OBJECTIVES

The main objectives of this report are:

(a) To provide the first step in an effort to develop a global knowledge base on the occurrence and characteristics of NORM in a wide variety of industrial and domestic activities;
(b) To identify and assess potential technologies that have been or could be used to help reduce the potential for or magnitude of potential exposures.

Such technologies could be applied:

(1) At the source to limit the amount of radioactivity extracted with the raw material;
(2) During the process in the form of recycling/reuse or treatment;
(3) At the back end of the process through special disposal practices;
(4) In the form of remediation approaches in cases where some form of remediation is needed.

This report is intended to help Member States gain perspective regarding the industrial processes that may lead to NORM generation and where, within these processes, NORM can accumulate and may eventually lead to an increased potential for exposure of the general public to radioactivity. To this end, this report includes a summary of a variety of processes that may involve NORM and some examples of the amounts and characteristics of NORM containing feedstocks, residues, by-products and products associated with these processes.

1.3. SCOPE

Industrial processes potentially involving NORM are addressed in this report. The emphasis is on processes involving NORM that, depending on country, may or may not be regulated in respect of their radioactive properties. Products, by-products and residues associated with industrial processes involving NORM are of interest here in so far as they may become wastes eventually. Past and future practices are considered in the context of either remediation of previously disposed residues or management of newly generated residues. Some information on uranium mining and milling is included in order to provide some perspective on how NORM is managed in that industry.

In order to identify suitable remediation strategies and technologies, an assessment is required of the potential sources of NORM, the typical processes leading to concentration enhancement and dispersal of NORM in the environment, and the resulting human exposure patterns (Fig. 1). To this end, the scope also includes an analysis of when and where in the relevant processes the concentrations of NORMs can be significantly enhanced or there is an enhanced potential for human exposure. Understanding these activities and

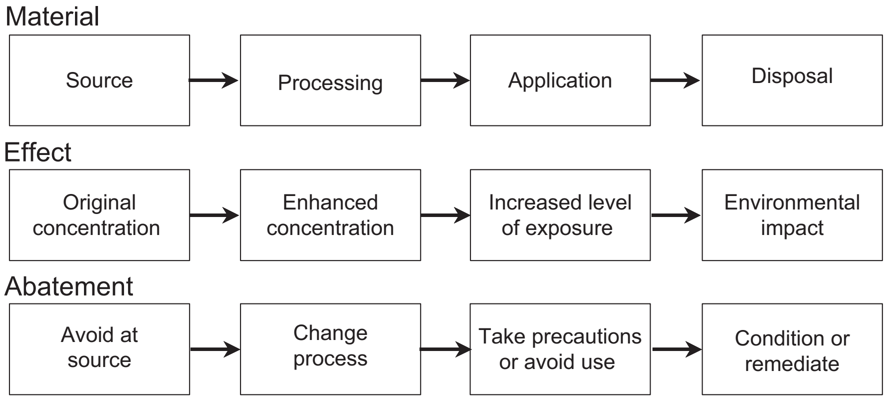

FIG. 1. Logic for the development of remediation technologies for NORM contamination.

processes is the basis for understanding the waste arisings, waste forms produced, related modes of disposal, and thus the potential for contamination situations that can lead to enhanced public exposure. In general, the types of information needed are those required to conduct an environmental impact statement, although in a more general sense.

The report also addresses possible changes in production and waste management technologies in order to reduce the arisings of wastes or their radioactivity, or to change their form into one more manageable or less harmful. Recycling and reuse of NORM containing residues is also addressed with an emphasis on the need for proper consideration of the potential for human exposures to recycled or reused NORM.

This report also serves as a source of information regarding technologies that have been or are being considered for use to remediate sites contaminated with NORM. Furthermore, because contaminated sites result from TE-NORM, technologies that have been applied, or are being considered for application, to limit the amount of residues and/or the activity concentration of NORM containing residues resulting from a given process are also discussed. Technological options discussed include changes in the industrial process and feedstocks for the process in addition to changes in waste handling and management procedures, and the cleanup/remediation of historical wastes.

While it is understood that exposure of the public can be kept low under normal operational conditions for most industries, absence or loss of institutional control over contaminated lands on which large amounts of NORM containing residues have been placed can result in redevelopment, intrusion and uncontrolled reuse of NORM containing materials. Thus, this report will also discuss the importance of properly considering such possibilities in the scenarios used to define the clearance levels to be applied for NORM.

Development of specific criteria related to the issues of clearance, exemption and transborder movement of radioactive materials will not be discussed. Similarly, issues such as radiation protection at the workplace and direct exposure to NORM in consumer products are not subjects of this report. In the European context such issues have already been discussed extensively [5–7] and are the subject of a variety of IAEA guidelines now in draft form.

Likewise, the sources, impacts and abatement technologies associated with ^{222}Rn accumulation in buildings resulting from decay of ^{226}Ra which has not been enhanced in concentration as a result of human activity will not be covered in this report. However, radon exposures that may occur as a result of NORM containing residues which have been deposited on the ground surface or not sufficiently covered to attenuate the radon dose are relevant in the context of this report. For the case of NORM disposed near or on the ground surface or used in building materials, radiogenic radon can be a major source of potential exposure, and thus, radiological dose to the public. Typical cases are, for example, plasterboard made from phosphogypsum or slag and fly ash additives in concrete.

Although not specifically addressed in this report, the issue of co-contaminants, such as heavy metals, also needs to be addressed, as these can have technological as well as regulatory implications. Remediation and waste management strategies and techniques have to be tailored in such a way that, when solving the NORM problem, no additional problems arise. For example, thermal waste treatment processes, which may be effective for the volume reduction of many wastes, will lead to the volatilization and subsequent accumulation/enhancement of polonium and lead isotopes along with mercury in residues.

1.4. RELATED IAEA PUBLICATIONS

The IAEA has developed a comprehensive programme, aiming at the collation and dissemination of knowledge about contaminated sites. The programme addresses appropriate methods for their characterization [8], assessment of their potential environmental and radiological impact [9], and applicable methods for their cleanup [10], quality control and monitoring [11] in accordance with internationally recommended safety criteria [12].

It is intended to list selected sites contaminated by NORM, for instance uranium mining and milling sites, in the Directory of Radioactively Contaminated Sites (DRCS) [8]. However, NORM containing wastes that have already been declared ‘radioactive wastes’ would be reported in the IAEA’s Net-Enabled Waste Management Database (NEWMDB) [13].

1.5. ORGANIZATION

The main text of this report begins with an overview of naturally occurring radionuclides, followed by a brief summary of processes that can lead to enhanced concentrations of naturally occurring radionuclides. Individual sections are then provided for the different industries that can involve such processes. Each of these industry specific sections includes a brief summary of the process, a description of where enhancement can arise within the process, ranges of amounts and activity concentrations of the NORM, and an overview of technologies that have been applied, or are being considered for application in order to limit the amount or activity concentration of NORM that arises. A number of issues associated with potential public exposures to residues containing NORM are summarized following the specific discussions for individual industries. A series of appendices describe in more detail specific examples of industries involving NORM in selected Member States.

2. NATURALLY OCCURRING RADIONUCLIDES

NORM comprises radionuclides associated with the ^{238}U and ^{232}Th decay chains as well as ^{40}K. These radionuclides are very long lived and have some progeny that are long lived, such as ^{226}Ra. The distribution of radionuclides in the geosphere depends on the distribution of the geological media from which they are derived and the processes which concentrate them at a specific location in specific media. The key to understanding these distributions, therefore, is to understand the distribution of the source materials and the physical and geochemical processes that lead to elevated concentrations of radionuclides under specific conditions.

The tables that follow provide a perspective regarding the concentrations of radionuclides in different natural materials. Table I includes a summary of typical concentrations of radionuclides in different geological media. Table II includes a summary of concentrations of radionuclides in natural resources that when exploited can result in enhanced NORM concentrations. It should be noted that the NORM problem often goes hand in hand with elevated concentrations of other elements that are considered toxic or hazardous, such as arsenic or heavy metals. Thus, in a comprehensive approach, the potential hazards associated with non-radioactive substances should also be considered.

3. PROCESSES ENHANCING CONCENTRATIONS AND THE POTENTIAL FOR PUBLIC EXPOSURE TO NORM

3.1. ENHANCED CONCENTRATIONS VERSUS ENHANCED EXPOSURE POTENTIAL

NORM in its unaltered natural state can pose potential radiological concerns. However, such unaltered NORM is mostly not amenable to regulatory control. On the other hand, exposures to NORM that has been altered in the course of exploitation of natural resources to meet human needs can, in principle, be controlled. There are two effects of human exploitation that

TABLE I. SUMMARY OF CONCENTRATIONS OF MAJOR RADIONUCLIDES IN MAJOR ROCK TYPES AND SOIL
(data from Ref. [14], other references are given therein)

	^{40}K		^{87}Rb		^{232}Th		^{238}U	
Rock type	Total K (%)	Bq/kg	ppm	Bq/kg	ppm	Bq/kg	ppm	Bq/kg
Igneous rocks								
Basalt, crustal average	0.8	300	40	30	3–4	10–15	0.5–1	7–10
mafic	0.3–1.1	70–400	10–50	1–40	1.6, 2.7	7, 10	0.5, 0.9	7, 10
salic	4.5	1100–1500	170–200	150–180	16, 20	60, 83	3.9, 4.7	50, 60
Granite (crustal average)	>4	>1000	170–200	150–180	17	70	3	40
Sedimentary rocks								
Shale, sandstones	2.7	800	120	110	12	50	3.7	40
Clean quartz	<1	<300	<40	<40	<2	<8	<1	<10
Dirty quartz	2?	400?	90?	80?	3–6?	10–25?	2–3?	40?
Arkose	2–3	600–900	80–120	80	2?	<8	1–2?	10–25?
Beach sands (unconsolidated)	<1	<300?	<40?	<40?	6	25	3	40
Carbonate rocks	0.3	70	10	8	2	8	2	25
Continental upper crust (average)	2.8	850	112	100	10.7	44	2.8	36
Soils	1.5	400	65	50	9	37	1–8	66

Note: Question marks indicate estimates in the absence of measured values.

are relevant in the case of potential effects of NORM on human health and the environment:

(1) The concentrations of NORM can be enhanced above its natural levels in a product, by-product or residue.
(2) The availability for release into the biosphere of the NORM in products, by-products or residues can be enhanced through physicochemical changes or simply due to the method by which the residues are managed.

TABLE II. NATURALLY OCCURRING RADIONUCLIDES IN MINERAL RESOURCES [15]

Element/mineral	Source	Radioactivity
Aluminium	Ore	250 Bq/(kg U)
	Bauxitic limestone, soil	100–400 Bq/(kg Ra)
	Bauxitic limestone, soil	30–130 Bq/(kg Th)
	Tailings	70–100 Bq/(kg Ra)
Copper	Ore	30–100 000 Bq/(kg U)
	Ore	20–110 Bq/(kg Th)
Fluorspar	Mineral	Uranium series
	Tailings	4000 Bq/(kg Ra)
Iron		Uranium series
		Thorium series
Molybdenum	Tailings	Uranium series
Monazite	Sands	6000–20 000 Bq/(kg U)
		Thorium series (4% by weight)
Natural gas	Gas, average for groups of US and Canadian wells	2–17 000 Bq/(m^3 Rn)
	Gas, individual US and Canadian wells	0.4–54 000 Bq/(m^3 Rn)
	Scale, residue in pumps, vessels and residual gas pipelines	100–50 000 Bq/(kg ^{210}Pb/^{210}Po)
Oil	Brines or produced water	Ranging from mBq to 100 Bq/(L Ra)
	Sludges	Ranging up to 70 000 Bq/(kg Ra)
	(scales)	Typically 10^3–10^4 Bq/kg, ranging up to 4×10^6 Bq/(kg Ra)
Phosphate	Ore	100–4000 Bq/(kg $U_{natural}$)
	Ore	15–150 Bq/(kg $Th_{natural}$)
	Ore	600–3000 Bq/(kg Ra)
Potash		Thorium series
		^{40}K
Rare earths		Uranium series
		Thorium series
Tantalum/niobium		Uranium series
		Thorium series

TABLE II. (cont.)

Element/mineral	Source	Radioactivity
Tin	Ore and slag	1000–2000 Bq/(kg Ra)
Titanium (rutile, ilmenite)	Ore Ore	30–750 Bq/(kg U) 35–750 Bq/(kg Th)
Uranium	Ore Slimes Tailings	15 000 Bq/(kg Ra) 10^5 Bq/(kg Ra) 10 000–20 000 Bq/(kg Ra)
Vanadium		Uranium series
Zinc		Uranium series Thorium series
Zirconium (zircon)	Sands Sands	4000 Bq/(kg U) 600 Bq/(kg Th) 4000–7000 Bq/(g Ra)

This section includes a brief overview of these two concepts and their impact on the potential for and the magnitude of radiological effects associated with the management of residues resulting from exploitation of natural resources. Emphasis is placed on the fact that it is not only technologically enhanced concentrations of NORM that are a concern but also NORM that has been altered from its natural state, and is thus more available for release into the biosphere or for direct human exposure.

3.2. TECHNOLOGICALLY ENHANCED CONCENTRATIONS

The concentration or dispersal of radionuclides, like that of any other chemical element, is controlled by its physicochemical properties in relation to the ambient conditions. Human activities change these ambient (environmental) conditions intentionally, for example in situ leaching of uranium ore, or unintentionally, for example the extraction and production of oil and gas. Changes in the physicochemical conditions can lead to the enrichment of the concentration of radionuclides, in particular products or residues in the course of industrial activities. A range of principal processes can be identified, for example:

(a) Extraction of groundwater (for drinking water purposes, as formation water in oil and gas recovery) typically changes the chemical boundary conditions (pH, redox potential and gas partial pressures), and chemical equilibria are disturbed, which can lead to the precipitation of scales (which scavenge, for instance, radium); conversely, 'mixing corrosion' resulting from the mixing of waters with different CO_2 partial pressures can lead to the dissolution of scales.

(b) Combustion processes volatilize certain nuclides, such as lead and polonium, and concentrate non-volatile nuclides, such as radium, in the ash.

(c) Physical disaggregation enhances dissolution and dispersal owing to increased surface areas.

(d) Sedimentation can lead to accumulation of naturally occurring radionuclides, which are often associated with heavy minerals, for example, rutile and zircon.

(e) (Preferential) adsorption on clays or organic sedimentary fractions can lead to enhanced concentrations in these fractions; size fractionation (sieving, floatation, etc.) leads to further concentration enhancement in fines and slimes.

3.3. ENHANCED POTENTIAL FOR HUMAN EXPOSURE TO NORM

The activities described in Section 3.1 could also lead to an increased availability of the radionuclides for potential human exposures. In some cases, the enhancement in concentration of NORM may be insignificant or relatively small, but a large amount of NORM could be disrupted from their natural state. This is most apparent in the case of the enhanced potential for human exposure as a result of metal or mineral mining. Mining activities often involve extraction of a given valuable fraction of a very large ore body, leaving the remainder as a residue. In this case, the concentrations of the NORM may not be significantly enhanced in the mining residues (e.g. tailings and gangue), but very large amounts of NORM are more available for release into the biosphere than they were in the undisturbed natural state. This illustrates human activities that do not necessarily enhance the concentration of NORM in products, by-products or residues can also be a concern, simply due to the increased potential for human exposure.

It should be noted that, when compared with the amount of declared radioactive waste that is disposed in radioactive waste repositories, the amounts of NORM containing residues are orders of magnitude larger. Management of such large amounts of residues poses a significant challenge for these industries.

The following sections provide a summary of the material flows for selected processes, including examples of the primary feed materials, end products, and the ensuing residues and existing contamination problems with an emphasis on the fate of the NORM. The intention is not to give a comprehensive description of each process, rather to describe the various processes or product streams in enough detail to raise the awareness of NORM within different industries and provide a perspective on areas where concentrations of radionuclides can be enhanced in products, by-products or residues or where the potential for human exposure to NORM in these materials is enhanced as a result of industrial activity. Indications are also given of where these exposures could be decreased.

4. FUELS

4.1. OIL AND GAS

4.1.1. Introduction

Oil and gas are used in a number of different ways, including: fuel for electricity production and transport, lubricants, and numerous industrial and consumer products. In terms of the raw materials, gas and oil originate from the breakdown of organic matter in a sedimentary formation (the source rock) and may become trapped in a suitable reservoir structure. Many types of reservoir structures are known, but all require that the vertical migration of oil and gas from the porous reservoir rock be prevented by an impervious covering known as the caprock (often rock salt or clay) [16]. Global crude oil production for 2000 is summarized in Table III and crude oil production since 1971 is illustrated in Fig. 2. Global natural gas production for 2000 is summarized in Table IV and production since 1971 is illustrated in Fig. 3.

Table I provided ranges of typical concentrations of radionuclides in different geological materials. Radionuclide concentrations are not exceptionally high in common sedimentary materials; however, elevated levels can be found in shales. The radionuclides tend to exist in equilibrium in rock formations. The formation water, often a brine with low sulphate concentration, preferentially dissolves radium relative to the parents uranium and thorium. Thus, radium and its progeny are present in larger concentrations than uranium and thorium in the water. As noted above, the fraction of water in the oil–gas–water mixture tends to increase during the time in which a reservoir is exploited. Thus, NORM in extraction and separation facilities for a specific reservoir becomes more prevalent over time. Radon emanating from the rock formation and resulting from decay of radium in the water will be present in the gas phase.

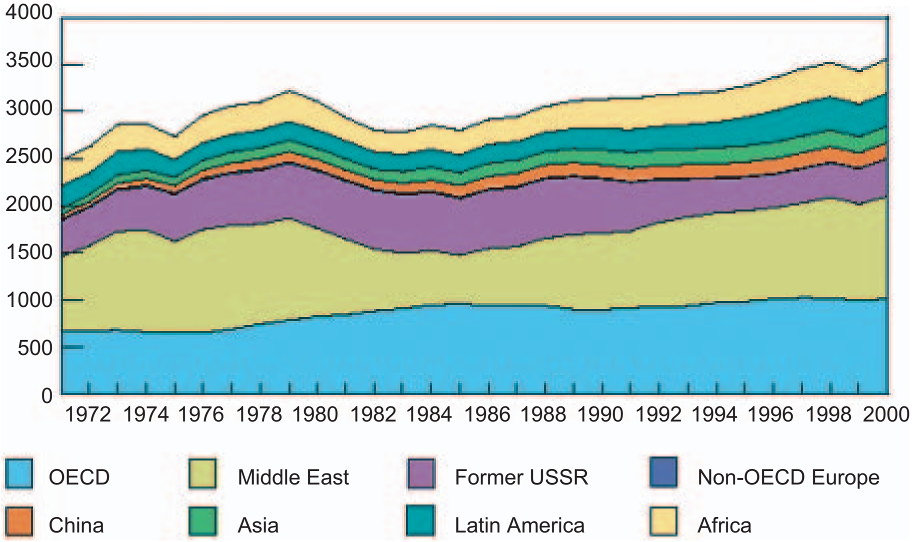

FIG. 2. Global crude oil production (in Mt) since 1971 (from Ref. [17]).

4.1.2. Processes

The general components of the oil and gas extraction and front end production process are illustrated in Fig. 4. In general, the oil–gas–water

TABLE III. GLOBAL CRUDE OIL PRODUCTION IN 2000
(from Ref. [17])

Producer	Mt	Percentage of world total
Saudi Arabia	427	12.0
United States of America	354	10.0
Russian Federation	322	9.1
Islamic Republic of Iran	186	5.2
Venezuela	172	4.8
Mexico	171	4.8
China	163	4.6
Norway	158	4.4
Iraq	127	3.6
United Kingdom	127	3.6
Rest of the world	1348	37.9
World total	3555	100.0

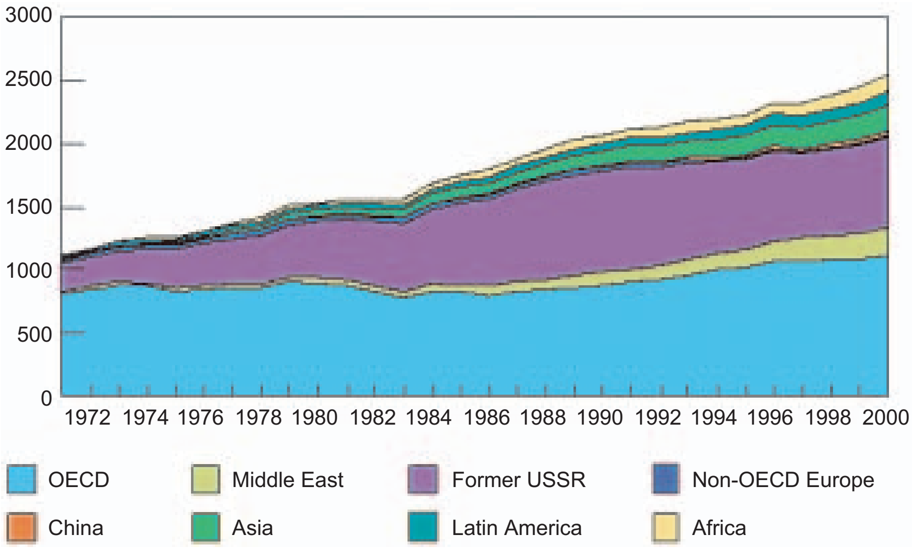

FIG. 3. Global natural gas production (in Mm^3) since 1971 (from Ref. [17]).

mixture is brought to the surface through the well tubing. This mixture is then processed through a separator which removes the gas. The gas is then processed and transported to a gas plant where it is divided into the different gas

TABLE IV. GLOBAL NATURAL GAS PRODUCTION IN 2000
(from Ref. [17])

Producer	Mm^3	Percentage of world total
Russian Federation	584 200	23.0
United States of America	548 044	21.6
Canada	180 618	7.1
United Kingdom	115 065	4.5
Algeria	89 348	3.5
Netherlands	72 864	2.9
Indonesia	67 830	2.7
Iran	63 744	2.5
Uzbekistan	56 001	2.2
Saudi Arabia	52 811	2.1
Rest of the world	706 341	27.8
World total	2 536 866	100.0

products. After the gas has been removed, the oil–water mixture is sent to a heater/treater and divided into the oil and water phases. The separated crude oil phase is distributed to stock tanks prior to being transported to a refinery. Additional gas which can be obtained in the heater/treater and in the stock tanks is also routed to the gas plant. After being separated from the oil, the formation water is routed to a water treatment plant and then discharged to the sea or held in (often unlined) open storage basins for further use or reinjected into the oil/gas bearing formation.

4.1.3. Residues

Oilfield equipment can contain hard radioactive scales and softer scale bearing sludges which appear as coatings or sediments [18]. Gas field equipment may be contaminated internally with films, coatings or plating of concentrated NORM [19]. The scales and sludges containing NORM are formed primarily during the extraction and production stages, starting when the oil–gas–water mixture is brought to the surface and also forming during the separation process. Initially, NORM is brought to the surface through the downhole tubing as part of the oil–gas–water mixture. Hard scales precipitate from the formation water onto the downhole tubing in response to changes in temperature, pressure and salinity as the water is brought to the surface. The scale is typically a mixture of carbonate and sulphate minerals. One of these sulphate minerals is baryte ($BaSO_4$), which is known to readily incorporate radium in its structure. When compared with other NORM containing wastes in the oil and gas industry, the concentrations of ^{226}Ra are the largest in these scales and are enhanced substantially from concentrations found in the natural environment.

During the separation process, different radionuclides in the decay chain are also separated. Figure 4 provides a rough illustration of the flow of the different radionuclides in the separation process. In the separation process, the deposits tend to be in the form of soft scales, sludges or films. Radium sludges tend to accumulate on the oil and water side of the separation process, especially in areas where there are changes in pressure or temperature and where the oil products are stored or water is discharged into (unlined) surface pits.

Likewise, radon and its progeny can be found in the gas and in films on the inside of the gas handling equipment. The radon appears to concentrate preferentially in the ethane and propane streams relative to the natural gas output of the plant [20]. Lead-210 and ^{210}Po that accumulate in films are especially problematic because they are more difficult to measure on the inside of piping and equipment due to the lack of a strong gamma decay. These

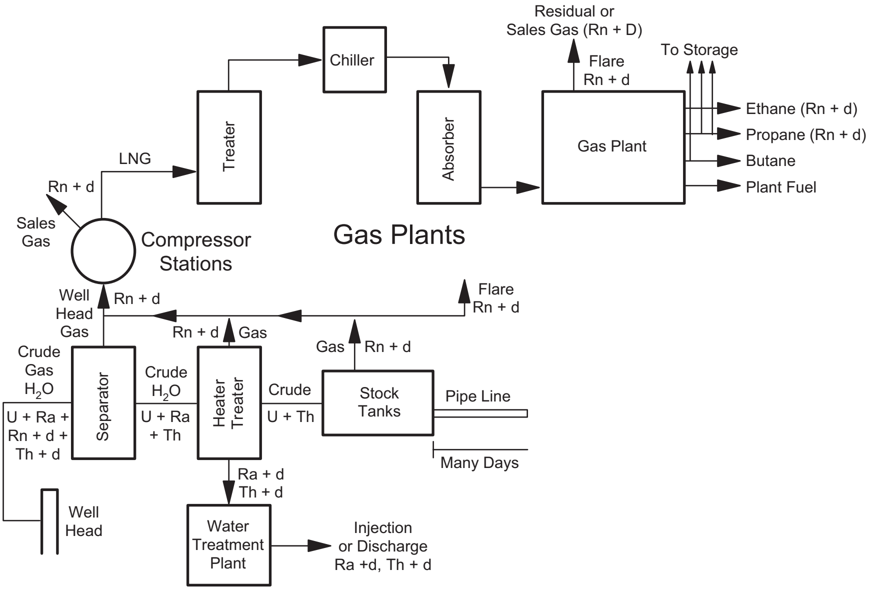

FIG. 4. Example flows of NORM in oil, gas and water separation ('d' stands for daughters). (By courtesy of M. Scott (Louisiana State University, Baton Rouge, Louisiana, USA).)

deposits can be found throughout the natural gas systems. In order to measure for ^{210}Pb and ^{210}Po, it is necessary to open the piping or valves and to swab the inside of the pipe or to use a pancake type Geiger–Müller detector [19].

As introduced above, the process residues containing technologically enhanced NORM associated with the oil and gas industry occur in the form of scales, sludges and films. There are also additional NORM containing residues or contaminated soils from the water discharges produced. The amounts and characteristics of these residues vary considerably in different installations [18]. Data for oil wells in the United States of America suggest that an average of roughly 100 t of scale per well is generated each year. Many wells are known to not currently yield residues containing NORM. Accounting for this yields an estimate of roughly 400 t of scale per year for each NORM generating well. Van Weers and co-authors [16] provide a number of examples of amounts of scales. It is known that the amount of scale increases as a well ages due to the increasing ratio of water to oil and in some cases the introduction of salt water into the formation to enhance recovery.

An American Petroleum Institute report [21] estimated that more than 140 000 drums (roughly 28 000 m^3) of technologically enhanced NORM containing residues (≳3300 Bq/kg) accumulate on an annual basis in the USA.

Estimates for the North Sea [22] suggest a somewhat lower value of 20 t of scale per well each year, although this value is based on a single, new well field which would be expected to generate less scale than some of the fields that have been operating for longer periods of time.

Another residue is the formation water that has been separated from the oil–gas–water mixture. Studies of the large quantities of water produced from wells at oil and natural gas drilling and production sites have indicated that a number of the wells yielded water with an average radium concentration in excess of 1.85 Bq/L [23]. Other data suggest that average radium concentrations in water from some wells can be as high as 111 Bq/L [24]. As a comparison, the radium concentration limit for discharges of water from US commercial nuclear facilities is approximately 2.2 Bq/L [25].

As noted above, the separated water is often re-injected into the oil bearing formation, which is considered in some Member States a sound waste management approach posing minimal potential impacts to human health, provided that there is no hydraulic connection to usable groundwater resources and that once the well is abandoned, the casing is properly cemented to prevent leakage into these aquifers. However, in many cases, the water is discharged into holding surface ponds (in the case of a land based well) or discharged directly into the sea (in the case of an offshore well). When discharged into ponds, the water is generally allowed to percolate into the ground or to evaporate. In this case, NORM in the water will concentrate in the bottom sludges or residual salts of the ponds. Thus, the pond sediments can become a residue posing a potential radiological health concern. The activity concentrations of radionuclides in these soils have been reported to be in the range from 1×10^4 to 4×10^4 Bq/kg.

Table V includes illustrative ranges for the activity concentrations of radionuclides in different residues associated with oil and gas. The largest concern in terms of radionuclide activity concentrations for NORM in the oil and gas industry involves the hard scales which form on the inside of the downhole tubing. At many oilfield sites the downhole tubing can become coated with scale deposits that contain radium. Because of the loss of flow capacity in the downhole tubing, these scales must be removed occasionally. When they are removed, they become a waste management concern. The composition of scale has been studied in detail for a number of years. NORM scale and sludges in production equipment have been shown to have radionuclide activity concentrations as high as several million Bq/kg [24, 26], which would place them at the upper end of the concentrations suitable for near surface disposal (these are extreme cases and not indicative of typical values).

Average radionuclide concentrations of 1000–100 000 Bq/kg are more common [23]. In terms of exposure, the scales tend to be relatively insoluble,

TABLE V. REPRESENTATIVE NORM ACTIVITY CONCENTRATIONS IN OIL AND GAS RESIDUES
(adapted from Ref. [29])

Material	Example activity concentrations (Bq/kg)
Scale in downhole tubing, pipes and other equipment for handling oil/gas and formation waters	^{226}Ra: Background to 15 000 000 (average 1000 to hundreds of thousands)
Sludges in separations and production equipment	^{226}Ra: 10 000 to 1 000 000
Sludges, films in natural gas supply equipment	^{210}Pb: Background to about 40 000
Sludges from soils beneath ponds of produced water	^{226}Ra: 10 000 to 40 000

thus the radionuclides would only be released slowly into the environment [27]. Furthermore, owing to the nature of the scales, the emanation coefficient of radon is substantially less than that of other materials such as mill tailings or sludges [28].

Sludges that accumulate in tanks and other settling areas within the production equipment tend to have less enhancement of NORM than the scales; however, the concentrations can still be a concern. Table V indicates ranges from background to roughly 1×10^4 to 4×10^4 Bq/kg. Sludges and films in natural gas equipment show a similar range of concentrations. Films of ^{210}Pb and ^{210}Po in gas plants have been found in higher concentrations [26]. The radionuclides in sludges are more available for release into the environment than was the case for the scales. Thus, although radionuclide activity concentrations are less than those of scales, potential exposures via the food chain and radon pathways are more likely.

4.1.4. Management of residues

Figure 5 illustrates the relative isolation of residues containing NORMs from the general public for a variety of possible disposal options. As the degree of isolation increases so does the capability for disposing of higher radium concentrations. Surface spreading and dilution of low activity NORM is a past practice that is now disallowed by most States in the USA with NORM regulations. The American Petroleum Institute has prepared a NORM disposal cost study [21] that includes an evaluation of different disposal options as

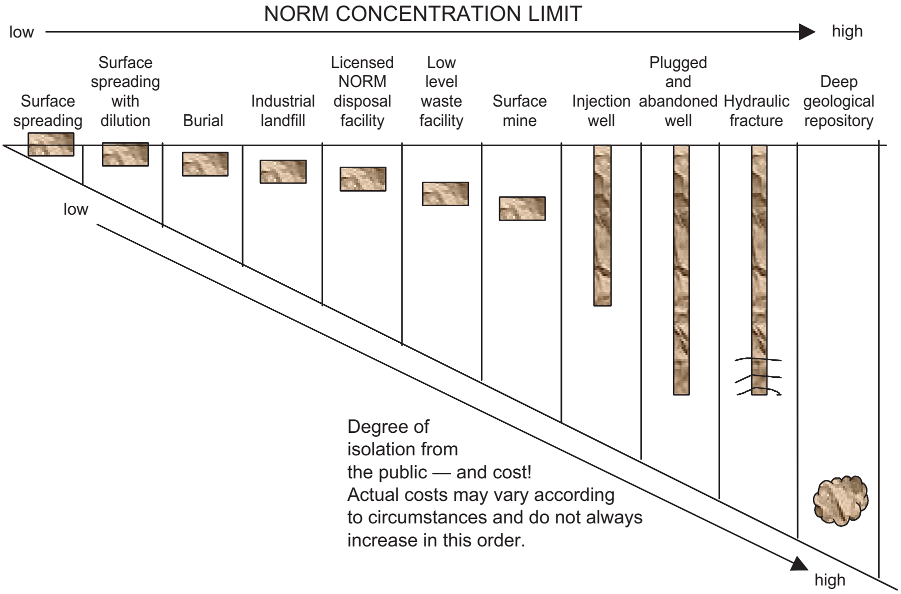

FIG. 5. Disposal alternatives for NORM wastes. Disposal of more concentrated wastes requires greater isolation of waste from the general public (from Ref. [30] based on an original American Petroleum Institute figure [31]).

illustrated in Fig. 5. An additional method of disposal is direct discharge into the sea of scales that have been ground to a small size. Although, in principle, this dilute and disperse management approach should lead to minimal health impacts, data have shown that large mounds of the scale can accumulate beneath a drilling and production platform.

Landfarming (Fig. 6) or other approaches involving mixing of NORM containing residues in surface soils are used for management of lower activity sludges and some scales. In this approach, the concentration of NORM is diluted in the soil to a level considered acceptable on the basis of applicable regulations.

Management approaches for the ‘produced’ water that is separated from the oil and gas vary from location to location. In some cases, both onshore and offshore, the separated water is reinjected into the oil/gas bearing formation. However, in other cases it is discharged into surface ponds for evaporation or discharged directly into the sea in the case of offshore rigs. When discharged into evaporation ponds, the resulting NORM containing sludges and salts require proper treatment and disposal.

The decommissioning of oil and gas production facilities gives rise to a waste stream of contaminated metal materials, mainly tubing and fittings [32].

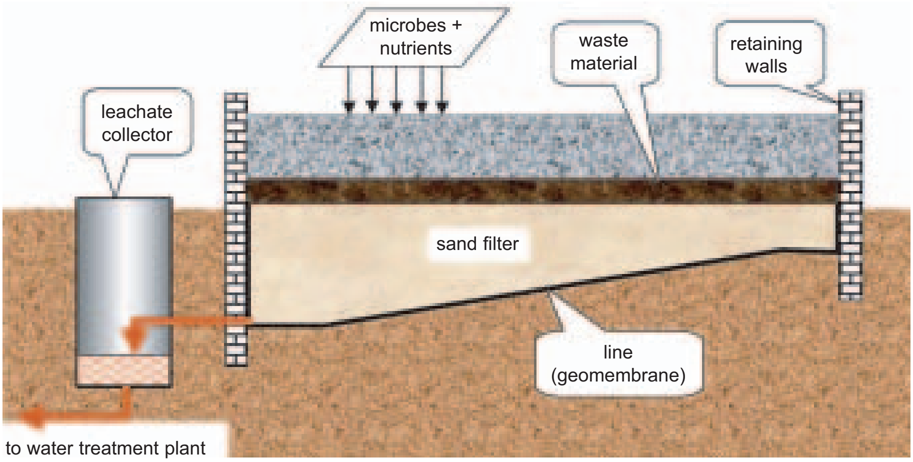

FIG. 6. Landfarming to treat oil production scales and sludge.

Here, contamination with ^{210}Pb is also of concern, as this can only be detected from the inside, owing to the low penetration depth of its radiation. However, sludges and scales present in a facility should be dealt with appropriately as part of the decommissioning strategy (Fig. 7). Some options for disposal of decommissioned offshore platforms are deep water disposal, creation of artificial reefs and partial removal to depths of 26 m or more [33]. These approaches take advantage of natural dilution in the vast amounts of sea water, but may have implications vis-à-vis the London Convention (LC) [34].

In reviewing the analysis of the NORM situation in the oil and gas industry in the European Union, and more specifically for North Sea offshore production [18, 29], one can conclude that very little need for environmental remediation is likely to arise. The routes of disposal for both operational and decommissioning residues vary from country to country and include discharge into the sea as well as land storage in low level storage facilities, such as COVRA in the Netherlands [35]. The agreements under OSPARCOM [36] aim to eliminate sea disposal and discharge. The situation is similar in the USA, although national requirements have not been developed for disposal of residues containing NORM from the oil and gas industry [21]. However, only limited information is available for other areas, where there may still be concerns about how these residues are managed and the issue of NORM has only recently gained more attention. Environmental contamination problems may arise when scales are dumped into the sea from offshore platforms or dispersed in terrestrial environments. The windborne dispersal of scales, when not properly managed, can be a problem in arid countries [37]. Abandoned sites with NORM contaminated soil, where produced

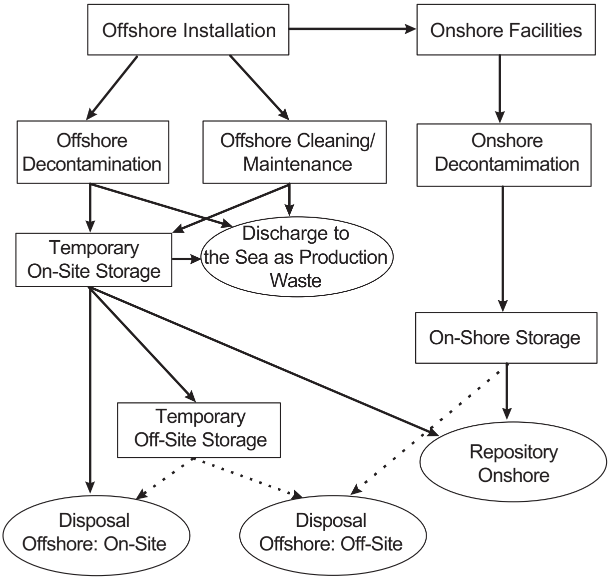

FIG. 7. Disposal chart — NORM waste in oil and gas production [33].

water was discharged into unlined ponds, could be of radiological concern when people move onto the site.

4.1.5. Residue management technologies

The interest in mobilization processes leading to deposition of scales is not exclusively related to NORM, as considerable scale formation interferes with the production process by reducing the free diameter of tubing and consequently increasing the flow resistance. Scale deposition also poses a risk by affecting the proper functioning of valves. The oil and gas industry has been developing and applying 'scale inhibitors' to help limit the amount of scale that forms on the inside of the pipe. A variety of different inhibitors have been applied to limit precipitation of the solids, which keeps the radium in solution rather than concentrated in the solid phase (see, e.g. Ref. [38]). Such inhibitors have been used with varying levels of success. In addition, methods for scale inhibition by injecting chemicals into reservoirs to change the formation/injection water chemistry have been developed and are now being tested in practice.

The same applies to the use of acids and proprietary mixes to dissolve scale in situ [16, 39]. The effective use of scale inhibitors, however, carries the radium isotope with the produced water through the production system. The radium can still accumulate, although in lower concentrations, in sludges or other residues associated with water handling.

Decontamination methods include mechanical scrubbing, shot blasting and high pressure water jetting. The resulting slurries are collected for disposal in low level radioactive waste disposal facilities. At least in the past, scales were also ground up and discharged from offshore installations or reinjected downhole. In certain instances scrap melting is also possible, when the NORM is collected in the slags and by scrubbing off-gases [16]. However, in some Member States recycling of radioactive contaminated metals is not an acceptable practice, and the materials must be disposed of.

Facilities where scale from pipes, tanks and other oilfield equipment was mechanically removed without awareness of its radiological contamination, and where the scale was left on-site, may also pose public health hazards to future occupants and nearby residents due to redistribution by dust generation or water erosion. Reuse of contaminated pipes as structural elements in buildings or playground equipment, or melting of contaminated metals for recycling to other uses, may also expose the public to unsuspected radiological hazards.

4.2. COAL MINING AND COMBUSTION

4.2.1. Introduction

Extraction of hard coal worldwide is about 3.7–3.8 Gt (Table VI) with consumption likely to increase in the future (Fig. 8). The biggest producers of coal are China, USA and India, which together produce more than two thirds of the world total.

Fossil fuels such as coal contain naturally occurring radioactivity from the uranium and thorium series and ^{40}K. The concentration of NORM in the coal, waste rocks and waste waters depends on the geological formation of coal seams. The concentration of NORM in the ash and other residues is closely

TABLE VI. WORLDWIDE PRODUCTION OF COAL (Gt)

Year	1992	1993	1994	1995	1996	1997	1998	1999	2000
All types of coal [40]	4.580	4.492	4.559	4.682	4.686	4.703	4.652	4.532	4.538
Hard coal [41]	3.486	3.431	3.553	3.715	3.797	3.821	3.758	3.666	3.639

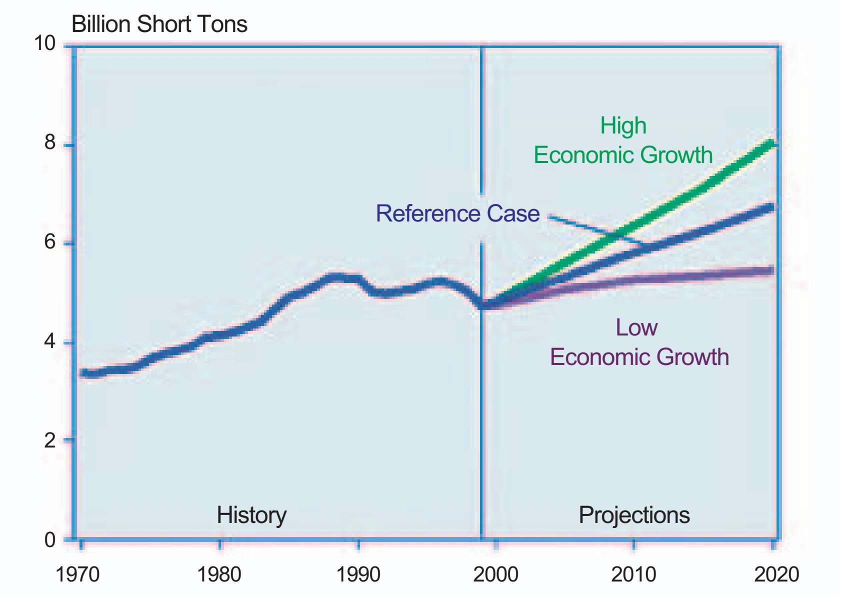

FIG. 8. Consumption of coal worldwide, with prognosis up to 2020 [40].

related to the NORM concentrations in the coal used as fuel. In order to accurately address these questions and to predict the mobility of radioactive elements during the coal fuel cycle (from mining to coal combustion), it is important to determine the concentration, distribution and form of radioactive elements in coal, waste rocks, waste waters, ash, slag and other residues [42].

As with other fuels, the concentration of NORM in coal or lignite depends on the geological setting. Coal forms by the accumulation of non-marine plant debris. As this organic sediment buried under stagnant swamp waters, increased pressure and temperature begin to break down the organic matter. The formation of coal involves a series of steps with the progressive release of volatile components, enriching the residue in carbon. These released gases include water, carbon dioxide and methane. As temperature and pressure increase, peat is transformed successively into lignite; bituminous or soft coal; and finally anthracite, or hard coal. Coal is characterized by the degree of carbonization, or in other words by its contents of carbon, ash (non-combustible inorganic matter), moisture and volatile materials. Heavy metals and radionuclides are often associated with the sulphidic accessories in coal.

Some examples of NORM concentrations in coal from selected countries are provided in Table VII. Note that the concentrations can be significantly

TABLE VII. EXAMPLE RANGES AND/OR AVERAGES OF RADIONUCLIDE ACTIVITY CONCENTRATIONS IN COAL (Bq/kg)

Country	^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th	^{228}Ra	^{40}K
Australia [44]	8.5–47	21–68	19–24	20–33	16–28	11–69	11–64	23–140
Brazil [45][a]	72		72	72		62	62	
Egypt [46]	59		26			8	8	
Germany [48, 49]			10–145			10–63		10–700
			32[a]			21[a]		225[a]
Lignite			<1–58			<1–58		<4–220
Former East Germany			10[a]			8[a]		22[a]
Greece [5][b]	117–390		44–206	59–205			9–41	
Hungary [48]	20–480					12–97		30–384
Italy [49][c]	23 ± 3					18 ± 4		218 ± 15
Poland [50]	<159					<123		<785
	18[d]					11[d]		
Romania [5]	<415		<557	<510	<580	<170		
	80[a]		126[a]	210[a]	262[a]	62[a]		
UK [44]	7–19	8.5–25.5	7.8–21.8			7–19		55–314
USA [44]	6.3–73		8.9–59.2	12.2–77.7	3.3–51.8	3.7–21.1		

[a] Average.

[b] Lignite.

[c] Lignite, average.

[d] Average for all coal seams.

higher in some areas. For example, concentrations of NORM greater than 1000 Bq/kg in coal have been reported in Croatia [43]. Examples of coal concentrations in gangue or waste rock from coal mining in China are provided in Table VIII.

TABLE VIII. OUTPUT-WEIGHED AVERAGE OF RADIOACTIVITY CONTENT IN GANGUE IN CHINA

Origin of gangue	Number of samples	Content		
		^{226}Ra	^{232}Th	^{40}K
Mining	526	53.1	61.2	530
Preparation	117	50.9	55.0	344
Total	643	53.0	61.0	524

4.2.2. Processes

Coal is excavated in open pits (mainly in the USA and Australia) or in underground mines (mainly in China, Europe and Brazil). The excavation of coal is accompanied by production of comparable quantities of waste rocks and large volumes of drainage water (Fig. 9) that can contain enhanced radioactivity. The mining activity can affect the overlying geological strata, creating new pathways for radon exhalation and, hence, its entry to dwellings. In such cases elevated radon concentrations in buildings can be observed [51].

A flow chart of the materials in a typical coal mining process is shown in Fig. 10.

The radioactivity of coal in seams is generally not enhanced compared with the average values for earth crust [53]. The same is true for most of the overlying and underlying rocks excavated together with coal and deposited on the surface as spoils (Table IX).

The conventional combustion of coal in coal fired power plants results in the generation of bottom ash, which is collected at the bottom of power plant boilers, and fly ash, which is collected prior to being discharged from the stack. Fine particles, or fly ash, are also emitted from the stack. Flue gas desulphurization (FGD), which is commonly being implemented on coal fired power

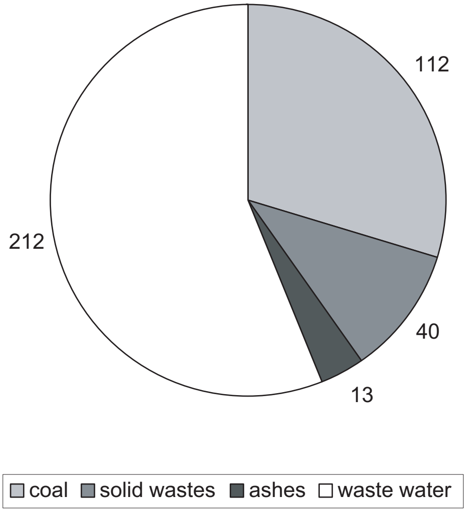

FIG. 9. Quantities of coal and wastes (in Mt) in the coal fuel cycle in Poland [52].

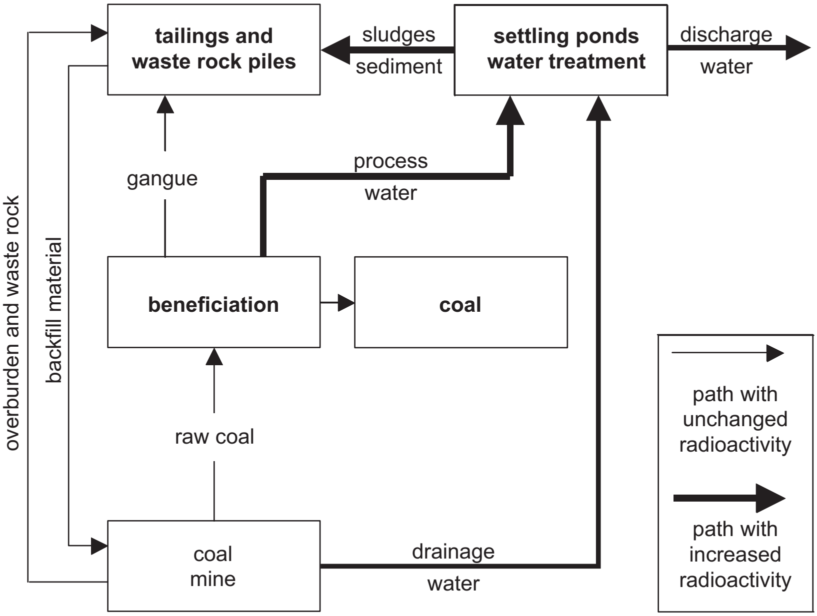

FIG. 10. Flow chart of the materials in the coal mining process.

plants, results in additional residues in the form of sludges or dry solids. The process flow for a typical coal fired power plant is shown in Fig. 11.

4.2.3. Residues

As shown in Fig. 11, most of the waste from a typical coal fired power plant is generated as fly ash, which is entrained with the hot flue gases of the combustion process. The remainder of the ash is heavier and settles to the

TABLE IX. AVERAGE ACTIVITY CONCENTRATION OF COAL AND WASTE ROCKS (Bq/kg)
(from Ref. [54])

Material	^{226}Ra	^{228}Ra	^{40}K
Coal	18	11	63
Waste rocks*	44–69	41–63	339–609
Soil in mining area*	20–25	20–23	252–307

* Values from two areas.

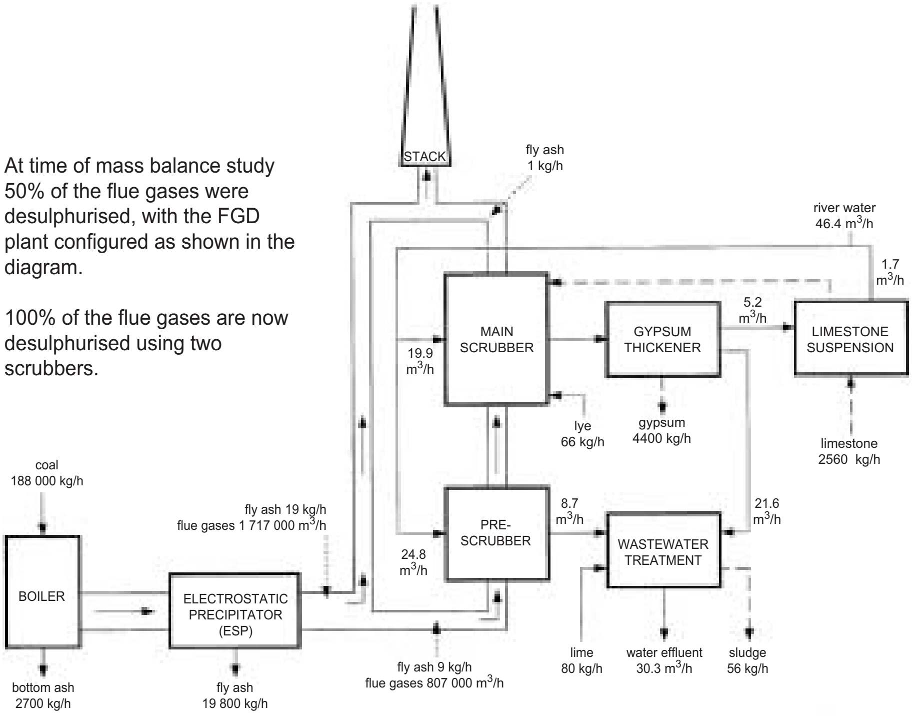

FIG. 11. Example of material flows in a coal fired power plant [55].

bottom of the boiler to form what is referred to as bottom ash. Volatile radionuclides, such as ^{210}Pb and ^{210}Po, accumulate in the fly ash and on the smaller particles emitted with the stack emissions. They can also accumulate on the inside of the stack as the exhaust gases cool. For the volatile radionuclides, there tends to be an inverse ratio between the particle size and the activity concentration. Table X provides some examples of the enhancement ratios for different radionuclides. The source in process materials and the subsequent partitioning of different elements in the various FGD residues is illustrated in Fig. 12. FGD residues are generated in large amounts as sludges and gypsum [47]; however, the NORM concentrations tend to be lower than in the ashes. Given that water and limestone are introduced into the system as part of FGD, it is important to understand the distribution of the elements in the source materials.

Tables XI and XII give examples of typical radionuclide concentrations found in residues resulting from coal combustion in a number of countries. These concentrations, however, can vary widely depending on the mineral

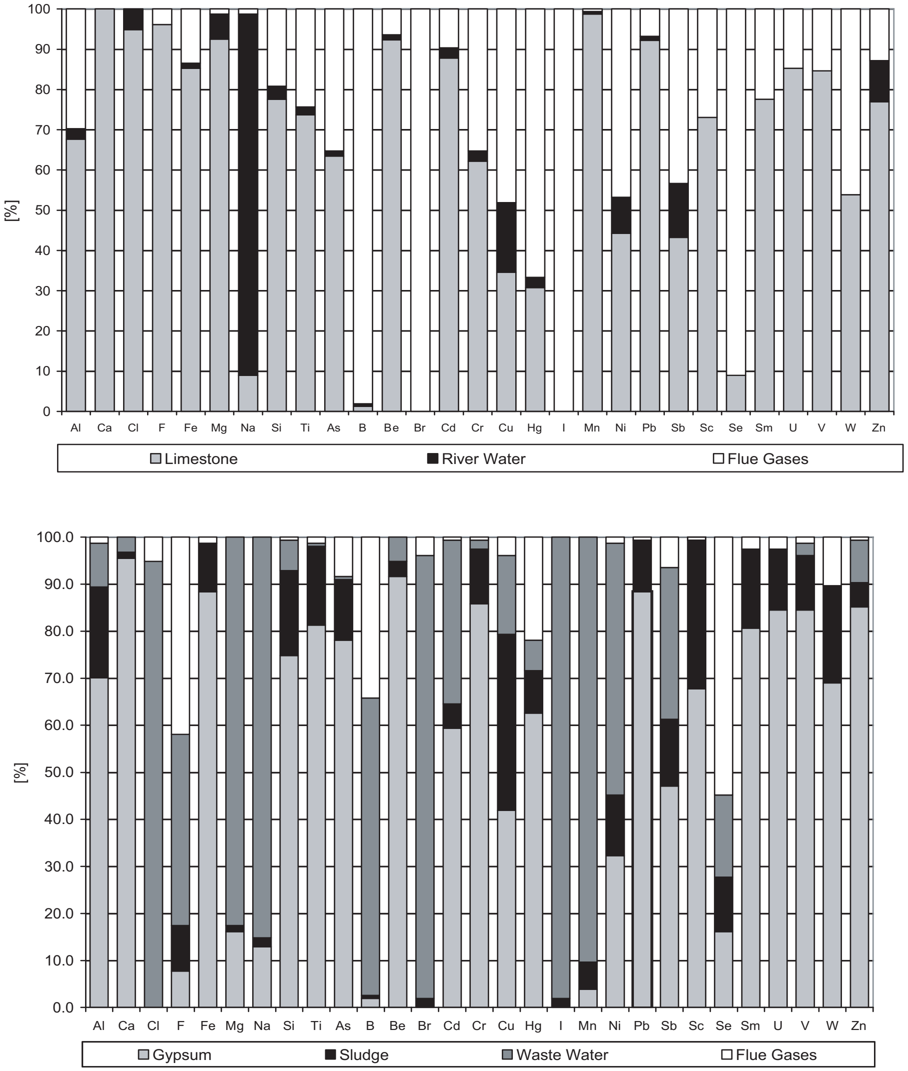

FIG. 12. Partitioning of elements in FGD process materials and resulting residues [45].

TABLE X. ENHANCEMENT FACTORS FOR RADIONUCLIDES IN ASH PARTICLES [5]

^{238}U	^{226}Ra	^{210}Po	^{210}Pb	^{232}Th	^{40}K
1–10	5–15	30–200	20–120	1–10	10–50

TABLE XI. MEASURED CONCENTRATIONS (Bq/kg) OF RADIONUCLIDES IN VARIOUS COAL BURNING RESIDUES

	^{238}U	^{234}U	^{230}Th	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	^{235}U	^{232}Th	^{40}K	Total
Fly ash emitted to the atmosphere from UK power stations [56]:											
Drax	109.7	121	34.5	53		188	171.3	<3.6	39.6		
Eggborough	84.9	88	30.6	74		125.4	139.8	<1.9	19.1		
Alberthaw	43.3	47.9	38.2	44.3		98	64.2	2.08	28.6		
High Marnham				<200		208	74				
Drakelow				<200		220	92				
West Burton				<400		290	158				
Polish coal fired power stations (1195 ash and 645 slag samples) [54]											
Ash (average)				146	102					631	
Ash (median)				131	101					654	
Ash (range)				18–870	16–275					35–1484	
Slag (average)				108	79					549	
Slag (median)				97	79					561	
Slag (range)				17–487	17–261					23–1103	
Croatian coal fired power plants [43]											
Fly ash	8700			2400	20			400		150	11700
Bottom ash and slag	3400			2000	60			200		290	5900
Brazilian coal and combustion products [57]											
Coal	72			72	62	72			62		
Bottom ash	156			120	84	70			96		
Fly ash	144			192	144	440			80		
US coal combustion wastes [58][a]											
Fly ash	96	96	67	111	96	200	207	5	63		1003[b]
Bottom ash and slag	26	26	19	26	22	52	52	1	15		255[b]

[a] Assumes that 80 wt% of the concentration is fly ash.

[b] Excludes contributions from other (short lived) radionuclides in decay chains.

TABLE XII. ACTIVITY CONCENTRATION RANGE (Bq/kg) IN COAL POWER PLANT RESIDUES (ASH, SLAG)

Country	^{238}U series	^{232}Th series	^{40}K
Hungary[a,b] [59]	200–2000	20–300	300–800
USA[a] [60]	100–600[a]	30–300[a]	100–1200[a]
Egypt [61]	16–41[a]	9–11[a]	
	41–90[b]	24–34[b]	
Germany [56]	6–166[a]	3–120[a]	125–742[a]
	68–245[b]	76–170[b]	337–1240[b]

[a] Fly ash or ash.

[b] Slags.

content of the coal, which, in turn, can vary with mining location and region of the country. There are also notable differences in residues from hard coal and lignite. Data from Brazil [57] show some enrichment during the combustion process for most radionuclides, but a significant enrichment for the volatile ^{210}Po in fly ashes. Ash data from Croatia for coal with relatively high concentrations of radionuclides are also provided in Table XI.

Depending on the hydrogeological conditions, the coal seams can be accompanied by waters containing high concentrations of radium. Radium is found in highly mineralized waters [62, 63], and in general radioactivity is correlated with water salinity. For example, in mine waters from the Upper Silesian Coal Basin the total salinity (total dissolved solids (TDSs)) may be sometimes higher than 200 g/L. The radium concentration in these waters can reach 390 Bq/L of ^{226}Ra and 120 Bq/L of ^{228}Ra. Radioactivity values in mine waters and associated sediments are exemplified in Table XIII. The radium in such brines can co-precipitate as (Ra, Ba)SO_4, provided that sufficient sulphate and barium are available. The radioactivity of these deposits can be thousands of times higher than that in waste rocks.

TABLE XIII. MAXIMUM VALUES OF RADIOACTIVITY IN MINE WATERS AND SUSPENDED SEDIMENTS DISCHARGED TO THE ENVIRONMENT
(data from Refs [64, 65])

	^{226}Ra	^{228}Ra
Discharge water (Bq/L)	63	28
Underground sediments (kBq/kg)	400	—
Sediments at discharge point (kBq/kg)	55	15

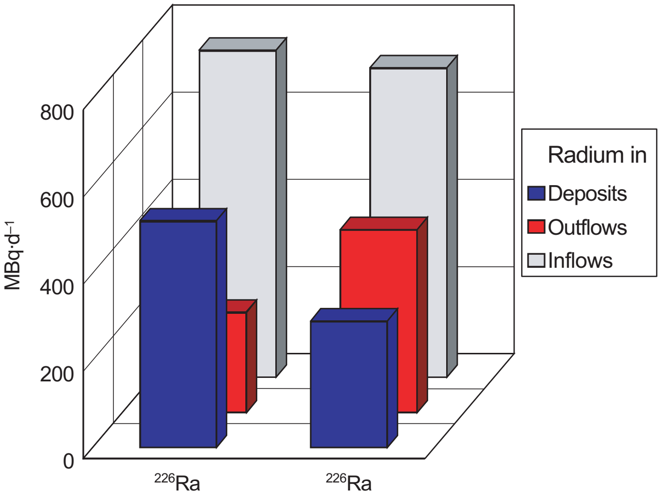

FIG. 13. Balance of radium in Polish coal mine waters [66].

The mining process itself, however, may lead to the dispersal of radionuclides from brines associated with either the coal or the host rock. Owing to the co-precipitation effects and dilution with other waters, the mine waters pumped from the mine to the surface are typically less radioactive (Fig. 13). Nevertheless high radium concentrations in the outflows were reported, for example, in Poland [51, 65] and Germany [64] (Table XIII). Thus, accumulation of ^{226}Ra and ^{228}Ra was observed in settling ponds and river beds in Poland [64] and Germany [67].

4.2.4. Management of residues

Most of the residues from coal mining are deposited onto the land surface. Only a fraction of the waste rocks is used for backfilling and for other purposes. The radioactive sediments generated underground are emplaced mainly in mined-out voids. In mines, where radium co-precipitates in settling ponds, the sediments are mixed with material used for backfilling and transported back underground [66].

Given the extent of solid fossil fuel burning and the amount of residues produced by power plants [45], the amount of residues in countries with many coal power plants is very large. The absence or inadequacy of flue gas filtering and scrubbing, in particular at smaller and older units, may result in the atmospheric dispersal of fly ashes. The scrubbing of flue gases in many larger

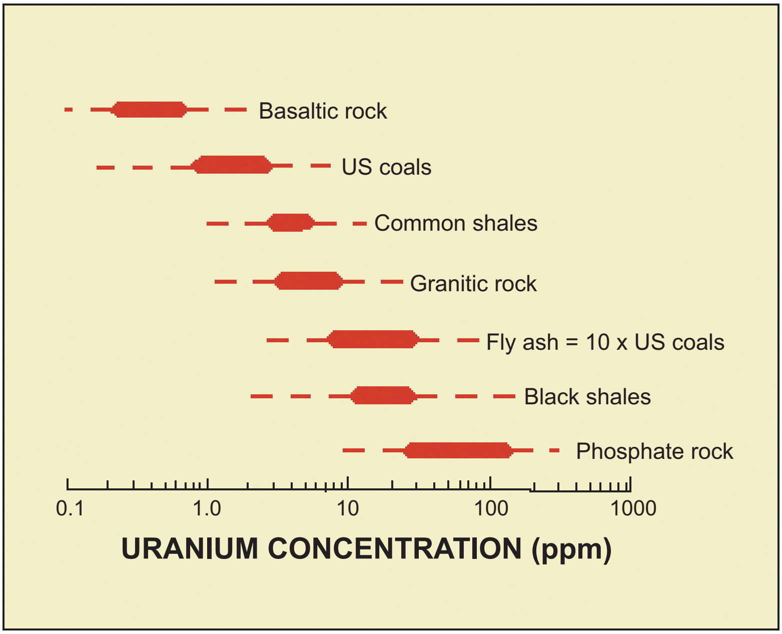

FIG. 14. Typical range of uranium concentrations in coal, fly ash, and a variety of common rocks [71].

coal power plants appears to be efficient enough that no significant contamination around the plants was found in Brazil [68], India [69] and Poland [70].

Other solid residues are either disposed of in engineered surface impoundments and landfills, or are backfilled into the mines. To a degree fly ash and gypsum are recycled as building materials, for instance, as additives to concrete or in lightweight building materials [20]. If used for the construction of houses, this may lead to exposure of the inhabitants to the radiation from the NORM contained in this material. While use of fly ash in concrete blocks for building construction may not be of concern in some Member States, others regulate the levels of permissible radiation in such concrete blocks. Resuspension of fly ash and other flue gas scrubbing residues from open disposal pits could be relevant pathways for exposure.

While coal burning power plants are mostly now fitted with flue gas scrubbing and ash management systems, the situation for domestic coal burning is quite different. The particulate emissions from coal based domestic heating systems and the potential for exposure can be considerable, in particular in less developed countries [72].

However, as the vast majority of coal and the majority of fly ash are not significantly enriched in radioactive elements (Fig. 14), or in associated radioactivity, compared with common soils or rocks [71], there does not seem to be any great reason for investigating abatement technologies.

5. WATER

5.1. GEOTHERMAL ENERGY PRODUCTION

5.1.1. Introduction

Geothermal energy is heat produced and stored in the earth. This energy can be economically extracted from high temperature crustal rocks, sediments, volcanic deposits, water, steam and other gases found at accessible depths from the earth's surface. The heat carrier is typically either groundwater or water injected into the system with direct contact to, and hence reacting with, the host rock. The three technology categories used to do this are geothermal heat pumps, direct use applications and power plants. Direct use applications and power plants utilize groundwater which either occurs naturally in an underground geothermal reservoir, heated by surrounding rocks, or that is injected into the underground geothermal reservoir. World locations where geothermal energy can be utilized are classified into three principal systems:

(1) *Hot igneous systems* — created by the buoyant rise of molten rock (magma) from deep in the crust. In hot igneous systems, the rock is either completely or partly molten, with temperatures greater than 650°C.
(2) *Hot dry rock systems* — heated low permeability rocks that may or may not have been molten at one time, with temperatures less than 650°C.
(3) *Hydrothermal systems* — consist of high temperature water and/or steam trapped in porous and permeable rock reservoirs usually found in porous sedimentary rock or in fractured rock systems, such as volcanic formations. Hydrothermal systems are vapour dominated systems, which contain mostly steam (with temperatures ranging from 180 to 270°C), and liquid dominated systems (with temperatures ranging from 30 to 350°C).

The first two categories contain the most heat energy, but the technology does not yet exist to exploit them. Research is currently being conducted to remove the technological barriers that prevent the development of these resources. Geothermal energy, though not yet a significant source of total world energy, is used in a wide number of countries in the Americas, Europe, Asia and Africa. Electricity generation in the calendar year 2000 from 22 countries was reported as 7974 MW [73]. Individual producing sites, using wells to extract the underground heat or fluids, can range from a single well to as many as hundreds in the geothermal fields of the USA.

The radioisotopes associated with geothermal energy are principally radon and radium. There are no available measurements of these radionuclides, which are found unconcentrated in the original ground fluids.

5.1.2. Processes

The utilization of geothermal energy requires boreholes to be drilled for the withdrawal of high temperature fluids from the ground, surface utilization equipment (e.g. steam turbines or heat exchangers and associated fluid handling equipment) and a fluid disposal system (e.g. percolation ponds or reinjection wells). The following discussion on the power plant process is modified from Ref. [74]:

(a) *Dry steam power plants* (Fig. 15). Steam plants use hydrothermal fluids, primarily steam. The steam goes directly to a turbine, which drives a generator that produces electricity. Water in the form of steam enters the power plant through underground extraction wells, and the steam which turns the turbine, thus losing its heat energy, is returned to the

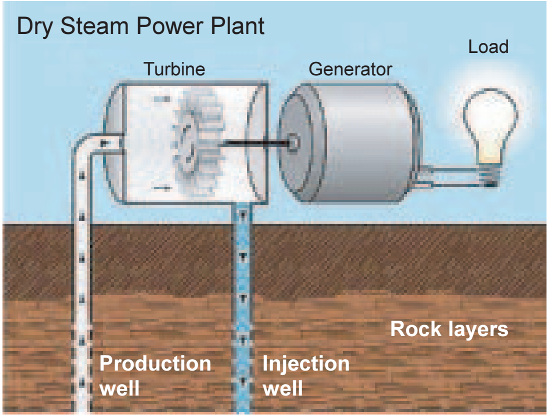

FIG. 15. Dry steam power plant [74].

underground reservoir by injection wells. These plants emit only excess steam and very minor amounts of gas.

(b) *Flash steam power plants* (Fig. 16). Hydrothermal fluids above 200ºC extracted from underground reservoirs by extraction wells can be used in flash plants to generate electricity. Fluid is sprayed into a tank held at a much lower pressure than the fluid, causing some of the fluid to rapidly vaporize, or 'flash', to steam. The steam then drives a turbine, which drives a generator. If any liquid remains in the tank, it can be flashed again in a second tank to extract even more energy. Only excess steam and trace gases are emitted. The fluids now cooled on the surface are reinjected by wells into the underground reservoir.

(c) *Binary cycle power plants* (Fig. 17). Most geothermal areas contain moderate temperature water (below 200ºC). Energy is extracted from these fluids in binary cycle power plants. Hot geothermal fluid derived from an underground production well and a secondary man-made (hence, 'binary') fluid with a much lower boiling point than water pass through a heat exchanger. Heat from the geothermal fluid causes the secondary fluid to flash to steam, which then drives the turbines. Because this is a closed loop system, virtually nothing is emitted to the atmosphere. Moderate temperature water is by far the most common geothermal resource, and most geothermal power plants in the future will be binary cycle plants. 'Spent' geothermal fluids are returned to the underground reservoir by an injection well.

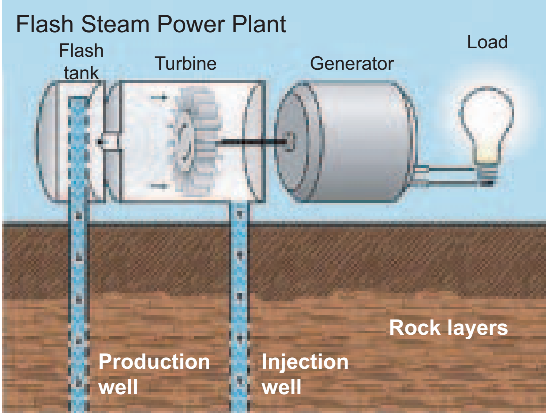

FIG. 16. Flash steam power plant [74].

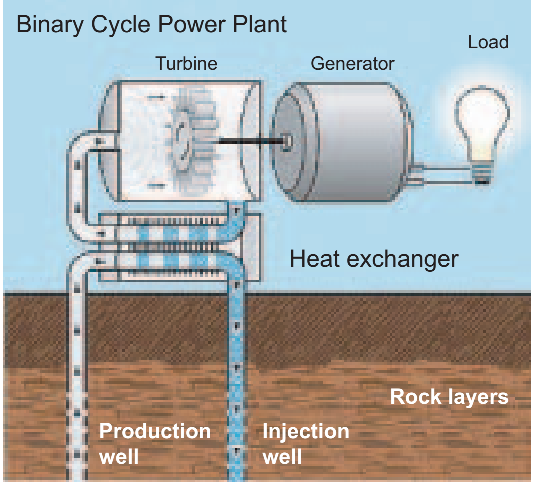

FIG. 17. Binary cycle power plant [74].

On the other hand, geothermal reservoirs of low to moderate temperature at 20–150ºC provide direct heat for residential, industrial and commercial uses. This resource is widespread around the world, and is used to heat homes and offices, commercial greenhouses, fish farms, food processing facilities, mining operations, and a variety of other applications. A recent survey of ten western US states identified more than 9000 thermal wells and springs, more than 900 low to moderate temperature geothermal resource areas, and hundreds of direct use sites. Direct use systems are typically composed of three components:

(1) A production facility — usually a well — to bring the hot water to the surface;
(2) A mechanical system — piping, heat exchanger and controls — to deliver the heat to the space or process;
(3) A disposal system — injection well or storage pond — to receive the cooled geothermal fluid.

5.1.3. Residues

NORM is found in the solid residues generated by the exploration and development of geothermal systems and in the extraction of the earth's geothermal energy for use in either producing electric power or supplying direct heat. Changes in the thermodynamic properties, such as pressure, temperature and volume, can cause precipitation and gas evolution because of the varying solubilities of some of the constituents present in fluids. The resulting fluids have different physicochemical properties than the original, and it is these properties that are passed on to subsequent processing stages. Scale can form by precipitation within equipment used for extraction and distribution to power systems. Sludge is formed by precipitation and sedimentation during the cooling process. Gas evolution can occur at any stage under appropriate conditions. These NORM containing residues include minerals that precipitate out of solution and form scale or sludge on the inside surfaces of the drilling and production equipment (e.g. steam turbines, heat exchangers, process lines, valves, turbines and fluid handling equipment) used to extract geothermal heat. Such residues contain barium, calcium and strontium salts (carbonates, sulphates and silicates), and silica, as well as significant concentrations of radium and radium decay products. Radium is slightly soluble and, consequently, can be brought to the surface and co-precipitated with barium and calcium salts onto the inside surfaces of drilling and production equipment, similar to the case of oil and gas production. The principal residues of concern are the scales in pipework and production equipment and the filter cake produced from treatment of spent geothermal fluids prior to their reinjection. Scales are hard, insoluble sulphate deposits that form on the inside of pipes, tubulars, filters, pumps, well heads and other water handling equipment.

Concentrations of TE-NORM in geothermal residues will vary with the geology and mineralogy of a geothermal resource area along with the physical and chemical changes that occur during energy extraction. As the TE-NORM is contained in a mineral matrix, this may lessen the radon emanation rates and leachability; however, few data are available on these particular scales.

Drilling operations for the production and injection wells require the use of special drilling mud and produce cuttings mixed with drilling mud. During the extraction process, geothermal fluids containing dissolved and suspended constituents are brought to the surface. Solids, such as rock cuttings, are removed from used drilling muds by circulation through special equipment, such as shale shakers, sand traps, hydrocyclones and centrifuges. After cleaning, the mud is recycled to the drilling operation and the removed solids are disposed of as waste. When drilling is completed, the used muds are discharged to reserve pits for storage or disposal. Although the solid waste generated during the drilling phase can be significant, precipitation and sedimentation from the geothermal fluids are the primary contributors to solid waste formation during energy extraction.

The only available estimate for annual waste generation is about 45 500 t for Imperial Valley, California, based on 1991 data [75]. The total can be subdivided into five waste streams, including: filter cake, 18 318 t; sulphur, 16 240 t; drilling, 9571 t; scale, 946 t; miscellaneous, 113 t. For filter cake and scale, these amounts translate to generation rates of 87 t per MW(net) and 0.58 t per MW(net), respectively. Power plants operating in Imperial County are estimated to produce approximately 19 200 t of filter cake and scale waste annually. On the basis of the average density of 1.3 $g\cdot cm^{-3}$, obtained from the final environmental impact statement for a disposal site receiving waste from liquid dominated geothermal facilities in Imperial Valley [76], this is equivalent to a waste volume of about 14 800 m^3. There are indications that up to an additional 40 000 t of silica scale may be produced annually from Imperial Valley [75]. This potential additional waste represents a source of significant uncertainty.

Similar to the case when oil and gas are produced, uranium and thorium and their radioactive decay products may be present in formations from which geothermal fluids are extracted. However, the available information indicates that no significant quantities of uranium and thorium are dissolved or entrained in geothermal fluids [75]. The primary radionuclides present in geothermal fluids are ^{224}Ra, ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{212}Pb and ^{222}Rn from the ^{238}U and ^{232}Th decay chains [77]. In addition, ^{40}K is often the only detectable radionuclide in water produced from Californian formations exploited for both geothermal energy and oil and gas [75], which suggests that ^{40}K may also be a significant contributor to NORM in geothermal waste, but there are no data to confirm this.

Very little information is available on the concentrations of TE-NORM in geothermal solid waste such as filter cakes, scale in piping and production equipment, and gases. Concentrations of NORM in geothermal residues will vary with the geology and mineralogy of the geothermal resource area along with the physical and chemical changes that occur during energy extraction. Table XIV (based on Ref. [58]) gives a breakdown of typical radionuclide concentrations found in residues from geothermal energy production.

An environmental impact report on four geothermal power plants located in the Imperial Valley of California [78] provides the results of analyses of filter cake and clarifier sediment samples. The data are summarized in Table XV.

Data for ^{222}Rn indicate that an emission rate of 1 $mBq\cdot d^{-1}$ per megawatt of power occurs at the Geysers field [76] in northern California. Radon flux rates were estimated to be 5.92 $Bq/(m^2\cdot s)$ from uncovered waste disposed of in a regulated landfill. The radon emanation coefficient was estimated to be 0.25. Radon concentrations in dry steam and ejector gas have been reported to vary widely, ranging from about 0.37 Bq/L to as low as 0.74 nBq/L. Average radon concentrations were tabulated and compared for different geothermal systems [79]. The results (Table XVI) indicate that concentrations are significantly

TABLE XIV. RADIONUCLIDE CONCENTRATIONS IN GEOTHERMAL ENERGY PRODUCTION WASTE [58]

Nuclide	Residue concentration (Bq/kg)
^{228}Th	930
^{228}Ra	3440
^{226}Ra	4880
^{210}Po	3550
^{210}Pb	3550
Total*	16 350

* Excludes contributions from other (short lived) radionuclides in decay chains.

higher in dry steam fields. In addition, wide variations were noted, even within individual geothermal fields.

5.1.4. Management of residues

Information on disposal of brines, sludges and other solid waste from these fields is not generally available, though the annual geothermal solid waste production rate in the USA is estimated to be between about 20 000 and 50 000 t. Over half of that is managed at a single disposal landfill facility, which

TABLE XV. RADIONUCLIDE CONCENTRATION RANGES IN GEOTHERMAL ENERGY PRODUCTION WASTE FROM GEOTHERMAL POWER PLANTS IN IMPERIAL VALLEY, CALIFORNIA, USA [78]

Nuclide	Residue concentration range (Bq/kg)
^{228}Th	Below detection level to 1628
^{228}Ra	Below detection level to 6771
^{226}Ra	370–9398
^{212}Po	Below detection level to 999
^{212}Pb	Below detection level to 1554
^{214}Pb	0.037–7622
^{214}Bi	0.037–6401

TABLE XVI. AVERAGE RADON CONCENTRATIONS BY TYPE OF GEOTHERMAL SYSTEM [79]

Type of geothermal system	Location	Average radon concentration (10^{-9} Bq/L)*
Dry steam	Larderello, Italy	1.28
Dry steam	The Geysers, USA	0.62
Flashing brine	Salton Sea KGRA, USA	0.11
Hot water	East Mesa, USA	0.01

* Concentrations given in condensed geothermal fluids. In steam fields, the concentration is inversely proportional to the corresponding volume of steam at the well head temperature and pressure.

receives waste from power plants located in Imperial County, California. The amount of TE-NORM waste generated annually is assumed to be equivalent to the estimate characterizing the production of filter cake and scale. Data on other world production facilities are not currently available.

Advanced biochemical processes have been investigated and are being developed commercially for geothermal brines and sludge [79, 80]. The purpose of these processes is to take advantage of certain biochemical agents that can extract trace metals and valuable salts, produce pigment free silica, and remove radioactivity to levels within exemptions. These biochemical processes may yield significant benefits to the geothermal industry by reducing the volumes of specific waste streams, producing cleaner waste fractions, generating secondary markets for processed waste, and reducing processing costs.

5.1.5. Environmental impacts

Environmental impacts from the generation of TE-NORM in the geothermal industry will continue to grow around the world as this resource is exploited. Radon gas emissions from the power plant stacks and facilities, as well as solid waste materials, provide the potential for human and environmental exposures. Most produced waters are re-injected into the geothermal source formation, so this should not represent a significant problem. Within the power plant, similar to the oil and gas industry, scales can accumulate in tubular piping, separators and steam containment vessels. The handling of the mineral scales can constitute environmental problems at sites where the scales are removed and stored, primarily through dust emissions and direct gamma radiation. As the TE-NORM is contained in a mineral matrix, this may lessen the radon emanation rates and leachability; however, few data are available on these particular scales. Disposal into landfill facilities has the potential for long term radionuclide transport through groundwaters should a site liner fail.

Sludges containing TE-NORM from geothermal steam plants may be stored on-site in pits or barrels. In addition to inhalation and direct exposure, any brines or sludges stored in ponds have the potential for site leakage into the ground, and potential for contamination of groundwater. Any sludges disposed in landfill facilities have the potential for more rapid leakage into surrounding land and groundwater should the landfill liner fail.

Potential misuse of the solid waste outside the boundaries of production plants could result in increased public and environmental exposures. Historical information indicates that silica scale produced in one US field has been used for roads, dykes and levees [81]. Mixing of the silica scale with asphalt and cement for road surfacing and bricks has also been reported [82]. In other applications, small amounts of silica scale have been used to construct artistic objects as part of the Community Geothermal Technology Program in Hawaii [83].

5.2. DRINKING WATER TREATMENT

5.2.1. Introduction

Public water supply systems derive their water from surface water bodies such as rivers, streams and reservoirs, or underground sources through drilled wells. Vast amounts of drinking water are obtained underground each year. For example, the fraction of drinking water obtained from groundwater in Germany, France, Italy, Spain and the UK was 65, 60, 50, 20 and 25%, respectively, in 1995 [84]. 5800 Mm^3 of water were delivered for 80 million inhabitants in Germany in 1995. About 78% of this water required treatment. In the USA, 1500 Mm^3 are produced annually, of which 50% is groundwater, nearly all requiring treatment for removal of impurities.

In most developed countries (public) water suppliers are required to limit the occurrence of contaminants such as metals and pesticides, and sometimes also radiation in drinking water. In the USA limits have been set for radium, uranium, alpha, beta and photon emitters in drinking water, and a draft standard has also been developed for limiting radon in those supplies [85]. In the European Union the need to, and the technical possibilities to, treat raw water were investigated in a recent project [86]. Water treatment plants occur in a variety of different sizes based on the population served. In Ref. [86] a summary is provided of the different classifications of waterworks in Germany and the populations served.

Surface water discharges by industries with NORM in their wastewater is suspected, but information on this is generally unavailable.

Groundwater is found almost everywhere on the earth. Albeit, the depth to the groundwater and the quality of different groundwaters vary greatly. Likewise, the concentrations of radionuclides in groundwater will vary depending on the local geology. For example, higher concentrations can be found in granite and uranium bearing formations. Naturally occurring radionuclides are leached into waters when they come in contact with uranium and thorium bearing geological media. Hence, the predominant radionuclides found in water include those of uranium, radium, radon and their decay products [87]. In general, the radionuclides of concern in groundwater are ^{226}Ra and its progeny (including ^{222}Rn). Concentrations of ^{226}Ra are known to vary in the USA from 4×10^{-4} to 4×10^{-2} Bq/L in surface water and from 0.02 to 0.93 Bq/L in groundwater [88–91]. Reports of ^{226}Ra concentrations as high as 7.4 Bq/L have been documented, but in most instances concentrations rarely exceed 1.85 Bq/L [88]. See also Ref. [92] for more references on radioactivity levels in water. The ^{228}Ra to ^{226}Ra concentration ratio has been reported to vary from 0.2:1 to 5:1; however, a ratio of 1.2:1 is most likely representative of average conditions in water extracted for human or agricultural use [87, 93]. The higher ^{228}Ra concentration is mostly due to the average crustal ^{232}Th and ^{238}U activity ratio of about 1.2:1 to 1.5:1. Natural geochemical enrichment or depletion processes may either increase or decrease this ratio. When present in groundwater, ^{228}Ra cannot be used to deduct the concentration of ^{226}Ra.

Although radium tends to be more soluble in typical water compositions, uranium and some ^{232}Th progeny may be found as well, and uranium activity levels as high as 24.1 Bq/L have been observed in both surface and groundwater samples [93, 94]. Concentrations of total uranium, ^{238}U plus ^{234}U, as high as 1.48 Bq/L have been described in groundwater samples from the Newark Basin in New Jersey [91]. Uranium concentrations in US surface and groundwater average about 0.037 and 0.11 Bq/L, respectively [94]. The isotopic ratio of ^{234}U to ^{238}U is known to vary, with higher concentrations of ^{234}U in both surface and groundwater. Ratios up to 28 have been reported, but more commonly a range of 1–3 has been found [88]. Higher ^{234}U concentrations are due to the alpha recoil process, which enhances the mobilization and solubility of the decay product (^{234}U) relative to the parent (^{238}U). After the alpha particle has been emitted, the uranium is often stabilized in solution as a very mobile uranyl carbonate complex.

The isotopes ^{230}Th and ^{234}Th are usually not detected in groundwater samples and are estimated to have upper mean concentrations of only 0.001 and 4×10^{-4} Bq/L, respectively [95]. Similarly, the presence of ^{232}Th in groundwater is believed to be very low, about 4×10^{-4}, and is rarely above

0.04 Bq/L, when detected [94]. The decay products of ^{222}Rn, the relatively long lived ^{210}Pb and ^{210}Po, are also generally found at very low concentrations in groundwater, 7×10^{-4} to 4.4×10^{-3} Bq/L [88, 95].

The concentration of radon in groundwater is known to vary significantly. In the USA again, for 2457 groundwater samples analysed for ^{222}Rn, statewide average concentrations ranged from 29.6 Bq/L in Mississippi to 513.6 Bq/L in New Hampshire [96]. Radon concentrations have been reported to be as high as 588.3 Bq/L in groundwater samples taken from the Newark Basin in New Jersey [94].

Although radionuclides are found in surface water, it has been found that groundwater is more likely to contain higher levels of this material due to the difference in chemical compositions. Some waters may require treatment to reduce concentrations to levels accepted by the regulators concerned. The principal problem with water treatment is that the very processes that are designed to remove radionuclides from the finished product — drinking or process water — concentrate those radionuclides in filters and sludges, and contaminate filtration equipment. The now contaminated equipment may require treatment and cleaning, and the concentrated waste, residual sludges, filters and other residues appropriate disposal.

Even in cases where radionuclide concentrations are significant, but not above standards, the customary water treatment, for example to remove hardness, can result in accumulation of NORM in process residues.

5.2.2. Processes

There are many different processes used for drinking water treatment. Some of these processes are specifically designed to remove NORM, such as ^{226}Ra. Many of these processes are designed to remove other contaminants and often also remove NORM at the same time. In general, the NORM specific processes will lead to wastes with higher NORM activity concentrations. In some cases, these concentrations can be substantial [97].

Radium is the most common target for removal by cation exchange resins and by a lime softening process [98]. A report prepared for the United Sates Environmental Protection Agency (USEPA) [97] describes several processes used to remove radionuclides from drinking water. The processes and target radionuclides are cation exchange (radium), greensand filtration (radium), co-precipitation with barium sulphate (radium), hydrous magnesium oxides (radium), anion exchange (uranium), coagulation/filtration (uranium), lime softening (radium and uranium), reverse osmosis (radium and uranium), granular activated carbon (GAC) (radon) and aeration (radon). A report prepared for the European Commission describes a general procedure for water treatment [97] which is not specifically directed at removing NORM. The process comprises four steps: aeration, removal of iron and manganization, pH adjustment and filtration. Figure 18 provides an overview of how this process is applied in Germany and the fate of NORM at each step of the process.

5.2.3. Residues

Examples of NORM containing residues from municipal water treatment are radioactive contaminated sludges and solids including filter sludges, spent ion exchange resins, spent granular activated carbon, as well as waters from filter backwash. Radium-selective ion exchange resins result in residues with much higher activity concentrations than those found in sludge, but in smaller

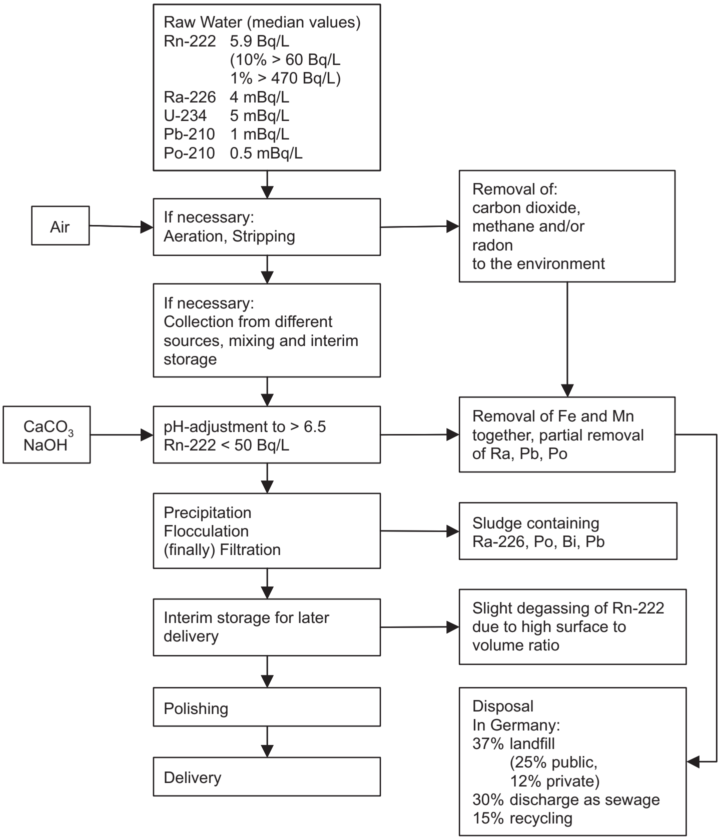

FIG. 18. Typical sequence of drinking water treatment [97].

quantities. In general, the more efficient and more specific the method of cation removal, the higher the specific activities are likely to be, resulting in a significant waste management problem. The concentrations can vary considerably from location to location because of the varying geological characteristics of different water sources in different regions of a country.

Water treatment sludges are placed in lagoons and may include lime sludge, backflush water, spent ion exchange media and sand filter elements. Disposal of sludges in lagoons results in the accumulation of radium in the bottom sediments, which may have to be dredged and disposed of properly. NORM containing sludge is also disposed of in sanitary landfills, discharged to sewers, injected in deep wells or spread on agricultural soils, while the decanted water is recycled. Some of these practices, however, are now forbidden in certain Member States due to the resulting contamination with heavy metals.

Given the amounts of water that are treated in a typical water treatment plant, the amounts of residues can be substantial. Surveys conducted by the American Water Works Association (AWWA), which conducts periodic surveys of the largest US water utilities [99–101], provided information on the volumes of waste generated through water treatment in that country. On the basis of survey data, the USEPA has estimated that, with some uncertainty, as much as 260 000 t of NORM contaminated residues are produced annually. That number may increase incrementally because new regulations now require additional treatment for uranium. For example, for the whole of the USA, it was estimated that roughly one million tonnes of NORM residues would be produced over a 20 year period from drinking water treatment, i.e. roughly 50 000 t/a [102]. Table XVII provides some examples of the amounts and radionuclide contents of NORM waste from different processes used in the USA.

In Germany, it is estimated that roughly 125 000 t of different types of sludge arise from surveyed waterworks each year for a throughput of 3600 Mm^3 of drinking water [67]. For the purposes of an assessment, it was assumed that roughly 800 t of dry sludge would result from delivery of about 20 Mm^3 of drinking water on an annual basis (a large waterworks). Medium and small waterworks were estimated to generate roughly 200 and 70 t of sludge, respectively, on an annual basis. Typical data for the concentrations of radionuclides for German water treatment are provided in Table XVIII.

5.2.4. Management of residues

The USEPA has addressed disposal management options, based on existing practices, for waste generated by water treatment and purification systems [103].

TABLE XVII. CHARACTERISTICS OF SELECTED WATER TREATMENT WASTES (COMPUTED LEVELS) [86]

(waste amounts based on an average throughput of 1 million gallons (≈380 000 L) of water per day)

Treatment method	NORM concentration in water (Bq/L)		Waste volume (kg/d)	Approximate radioactivity (Bq/L)
	Raw	Finished		
Coagulation/filtration (U)	1.85	0.37	10[a]	148
Lime softening (Ra)	0.93	0.46	22[a]	20.6
Ion exchange (Ra)[b]	0.93	0.19	97[c]	7.7
Reverse osmosis (U)	1.85	0.56	130[c]	11.8

[a] Sludges.

[b] Does not include ion exchange resins, which have much higher radioactivity levels.

[c] Waste.

The USEPA [104] indicates that NORM residues from drinking water treatment may be disposed of in landfills or lagoons or can also be used as agricultural conditioners. In the case of lagoons, evaporation is used as the means to reduce the volume of the waste. However, this results in a lower volume of residues with a higher activity concentration of radionuclides. In

TABLE XVIII. TYPICAL DATA FOR SLUDGE CONTAINING NATURAL RADIONUCLIDES PRODUCED BY A LARGE WATERWORKS IN GERMANY [97]

Weekly throughput of water	420 000 m^3
Total activity of this water assuming very conservative estimate of concentration	3 654 000 000 Bq
Total activity taking maximum measured radionuclide content in Germany	588 000 000 Bq
Total activity using median value for tap water in Germany	8 400 000 Bq
Produced sludge per week	15 t
Dry solids in sludge	6 t
Specific activity of dried sludge for different water concentrations:	
— High conservative concentration	600 000 Bq/kg
— Maximum measured concentration	100 000 Bq/kg
— Median concentration for tap water	1 400 Bq/kg

cases where the concentrations of radionuclides are higher, it may be necessary to dispose of the residues in a licensed radioactive waste disposal facility. Given that NORM is long lived, it may be a problem to dispose of these wastes in a near surface repository [30]. Table XIX summarizes potential disposal practices for NORM containing residues resulting from drinking water treatment in the USA. In Germany, it is estimated that 37% of sludges are disposed of in landfills, 30% are treated with sewage and roughly 15% are recycled [97].

5.2.5. Environmental impacts

The vast majority of residues from water treatment are in the form of sludges. However, ion exchange resins and filter media can be a significant radiological concern given that very high concentrations of radium are possible in those residues. From a groundwater protection perspective, disposal of sludges is likely to be the greatest concern, because the radionuclides on resins or filter media would not be expected to be as mobile. The use of lagoons and evaporation ponds to dispose of residues may result in contamination of underlying groundwater bodies. This then may require long term stewardship or other measures to prevent future human and environmental exposures due to migration, or due to re-use of the water or residues. Disposal of TE-NORM into sanitary sewer systems can potentially result in accumulations of those radionuclides in sewage sludge. Depending on the concentration levels found in the sludge, special considerations may need to be given to the re-use or disposal of this waste. External exposure and potential radon exposures are

TABLE XIX. DISPOSAL PRACTICES FOR NORM FROM WATER TREATMENT FACILITIES IN THE USA [86]

Liquid waste disposal	Solids/sludge disposal
Direct discharge to surface water	Temporary lagooning (surface impoundment)
Deep well injection	Disposal in landfill
Drying or chemical precipitation	— without prior treatment (resins, filter media, granular activated carbon)
Irrigation	— after temporary lagooning
Sanitary sewer discharge	— after mechanical dewatering
	Application to land (soil spreading/conditioning)
	Disposal at licensed low level waste facility

likely to be of more concern for resins and filter media, because the concentrations of NORM would not be expected to decrease substantially over time.

5.3. WASTEWATER TREATMENT

It was noted during screening exercises that sewage sludges and ashes from incinerated sludges sometimes have elevated levels of radionuclides [105]. The sources are either elevated levels of NORM in groundwater or (authorized) discharges into the sewerage system by various industries. The organic solids in the sewage concentrate radionuclides, and the incineration of the sludge further concentrates them.

The common practice of dispersing sewage sludges or incinerator ashes onto agricultural land can lead to the accumulation of radionuclides and, in particular, heavy metals, in the soils and eventually their uptake into crops. For this reason various countries have regulated this practice over the last few years or are in the process of doing so; see, for example, Ref. [106].

5.4. INDUSTRIAL PROCESS WATERS

Several industries, for instance the paper industry, are heavy users of process water. Owing to the substances involved, significant radionuclide concentrations can accumulate in various products, intermediates and wastes. Paper pulp, for instance, is capable of scavenging heavy metals and radionuclides. Certain chemical processes may lead to the precipitation of scales inside tubes and other vessels, which have to be removed and require special treatment when radioactively contaminated.

For instance, in December 1999, the radiation detectors at a steel recycling yard in eastern Ontario found scrap piping from a pulp and paper mill to be radioactive [107]. The piping was returned to the mill, where the owners started an investigation and found that the pipes had been removed from the pulp bleaching process some years previously. The field on-contact with the scale inside the pipe was 200 μR/h. A scale sample contained about 15 kBq/kg of ^{226}Ra and about 4 kBq/kg of ^{230}Th.

5.5. IRRIGATION

As with drinking water, (ground)waters used for irrigation may contain NORM that may accumulate in certain soil fractions and eventually transfer into plant materials.

For instance, in irrigated areas along the Arkansas river valley in southeastern Colorado, uranium and salts are actively leached from marine shales (Fig. 19). These contaminated saline irrigation waters eventually return to the river, where uranium levels increase to concentrations as high as 200 ppb and, because of the accompanying high salinity, wetlands in this area do not trap uranium. In other much publicized wetland areas such as the Kesterson Wildlife Refuge in California, uranium and selenium contamination is responsible for wildlife death and deformities [108]. Many other irrigation systems in semi-arid areas that drain farmland on marine shales face similar stresses on water quality.

6. METALS

6.1. MINING AND PROCESSING METHODS

6.1.1. Open pit mining

Open pit mining techniques are employed to exploit ore deposits relatively close to the surface of the earth. Topsoil is typically removed

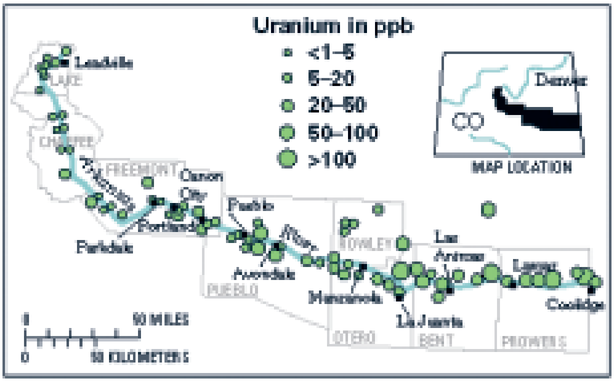

FIG. 19. Uranium in surface waters of the Arkansas river valley, southeastern Colorado, April 1991 (from Ref. [108]).

separately and stockpiled. Overburden, the material overlying the deposit, is removed using scrapers, or with trucks and loaders or mechanical shovels. Depending on the extent of consolidation, the overburden may be broken up by bulldozers or blasted prior to removal. Overburden may be stockpiled outside the pit or placed in mined-out portions of the pit once pit development has progressed to an acceptable point. Mining economics typically require that overburden haulage be minimized. Once the ore body has been exposed, it is ripped, loaded into trucks and transported to an on-site stockpile. The ore can then be moved from the stockpile to the mill site as required.

The depth to which an ore body is mined depends on the ore grade, nature of the overburden and stripping ratio. Stripping ratios describe the amount of overburden that must be removed to extract one unit of ore. One report indicates that stripping ratios for open pit uranium mines range from 10:1 to 80:1 with an average of around 30:1 [109]. The primary advantage of surface mining is the ability to move large amounts of material at a relatively low cost, in comparison with underground operations.

Waste material, including overburden, is often returned to mined-out areas during mining to reduce haulage costs. Ore, protore and low grade mineralized rock are outlined, and plans developed for mining and stockpiling these materials. Often parts of an ore body delineated by drilling cannot be economically mined by open pit methods. Where parts of the deposit lie adjacent to the bottom of the planned pit, underground mines may be developed from the pit bottom to recover these ores.

6.1.2. Underground mining

A variety of techniques are employed in underground operations depending on the distribution and orientation of the ore deposit. In general, underground mining involves sinking a shaft (or driving an adit) near the ore body to be mined and extending levels from the main shaft at various depths to the ore. Shafts, adits, drifts and cross-cuts are developed to access and remove the ore body. Ore and development rock, the non-ore bearing material generated during mining, may be removed either through shaft conveyances or chutes, and hoisted in skips (elevators) to the surface or used to backfill mined-out areas. Ore is placed in stockpiles while development rock brought to the surface is placed in waste rock dumps. As underground mining techniques are able to leave much of the non-ore bearing material in place, the ratio of waste (development) rock to ore is much lower than stripping ratios in open pit mines. Ratios of waste rock to ore range from 1:1.5 to 1:16 [109]. As with surface mining operations, ores and subgrade ores may be stockpiled on the surface. These materials may be beneficiated as market conditions allow or left with mine development rock in the waste rock piles.

Because of the high costs associated with moving rock underground, a minimum of waste rock is generated, some weakly mineralized rock is generated, and a high proportion of subore and ore is mined. Because of the high costs of moving such materials, some waste rock may be used underground as backfill material in mined-out areas [109]. The extracted ore is stockpiled at the surface or transported by truck directly to the mill, protore may be stockpiled on-site for later shipment if economic conditions warrant this, while the waste rock is placed in different areas.

6.1.3. Solution mining

Solution mining is a general term used in the uranium and some other mining industries to describe operations in which a leach solution, referred to as the lixiviant, is employed to extract the target metal from subsurface ore deposits. The chemical reactions involved during in situ leaching are the same as those involved in the chemical aspects of a metal ore mill. A number of solution mining techniques have been explored since the 1960s, including in situ leaching, stope leaching and borehole mining. Stope leaching involves the injection of lixiviant into areas that have previously been hydraulically isolated and backfilled with low grade ore or that contain low grade ores to begin with. The lixiviant can be recovered from a sump or well drilled to the bottom of the mine workings. Like other solution mining techniques, this method allows the recovery of metal from ores not economically mineable using conventional methods.

During in situ mining, barren lixiviant is pumped down injection wells into the ore body; production wells then bring the pregnant leach solution to the surface for further beneficiation. Numerous well patterns have been investigated since the early 1960s, when in situ mining techniques were first employed. They are intended to create a hydrodynamically well controlled flow regime for the lixiviant, thus minimizing dispersion losses and the contamination of surrounding aquifers. When in situ leaching is completed, the ore body and aquifer are placed in a restoration phase, as required under State regulatory programmes and mine permits. Typically, the aquifer must be restored to its prior classification for water use regarding the presence of metals, organics, pH level and radioactivity.

In some instances, it has been impossible to return the water quality to its original baseline conditions. The contaminants most often found in groundwater that exceed the drinking water contaminant limits include uranium, radium, selenium, molybdenum, manganese, nitrate, sulphate, arsenic and iron [110]. The target metal can be recovered during the early stages of the restoration process as the water from the production wells passes through the ion exchange system. Eventually, uranium recovery is abandoned while restoration continues. A rinse of multiple aquifer pore volumes is typically required to reach a satisfactory level of restoration. The number of pore volumes required depends on the ease with which the aquifer returns to baseline conditions and the permit requirements established in the State permits [111, 112]. Wastewater that is not recirculated in the wells is usually placed in an impoundment (usually with a liner to prevent groundwater seepage) and evaporated. This water and the evaporite or sludge at the pond bottom is considered mining waste and thus NORM in most countries, and treated as solid or hazardous waste depending on national or local governmental requirements. In some cases this material is removed for disposal, in other cases it is left on the site after being covered with a capping material.

6.1.4. Beneficiation

Beneficiation of ores and minerals means crushing; grinding; washing; dissolution; crystallization; filtration; sorting; sizing; drying; sintering; pelletizing; briquetting; calcining to remove water and/or carbon dioxide; roasting; autoclaving, and/or chlorination in preparation for leaching (except where the roasting (and/or autoclaving and/or chlorination)/leaching sequence produces a final or intermediate product that does not undergo further beneficiation or processing); gravity concentration; magnetic separation; electrostatic separation; flotation; ion exchange; solvent extraction; electrowinning; precipitation; amalgamation; and heap, dump, tank and in situ leaching. Beneficiation of conventionally mined ores involves crushing and grinding the extracted ores followed by a leaching circuit.

6.1.5. Mining and milling residues

The mining and processing of metal ores, other than uranium, may also generate large quantities of NORM wastes. These wastes include ore tailings and smelter slag, some of which contain elevated concentrations of uranium, thorium, radium and their decay products that were originally part of the process feed ore. The extraction process for some ores can yield a waste product (e.g. tailings or slag) that has a higher radionuclide concentration than that of the original ore. Tailings are the solid materials remaining after physical or chemical beneficiation (washing, flotation, grinding and drying) has removed the valuable metal constituents from the ore. Slag is the vitreous residue mass left from the smelting (blast furnace melting and conversion) of metal ore for extraction and purification. A USEPA study [58] describes NORM wastes from the mining and processing of three categories of metals: rare earth metals (see below), special application metals and metals produced in bulk quantities (i.e. large volumes) by industrial extraction processes. Special application metals are regarded as metals that have unique commercial and industrial uses, and include hafnium, tin, titanium and zirconium. Metals mined and processed in bulk for industrial applications include aluminium, copper, iron, lead, zinc, and precious metals such as gold and silver. The level of NORM found in metal ores depends more on the geological formation or region than on the particular mineral being mined. The mining technique and its selectivity can be an important variable that controls the NORM content in wastes. For instance, the NORM content of adjacent ore veins or other deposits might vary considerably and selecting the appropriate portion can reduce the NORM in the waste stream. Table XX, based on Ref. [58], gives a breakdown of typical radionuclide concentrations found in NORM wastes from each of the three categories of mined metals.

Research at South African gold mines has shown that up to 70% of the chemical contaminant load found in mine water discharges to the environment

TABLE XX. RADIONUCLIDE CONCENTRATIONS IN METAL MINING AND PROCESSING WASTES
(from Ref. [58])

Nuclide	Rare earth metals (Bq/g)	Special application metals (Bq/g)	Large volume industry metals (Bq/g)
^{238}U	33.3	1.59	0.37
^{235}U	1.67	0.08	0.02
^{234}U	33.3	1.59	0.37
^{231}Pa	1.67	0.08	0.02
^{232}Th	74	0.81	0.37
^{230}Th	33.3	1.59	0.37
^{228}Th	74	0.81	0.37
^{227}Ac	1.67	0.08	0.02
^{228}Ra	74	0.81	0.37
^{226}Ra	33.3	1.59	0.19
^{210}Po	23.3	1.11	0.13
^{210}Pb	23.3	1.11	0.13

Note: Original data where given in pCi/g.

originate from pyritic oxidation processes in the underground stopes [113]. The impact of ore remains during dewatering of some underground areas and flooding following mine closure may hence be significant and has been investigated in various regional studies in South Africa, especially with regard to the amount of salts discharged to river systems [114, 115]. Chemical (especially heavy metal) pollution, caused by the penetration of such water from abandoned mines into adjacent operating mining areas, is of particular concern. Radioactivity still needs to be integrated into many studies on chemical pollution problems.

The systematic analysis of certain industrial processes is complicated by their complex and interlinked materials streams. Typical examples are the heavy mineral, rare earth and ceramics industries that are discussed later on. The primary feed material in all cases may be placer deposits, the heavy mineral content of which is split into different processing streams which may become reunited further on as waste streams of one industry join the feed streams of another. A discussion based on individual radionuclides may also not be appropriate, as frequently several occur together.

In the following sections the material flows for selected processes, types of primary feed materials or particular end products, and the ensuing waste and contamination problems, are discussed in more detail. The intention is not to give a comprehensive geographical coverage, but to describe the various processes or product streams in detail, in order to raise the awareness for the potential occurrence of NORM.

6.2. MINE WATERS

Mine waters can be of particular environmental concern, even a long time after the mining operation has ceased. Water is one of the main pathways by which NORM may be dispersed from mining and milling operations into the environment. Mining, be it open pit or underground mining, almost always requires the dewatering of the zone to be mined. Many (sedimentary) rocks contain sulphidic minerals and many ores are either sulphidic or associated with such mineral parageneses. Upon exposure to ambient air these sulphides are oxidized — a process generating acid. The acid in turn dissolves other mineral constituents. Also, many metallic elements, including radionuclides, exhibit higher solubility at low pH values. These phenomena, subsumed under the terms acid mine drainage (AMD) or acid rock drainage (ARD), have been in the focus of scientific and technical attention for some time [116, 117]. AMD is a worldwide problem.

Elevated radionuclide concentrations have been reported frequently in AMD/ARD. For instance, ^{238}U and ^{226}Ra concentrations of 175 and 0.29 Bq/L, respectively, have been measured in the ARD of the uranium mining and milling facility of Poços de Caldas, Brazil [118]. Similar ^{238}U concentrations in AMD of a coal mine in Brazil are reported in Ref. [39], but saline waters from German hard coal mines have even higher radium concentrations, up to 63 Bq/L of ^{226}Ra and up to 28 Bq/L of ^{228}Ra [67].

In situ leaching processes may result in the development of contaminated water plumes that extend beyond the boundary of producing well fields. Examples of this have been reported in the copper and uranium mining industries [119, 120].

Several options were explored for conditioning and disposal of residues from the treatment of mine waters and leachates from tailings ponds at the former Wismut uranium mines in Germany [121]. Near surface disposal in waste rock piles was considered less favourable due to the lower specific activity of the waste rock. Either disposal underground in the mines, above the final groundwater level, or in tailings ponds are the chosen solutions.

6.3. ALUMINIUM

6.3.1. Introduction

Aluminium is mined on a large scale as bauxite (containing about 50% $Al_2O_3{\cdot}2H_2O$). Australia, Guinea and Brazil are the largest producers of bauxite ore in the world (Table XXI).

The uses for aluminium in modern society are countless: cans and foils, kitchen utensils, building cladding, structural engineering applications where a strong, light, easily machined material is needed. Although its electrical conductivity is only about 60% that of copper per area of cross-section, it is used in electrical transmission lines because of its lightness and price. Its various alloys are of vital importance in the construction of modern aircraft and rockets. Evaporated in a vacuum, it forms a highly reflective coating for both visible light and radiant heat. These coatings form a thin layer of protective oxide and do not deteriorate as do silver coatings. These coatings are used for telescope mirrors, decorative paper, packages and toys, and in many other applications. The oxide, alumina, occurs naturally as ruby, sapphire, corundum and emery, and is used in glass making and for refractories. Synthetic ruby and sapphire are used in the construction of lasers.

The high electricity consumption for the electrolytic refining step (Section 6.3.2) has caused environmental concern, and recycling is strongly

TABLE XXI. WORLD BAUXITE MINE PRODUCTION ESTIMATES (10^3 t) [122]

Country	1999	2000
Australia	48 400	49 000
Guinea	15 000	15 000
Brazil	12 900	13 200
Jamaica	11 700	11 200
China	8 500	8 000
India	6 200	6 200
Venezuela	4 190	4 400
Suriname	4 000	4 000
Russian Federation	3 750	3 900
Guyana	3 300	3 300
Other countries	9 200	8 900
World total	127 000	127 000

encouraged, in particular for 'disposable' or one way products, such as cans and foils.

The bauxite ore can contain significant concentrations of radioactivity as shown in Table XXII. This radioactivity can be due to either or both of ^{238}U and ^{232}Th, depending on the ore gangue mineralogy. Some of the contained radioactivity may be associated with trace quantities of minerals which commonly have radioactivity associated with them, such as ilmenite and monazite. Some disequilibrium of the radionuclides in bauxites has also been observed.

TABLE XXII. SOME REPORTED CONCENTRATION RANGES OF RADIOACTIVITY (Bq/kg) IN BAUXITE ORE AND RED MUD [123]

	Bauxite	Red mud
Uranium series radionuclides	10–9000	100–3000
Thorium series radionuclides	35–1400	100–3000
Actinium series radionuclides	120–130	—
^{40}K	10–600	10–100

6.3.2. Processes

Bauxite is washed, ground and dissolved in caustic soda (sodium hydroxide) at high pressure and temperature (Bayer process). The resulting liquor contains a solution of sodium aluminate and undissolved bauxite residues containing iron, silicon and titanium. These residues (so-called ‘red mud’) sink gradually to the bottom of the tank and are removed (Fig. 20). The clear sodium aluminate solution is pumped into a large tank, the precipitator. Fine particles of alumina are added to seed the precipitation of pure alumina particles as the liquor cools. The particles sink to the bottom of the tank, are removed, and are then passed through a rotary or fluidized calciner at 1100°C to drive off the chemically combined water. The result is a white powder, pure alumina (Al_2O_3). The basis for all modern primary aluminium smelting plants is the Hall–Héroult process, invented in 1886. Alumina is dissolved in an electrolytic bath of molten cryolite ($NaAlF_4$) within a large carbon or graphite lined steel ‘pot’. A very high current, typically 150 000 A, is passed through the electrolyte at low voltage. This electric current flows between a carbon anode, made of petroleum coke and pitch, and a cathode, formed by the thick carbon or graphite lining of the pot. Molten aluminium is deposited at the bottom of the pot and is siphoned off periodically, taken to a holding furnace, often but

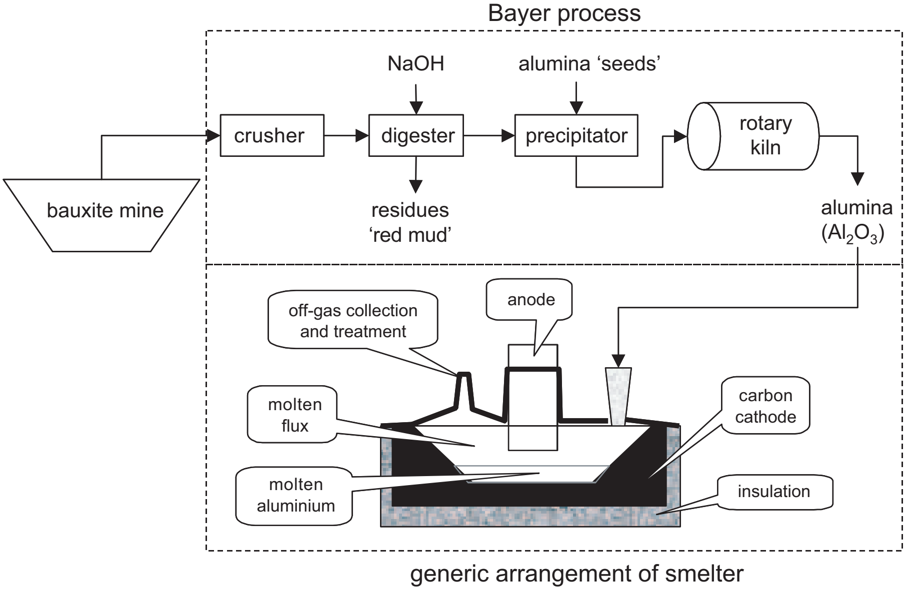

FIG. 20. Aluminium refining and smelting.

not always blended to an alloy specification, cleaned and then generally cast [124].

6.3.3. Residues

This residue, red mud, accumulates in large quantities on a worldwide scale: since the average alumina content in bauxite ore is 50%, the figures in Table XXI can be halved to give an estimate of the red mud arisings. The disposal of red mud presents a major problem, from both an ecological and an economic point of view [125]. Depending on the quality of the bauxite and the layout of the Bayer process, between 1.1 and 6.2 t of red mud are generated per tonne of alumina produced. Depending on the feed materials to the Bayer process (i.e. the bauxite ore and chemicals added), the resulting red mud is made up of a mixture of some original ore minerals and process determined precipitates, but consists of up to 60% hydrous ferric oxides, which have a high affinity for cations, including radionuclides and heavy metals. Reported concentrations of radioactivity in red mud are given in Table XXII [6, 8, 126, 127]. In some cases, depending on the bauxite ore grade, a threefold increase in the radionuclide content from ore to red mud is reported, and it is generally assumed that most of the radioactivity in the ore arises from the fine particle sized red muds. Uranium has been observed in alkaline leach solutions and may need to be given further consideration in regard to the potential for egress as seepage water to the environment and/or contamination of products and/or waste streams from sodium aluminate solution purification and alumina manufacture. There is no literature available which describes the nature of the radioactivity in the red mud. The siting of the radioactivity, and whether or not it is in equilibrium, is a subject for further study.

6.3.4. Management of residues

The major environmental issue in the industry is associated with the disposal of the red mud, which arises as an alkaline slurry. More than 70 million dry tonnes are produced each year [125]. The typical procedure is to dispose of the red mud onto waste heaps or in surface depressions. However, these red mud heaps can result in the percolation of caustic leachates into underlying aquifers. A more recent development has been to build engineered tailings ponds with clay and geotextile liners. These ponds, which may be 40–50 hectares in area, create other problems. Ponds were designed to hold several years production of mud, but they had poor settling characteristics and drained slowly only after they were full. Some have consequently been abandoned.

Without proper close-out procedures, this can result in airborne dust pollution once the surface layer has dried out.

Airborne dusts are a major potential pathway to exposure from the radioactivity in red mud. During operation the heaps are kept wet to minimize dust and after the heaps have been built the waste is covered with coarser residue and soil to reduce dust arising and radon/thoron emanation to meet regulatory requirements for the end use of the land.

Considerable attention has been given towards making further use of red mud. It has been used in the past, for example, in the manufacture of building materials [123], although this may lead to exposure under local conditions due to higher than average radionuclide levels. Red muds are also being used to improve the cohesive and water retention properties of sandy soils (soil conditioner) [127, 128], although there is some resistance to this [129], primarily because of the presence also of other potentially polluting constituents. There are also moves to use the mud for the treatment of acidic mine wastewaters [130, 131]. The potential exists for further useful applications of red mud; however, the NORM content of the mud needs close consideration in any application, for example the potential to have ^{210}Po and ^{210}Pb in dusts if the mud is thermally processed.

6.4. IRON

6.4.1. Introduction

Iron ore almost always consists of iron oxides, the primary forms of which are magnetite (Fe_3O_4) and haematite (Fe_2O_3). Iron ore is the primary source of pig iron for the world's iron and steel industries. It is therefore essential for the production of steel, which in turn is essential to maintain a strong industrial base. Almost all (98%) iron ore is used in steel making. Iron ore is mined in about 50 countries, but the seven largest of these produce about three quarters of total world production (Table XXIII). Australia and Brazil together dominate the world's iron ore exports, each having about one third of total exports [112].

Owing to their geochemical properties, iron ores, in particular when they are weathered, are good scavengers for radionuclides and heavy metals.

6.4.2. Processes

Owing to the large quantities produced and the nature of the deposits, iron ore mines are typically open pit mines. Lump iron ore is exported around

TABLE XXIII. WORLD IRON ORE MINE PRODUCTION ESTIMATES (10^3 t) [122]

Country	1999	2000
China	209	215
Brazil	190	190
Australia	155	158
Russian Federation	81	80
India	68	68
USA	58	61
Ukraine	48	50
Canada	34	34
South Africa	30	30
Sweden	19	19
Mauritania	12	12
Kazakhstan	9	10
Other countries	82	80
World total	994	1010

the world, but is sometimes pre-treated thermally to improve its characteristics in the blast furnace for the production of iron. At many sites iron ore fines are thermally processed, after agglomeration, in pelletizing or briquetting processes.

There is an increasing trend to carry out a substantial degree of pre-reduction of ore to enable its use in various direct steelmaking processes. The thermal pre-treatment processes give rise to fumes and dusts, which are recovered and recycled. Recycling can give rise to accumulation of volatile components, including volatile radionuclides such as ^{210}Po and ^{210}Pb. An overview of the process is shown in Fig. 21.

6.4.3. Residues

Ferrous slags are valuable co-products of iron and steel making. In 2000, about 19 Mt of domestic iron and steel slags were consumed in the USA. Of this, iron or blast furnace slag accounted for about 57% of the tonnage sold. Steel slags, produced from open hearths, basic oxygen furnaces and electric arc furnaces accounted for the remainder.

The presence of small concentrations of uranium in feedstocks (iron ore, coke, limestone, etc.) for iron and steel making can result [132] in enhanced

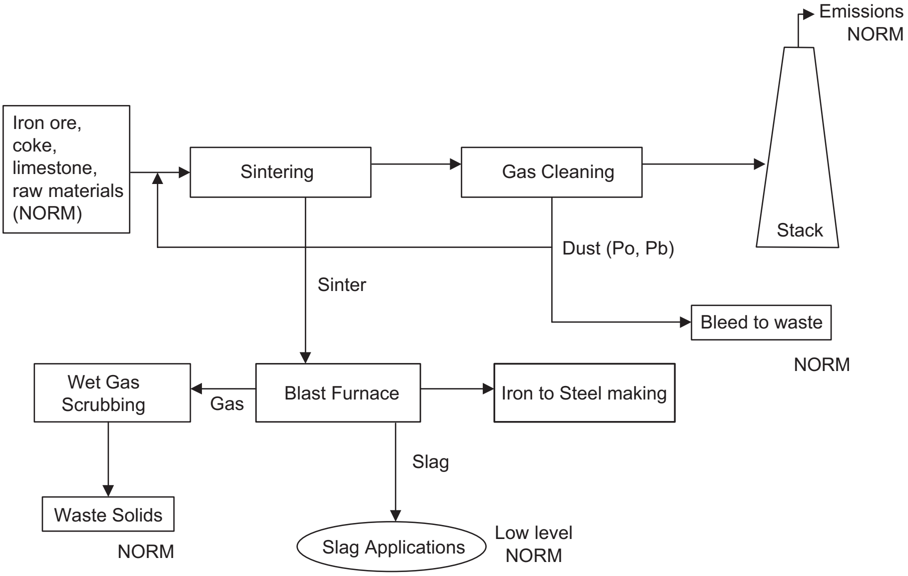

FIG. 21. Generalized flowsheet of iron production from iron ore.

levels of ^{210}Po and ^{210}Pb in recirculated thermal processing plant off-gas cleaning dust. Depending on the site specific gas cleaning processes used, the concentrations of these radionuclides in the small quantities of collected dust can be such that any bleed-off dust must be examined for its potential to contribute to human exposure and environmental pollution. However, most of the uranium progeny, and therefore radioactivity, occurs at lower concentrations in downstream process streams including slags and off-gas cleaning sludges, as they are not recirculated and therefore do not accumulate. Although the concentrations in iron and steelwork slags are low, there may be potential for exposure if such products are subsequently used by other industries, such as in cement manufacture, where the workforce might be exposed to a dusty workplace. The major uses of iron slag were for road bases (29%), asphaltic aggregates (19%), cement and concrete applications (18%) and fill (9%). Steel slags were mainly used for road bases (30%), asphaltic aggregates (27%) and fill (13%) [122]. In older houses in Europe, metal smelting slags were also widely used, for example, for infilling the space between the ceiling of one floor and the floorboards of the floor above. Radium-rich slags from metal production (100–200 Bq/kg) were sometimes used. Slag was also used in earlier years as a building material. Owing to the mineralogical, i.e. glassy, properties of slags, any radioactivity contained in them is unlikely to migrate.

Radioactivity has been measured in iron sintering plants in the Netherlands and the UK [133–135], but some major world producers have not indicated whether there is radioactivity in their processes. The ores used for metal production in, for example, Sweden at present normally have low levels of natural radioactivity [136]. Some steel companies deal with NORM (in association with other hazardous components) in their processes at concentrations of less than 100–1 000 000 Bq/kg in quantities of up to millions of tonnes per annum. Concentrations of radioactivity in wastes for disposal of up to 100 000 Bq/kg have been reported. The quantities of NORM waste involved were recently published in an EU report [6].

Consideration has been given to the recovery of valuable metal from steelwork wastes, and treatment options were reported on from furnaces in the Netherlands [134]. Studies are under way on the potential to treat wastes, such as dusts containing ^{210}Po and ^{210}Pb, to reduce the volumes of waste and possibly to condition the waste into an appropriate form for disposal.

Downstream intermediates and wastes containing low concentrations of radioactivity will need to be assayed to ensure that activity is not accumulating as TE-NORM, such as ^{226}Ra bearing scales in gas scrubbing processes, which need to be handled appropriately.

6.5. COPPER

6.5.1. Introduction

Copper continues to be one of the most important base metals in industrial societies because of its high electrical and heat conductivities, its corrosion resistance and ductile properties. The major application is in electrical installations and electronics as wires and conductive layers. It is also used extensively as a major compound and alloying metal in brass, bronze, German silver for armatures, mechanical parts, coinage, etc. Copper compounds, such as copper sulphate, are used as anti-foulants in agriculture and marine engineering, and as a precipitation agent in water treatment.

Copper is sometimes found as the native element and is present in many minerals. Important production deposits are listed in Table XXIV. The most important copper ores are the oxides, sulphides and carbonates.

Owing to their geochemical properties, silver, gold and uranium are often associated with copper. Historic copper mining wastes are processed at various places for these elements. In more recent projects, such as at Olympic Dam in Australia, separation is integrated into the overall milling process, thus

TABLE XXIV. WORLD COPPER ORE MINE PRODUCTION ESTIMATES (10^3 t) [122]

Country	1999	2000
Chile	4 382	4 500
USA	1 600	1 450
Indonesia	740	850
Australia	735	760
Canada	614	650
Peru	536	530
Russian Federation	530	520
China	500	510
Poland	460	480
Mexico	362	390
Kazakhstan	374	380
Zambia	260	260
Other countries	1 500	1 600
World total	12 600	12 900

enhancing the commercial value of the operation and reducing the radionuclide contents in the waste stream (by 30%).

6.5.2. Processes

Much modern copper mining is done in open pits from where the ore is hauled to a mill. In some locations, the in situ leaching process has also been used successfully to extract copper from deep ore horizons [119]. The method of extracting copper from ore has been improved with modern technology, the exact process varying with the type of ore. The non-copper minerals in the ore, called gangue, also vary.

The first step in milling is crushing. The crushed ore is mixed with water and ground down in a ball mill. The next step is a flotation process, where the copper minerals are suspended in a froth and then skimmed off, while the unwanted components of the ore sink into the slurry. The unwanted components, called tailings, sink to the bottom and are separated. The froth containing the copper is dried, and the product is called copper concentrate. It contains 15–33% copper.

Sometimes the ore cannot be processed by flotation. In such cases leaching processes, for example heap leaching, with or without microbiological action (bioleaching) are employed. After leaching, the solution is processed by solvent extraction and electrowinning to produce metal cathodes. Older technologies would have resulted in less pure products (concentrate) that must be smelted or refined to remove the rest of the impurities.

Copper sulphide concentrates can be smelted using several technologies, depending inter alia on the concentrate mineralogy. In the smelting furnace, volatile impurities rise to the surface as gas bubbles. Other impurities form a liquid — molten slag — that is less dense than the copper melt and rises to the top. The slag containing impurities such as iron, calcium oxide and silica is skimmed off the top. The melt containing up to 50% copper, called copper matte, still contains impurities including iron and sulphur. The copper matte goes through a converter. Applying air forces silica back into the copper matte to gather up the impurities and make more slag, which is then separated off. The resulting metal contains up to 99.5% copper and is known as blister copper.

For use in electrical wire and instruments, copper must be of even higher purity, which is achieved by electrolytic refining. Blister copper is poured into molds to make slab-like anodes of about 1 m × 1 m and 7 cm thick. The copper anodes are lowered into tanks containing a solution of copper sulphate in sulphuric acid. They are arranged so that hanging between them are sheets of pure copper that act as the cathodes. An electric current is passed through the tank. The anode copper slabs dissolve. Impurities sink to the bottom as sludge. Pure copper is deposited on the cathode sheets.

6.5.3. Residues

Copper slags, like other metal smelting slags, may find further uses, for example as grits for ‘sand’ blasting applications [137].

Investigations at gold and copper mines in Cuba [138] have indicated that the ^{226}Ra concentrations range between 53 and 7216 Bq/kg in various minerals and residues. Water samples had a ^{226}Ra concentration of between 4 and 160 Bq/L. A study on NORM in the copper industry of Arizona in the USA [119] reported that groundwater from six mines had ^{226}Ra concentrations ranging from non-detectable to 4.81 Bq/L, ^{228}Ra from non-detectable to 4.51 Bq/L and total uranium from non-detectable to 7.73 Bq/L. That same study found extremes for NORM in surface water, particularly for uranium nuclides such as ^{238}U, that ranged from 0.004 Bq/L to 25 Bq/L in 19 samples taken from five mines. Some of the uranium deposits exploited in eastern Germany are associated with copper shales. Uranium concentrations in these shales are quite variable, but may reach up to 2 g/kg [113].

6.6. GOLD

6.6.1. Introduction

The major use of gold is in the manufacture of jewellery, while a significant amount of gold is also retained by central banks in Member States to support the value of their currencies. Industrial uses relate to the high electrical conductivity of gold, together with its malleability and resistance to corrosion, for example the manufacture of components for electrical products and equipment. The high reflective power of gold is relied upon when used in window pane coatings, as shielding material to protect spacecraft and satellites from solar radiation, and in industrial and medical lasers using gold coated reflectors to focus light energy. Because gold is biologically inactive it has traditionally been used in dental care, has become a tool in medical research, and is also used in the treatment of arthritis and other intractable diseases. More recent research seems to indicate a useful application for gold in catalytic converters to reduce vehicle emissions.

While only low levels of NORM are associated with gold deposits in some geological formations (e.g. in the Archaean greenstone belts at Kalgoorlie in Australia and in parts of Africa), it was found that the detailed information available warranted its inclusion in the discussion here as an example of what can be done in other types of mining. The discussion below will, therefore, focus on those formations with enhanced levels of NORM. A classification of such formations as uranium deposits is presented, for instance, in Refs [140, 143]. Some of the better known deposits are the placer gold deposits of the Witwatersrand Basin in South Africa [142] and, together with copper, the predominantly lode gold deposits in the Jacobina and Carajás regions of Brazil, the Copper Belt of Arizona, USA [119], and Olympic Dam in Australia [141].

World annual gold production at the end of the twentieth century amounted to around 2400 t/a, with details for the larger producers indicated in Table XXV. In South Africa the average gold ore grade is around 5 $g \cdot t^{-1}$ [143]. Extrapolating this to the world gold production above gives a global raw materials extraction rate of around 5×10^8 t/a or 3×10^8 m^3/a. Because the volumes of waste rock, products and by-products permanently removed from the ore are relatively small, tailings production would not be much different from the amount of ore extracted. The total inventory of mine tailings in South Africa is estimated for gold mines alone at around 6×10^9 m^3 [143]. The global extraction of raw materials may be more difficult to obtain except through extrapolation from the gold production information.

TABLE XXV. WORLD GOLD PRODUCTION [122]

Country	1999 (t/a)	2000 (t/a)	2000 (%)
South Africa	449	440	18
USA	341	330	14
Australia	303	300	12
China (estimated)	170	170	7
Canada	158	150	6
Peru	128	140	6
Indonesia	130	120	5
Russian Federation	104	105	4
Other countries	735	665	27
World total (rounded)	2500	2400	99

6.6.2. Processes

Small scale mining seems to be a diminishing practice, but may still be relevant in some developing countries. In large scale mining, shallow and deep deposits are still mined mostly through conventional open pit and underground mining methods, respectively, in some cases on a very large scale. Almost all ore and waste rock is brought to the surface for processing.

Gravity concentration was generally used as the main extraction process for gold in the early days. At present, cyanide leaching (mostly in leach tanks, but also through heap leaching) is almost exclusively being used together with carbon adsorption and elution, followed by gold precipitation and melting, otherwise by electrowinning from the leach solution. Gravity concentration may still be used in small scale operations or as an initial step in large scale operations to prepare a low cost cyanide leach solution for electrowinning, but the low gravity stream will probably still go through a cyanide leaching process.

Mined ore, pulped and milled, is the main feed material for gold extraction. Various but generally small amounts of pyrites from the same ore body may be present in the process stream. Occasionally (e.g. at some South African mines previously or still involved in uranium production), pyrites may be separated and calcined to iron oxide in sulphuric acid plants (Section 16.1). The calcined material is usually subsequently also processed for gold extraction.

When performed in a slightly alkaline environment, the selectivity of the cyanide leaching process for gold and other precious metals is good and only insignificant amounts of radioactivity are partitioned into this stream. These may end up in slimes from the electrowinning process or in slag from melting furnaces. Activity concentrations are, however, very low.

Disregarding the relatively small amount of radioactivity that may be in extracted uranium products (e.g. at some mines in South Africa) and the minor residues mentioned above, the radionuclide content of the tailings remains very similar to that of ore.

6.6.3. Residues and their management

6.6.3.1. Form

The residues from gold mines include tailings, waste rock and wastewater from either dewatering operations or as fissure water, the latter, however, usually ending up as process water. The remaining underground ore also constitutes a residue to be considered in terms of its influence on mine water.

6.6.3.2. Tailings

During their use, the maintenance of tailings impoundments includes the maintenance of drainage systems and paddocks to control seepage and surface run-offs and even the slime discharge operations. Tailings impoundments, which cannot be reprocessed economically, can also serve as burial sites for limited quantities of other homogeneous waste. In South Africa residues with activity concentrations below 1×10^6 Bq/kg and diluted to below 1×10^5 Bq/kg, either created during normal operations or during demolition (e.g. building rubble), can be diluted or buried in tailings impoundments. Materials with concentrations above 1×10^6 Bq/kg are presently stored pending a solution for their disposal. Their impact when disposed of in tailings impoundments is presently being investigated [144].

Rehabilitation of tailings impoundments involves the stabilization of impoundments and the establishment of vegetation, which sometimes requires specialized pre-treatment and selection of suitable plant species. Owing to the large areas involved and the associated cost, capping may only be performed on a limited scale, based on site specific assessments (e.g. to prevent chemical pollution of shallow groundwater sources or where exhaled radon affects inhabitants living very close to the source). While rehabilitation of tailings impoundments sometimes starts during their use, for others this process only follows when the impoundment becomes redundant. Better located impoundments are created through the reclamation of the older impoundments still containing economically recoverable gold.

6.6.3.3. Waste rock

The inventory of waste rock is usually much smaller than that of the tailings. In South Africa it amounts to an estimated 6 × 10^8 m^3 (minus the amount reused) from underground mines [145]. Water ingress into waste rock dumps may generate significant chemical pollution due to seepage of contaminated water requiring implementation of active water management measures [113].

It is important to note that many waste rock dumps are of a temporary nature, often being processed through the plant at the end of life of a mine or being crushed for use in civil construction projects. When applicable, measures to prevent the unauthorized use of the material may be appropriate.

In South Africa waste rock has been used in the past for construction work on mines, and contractors were also allowed to crush and sell waste rock. Re-use still presents a useful management option, but is presently only allowed following formal authorization for unrestricted use (e.g. residential construction) or restricted use (e.g. the construction of roads and bridges). External gamma radiation and indoor radon doses around 20 and 80 μSv/a, respectively, have been assessed for residential floor slabs containing waste rock with nuclide concentrations within the range 10–50 Bq/kg as an aggregate [146]. The use of waste rock as capping material for tailings impoundments is also being investigated [147].

6.6.3.4. Exposed ore and residual ore

This section relates to reef sections and ore not removed from mining areas and exposed to water and oxygen. Underground water is typically collected in settlers at the bottom of the mine, where lime is added to precipitate heavy metals. This also reduces radioactivity, especially uranium and thorium concentrations [148]. The sludge is sometimes removed to gold or uranium extraction plants for processing, but may also be pumped onto tailings impoundments. Overflow water from settlers is mainly recycled as process water, but excess water may be discharged to the environment.

For the larger dewatering and post-closure mine flooding problems, various methods for water treatment have been investigated (e.g. reverse osmosis, ion exchange, precipitation methods and anaerobic bacterial techniques) [149]. The GYPSIX method starts with the addition of lime, with subsequent treatment of the water with cation and anion ion exchange resins for regeneration. Various alternative acid and alkali regeneration and subsequent precipitation processes may result in marketable products [149]. Other investigations [114,115] stress the importance of proper planning of mine closure on a regional basis, with the objective of minimizing the volumes and improving the quality of contaminated water that may require treatment prior to discharge. Radiological impacts are treated as part of some studies but still need to be integrated into others.

6.6.3.5. Contaminated areas and materials

NORM contamination in ore processing areas mainly relates to inevitable spills of process materials or scales formed during operations. Activity concentrations are mainly similar to those of the associated process materials but are higher in scales or where selective sedimentation has caused concentration. Although cleanup operations are performed continuously, final cleanup operations are more likely only to be performed during the decommissioning phase.

A typical process area may cover several hectares of land, while each mine may have several process areas. The combined surface area of process areas at South African gold mines is estimated to be of the order of a thousand hectares. Excavations to clean up such areas typically go down from a fraction of a metre to a few metres.

Enhanced concentrations of radionuclides in the underground water of gold mines contaminate some materials such as underground wood. When ending up in process water, the underground water also produces radioactive scales on pipes and process equipment from underground areas and gold plants. Uranium nuclides seem to be the dominant source of radioactivity in the scale. Subsequent detachment of scales normally also causes the contamination of surrounding soils, especially in mine salvage yards but in the past also during storage and processing at recyclers. During an investigation of the upper range of the problem in South Africa, scales with total activity concentrations within the range 9.0×10^4–3.3×10^5 Bq/kg have been collected in the salvage yard of a gold plant, causing activity concentrations up to around 1×10^4 Bq/kg of scrap [150]. The average value for the complete gold industry is, however, estimated at only a few per cent of this [151]. Following studies which indicated that doses from the resulting steel are trivial, most of the scrap from gold mines in South Africa is presently recycled via authorized smelters [150, 151]

An estimated 4×10^4 m^3 of bulk wastes (e.g. scales, rubbers, plastics, wood, ceramics, but also decommissioning waste) will eventually be produced by South African gold mines, including associated uranium and sulphuric acid plants [144]. While most of these may find their way to tailings impoundments, those with activity concentrations above 1×10^6 Bq/kg are presently stored on different mining sites pending a solution for their disposal. The profitable recovery of gold from residues, subsoil, scales, concrete surfaces and even wood during demolition of gold reduction plants provides a great financial incentive for proper cleanup operations [152]. This involves the recycling of these materials through the milling and extraction plants. The recycling and reuse of equipment (e.g. metal scrap and electric motors) is normally also maximized.

6.6.3.6. Former mine land

These are lands from previous mining operations, including sites contaminated through previous metallurgical plant operations and sites from which tailings impoundments have been removed for reprocessing. Cleanup criteria are aimed in the first instance at decontamination to legally defined levels below which the land can be excluded outright from any further regulatory concern, but they may also be influenced by future land use (e.g. suitability for residential development or restricted to industrial use only). More than 20 such sites have been or are in the process of being remediated in South Africa [153].

Cleanup of former mine land and other contaminated sites is mainly performed by excavating the contaminated soil to approved levels, predetermined by pathway modelling of doses associated with residential, industrial, recreational or other use [154]. Other options include cleanup to levels statistically indistinguishable from those of surrounding unaffected land or to an optimized level below a given reference level. When economic values justify this, excavated residues may be recycled for the recovery of gold. Otherwise material has been removed to a tailings impoundment of a nearby licensed mine [155].

6.6.4. Radionuclide contents

Globally, NORM levels within gold deposits vary greatly within an overall range of 10–15 000 ppm with smaller variations within specific deposits. Within the Witwatersrand Basin the overall range is 10–750 ppm with the average contents for a reef more likely to be in the range 20–350 ppm [144]. Uranium may be extracted as a by-product from these reefs if they contain above about 350 ppm of the metal. NORM concentrations in the Jacobina and Carajás deposits are lower at around 9 ppm U and 12 ppm Th [156]. In Arizona, NORM is associated with various metal deposits, for example porphyry and vein deposits of copper, lead and precious metal sulphide ores. The estimated uranium contents range widely, from around 55 ppm in copper sulphide ore in schists and porphyry copper deposits at Globe and Miami to 7600–14 000 ppm in copper oxide veins in altered andesite at the Copper Squaw underground mine [118]. In Australia the average grade of the Olympic Dam breccia complex deposit is given as 600 ppm U_3O_8 [141].

Where gold is extracted as a by-product from primary copper ore, the radioactivity problem with regard to gold mainly originates from NORM in the anode slimes generated during the final stage of the copper extraction process, from which gold is extracted. In Australia, the slimes can in extreme cases contain ^{210}Pb at (1–3) × 10^5 Bq/kg and ^{210}Po at 6 × 10^5 to 1 × 10^6 Bq/kg, but these are refined on-site. More of a problem are slimes containing ^{210}Pb at (1.5–2) ×10^4 Bq/kg and ^{210}Po at (6–8) × 10^3 Bq/kg produced in relatively small quantities, which are removed from the site for refining elsewhere [157]. No published information is available and the problem is currently being investigated.

6.6.5. Environmental impacts

The impacts of oxidized pyrites and associated acid leaching from tailings, waste rock piles and ore remains on aquatic systems are likely to cause the largest impacts from gold mining. A general discussion on this subject is presented in Section 3. The impact of the South African gold mining industry in terms of water consumption from a river was found to be represented by age weighted annual doses within the range 0–0.5 mSv/a [158], with the higher doses coming from mine release points. That study seems to confirm results of an earlier study concluding that the radiological impact of South African gold mine releases on surface water sources is reduced over relatively short distances to almost background levels [159]. Presently the study is being extended to the river sediments [160]. Studies done as a regulatory requirement at South African mines indicated the radon and dust doses to be even lower. For radon, background concentrations were also measured within the range 14–111 Bq/m^3 with an average of 48 Bq/m^3 and a standard deviation of 19 Bq/m^3 [161]. With this background distribution the contribution of the mine tailings sources became statistically insignificant at the locations of the critical groups. The studies covered only the existing situation at mines, and post-closure assessments still need to be done.

6.7. MINERAL SANDS AND RELATED INDUSTRIES

6.7.1. Introduction

Heavy minerals, defined as those with a density above 2.8 g·cm^{-3}, are minor constituents of a wide range of rocks, and comprise a wide variety of minerals, including oxides, phosphates and silicates. They are typically harder than the other minerals in the rocks from which they originate and, therefore, survive the erosion and transport processes. Hence, economic deposits of heavy minerals occur predominantly when they have been concentrated by marine, alluvial and/or wind processes and are called placer deposits.

Heavy minerals such as monazite, zircon, xenotime, ilmenite and rutile and ores such as cassiterite are the raw materials in the extraction processes of certain metals or their compounds. Zirconium, titanium, thorium, tin and the rare earth elements (REEs) are the major target elements. REEs comprise 16 chemical elements, including those with atomic numbers 57 (lanthanum) through 71 (lutetium), as well as yttrium (atomic number 39), which has similar chemical properties.

Indicative production quantities of these minerals worldwide are given in Table XXVI. Thorium production is discussed in more detail in Section 8.3.

Tin is used to coat other metals to prevent corrosion. It is also used as an alloying agent, for example in bronze, and as a biocide in antifouling paint. A crystalline tin–niobium alloy is superconductive at very low temperatures.

Titanium metal is used for alloys with aluminium, molybdenum, manganese, iron and other metals. These alloys of titanium are used principally in the aerospace industry. Titanium is a component of human joint replacement parts, including hip ball and sockets. It has excellent resistance to sea water and is used for propeller shafts, rigging and other parts of ships exposed to salt water. A titanium anode coated with platinum provides cathodic protection

TABLE XXVI. APPROXIMATE PRODUCTION FIGURES FOR VARIOUS HEAVY MINERALS [162]

Zircon	$ZrSiO_4$	100 000 t/a in the USA 400 000 t/a in Australia 400 000 t/a in South Africa 150 000 t/a from various other sources
	ZrO_2	1 070 000 t worldwide in 2001
Rutile	TiO_2	414 000 t worldwide in 1998
Ilmenite	$FeO{\cdot}TiO_2$	3 966 943 t worldwide in 1990, includes leucoxene
Baddeleyite	ZrO_2	20 000 t/a in South Africa (now discontinued)
with by-products such as		
Monazite	$(Ce,La,Y,Th)PO_4$	~23 000 t worldwide in 1990, excluding the USA
Xenotime	$Y(PO_4)_2$	Total world Y production from xenotime and monazite was 2400 t in 2001.

from corrosion by salt water. Titanium dioxide is used extensively as white paint pigment, as it is permanent and has good covering power, and is not toxic as opposed to, for example, lead white. TiO_2, in fact, accounts for the largest use of the element (>95%).

The last few decades have seen an increasing use of REEs, for instance as polishes, catalysts, incandescent and other special glasses, steel additives and as burnable poisons in nuclear reactors. $EuVO_4$ and Eu_2O_3 phosphors provide the red colour in colour television tubes.

Of world zirconium production, more than 95% is used as zircon in foundries (30%), refractory manufacture (30%), ceramics (30% as glazes) and glass making. The majority of the zircon used is consumed as a zircon ‘flour’ with a particle size of around 1 μm. Zirconium metal is used in the nuclear industry for cladding fuel elements since it has a low absorption cross-section for neutrons.

Niobium metal is a component of some stainless steels and also alloys with non-ferrous metals. These alloys have good strength and other properties, and are used in pipeline construction.

6.7.2. Occurrence

Heavy minerals occur in mineral sand placer deposits and in veins, or are disseminated predominantly in alkaline intrusions in hard rock. Well known placer deposits in the Indian Ocean region are the cassiterite sands of Southeast Asia (Malaysia, Thailand and Indonesia), the tin province of Australia along the West Pacific, heavy mineral placers on the coasts of Mozambique, South Africa [163], western Australia, northeast Sri Lanka and the western and eastern coasts of India, with other smaller deposits in the USA.

Ilmenite ($FeTiO_3$), rutile, leucoxene and sphene are sources of titanium and are usually associated with iron. Titanium is also readily mined in one of the purest forms, rutile (TiO_2), from beach sand. The deposits are mainly located in the Americas, Australia, sub-Saharan Africa, Scandinavia and Malaysia.

Monazite ($[Ce,La,Y,Th]PO_4$), xenotime (YPO_4), bastnaesite ($[Ce,La][CO_3]F$) and others are the source of REEs. Since some REEs behave (geo-)chemically rather similarly to uranium or thorium, the latter may be found associated with them. Monazite forms in phosphatic pegmatites but is actually a standard trace constituent in many ordinary igneous, metamorphic and vein filling rocks. Notable occurrences of monazite are widespread and diverse. They include beach and river sand deposits in India, Australia, Brazil, Sri Lanka, Malaysia, Nigeria, and Florida and North Carolina, USA. In Malaysia [164] the minerals are also by-products of ‘amang’ processing plants.

There are notable occurrences of xenotime in Arendal, Hittero and Tvedestrand, Norway; Madagascar; several locations in Brazil; and in Colorado, California, Georgia and North Carolina, USA.

Large resources of yttrium in monazite and xenotime are available worldwide in both ancient and recent placer deposits (monazite and xenotime), weathered clay deposits (ion adsorption ore), carbonatites and uranium ores. It is probable that the world's resources are very large relative to expected demand.

Cassiterite occurs in the form of placers in alluvium, as well as lodes in hard rock. Granites have been shown to contain significant amounts of radium [165]. According to Ref. [166], accessory minerals containing uranium and thorium are commonly found in granitoids, alluvium and residual soils derived from granitoids. Thus, it is not surprising to observe that alluvial tin ores, in for example Malaysia, are associated with NORM due to the presence of minerals such as monazite, zircon ($ZrSiO_4$), xenotime, ilmenite, struverite (Ta/Nb bearing TiO_2), columbite ($[Fe,Mn][Nb,Ta]_2O_6$), tourmaline and others.

Bastnaesite deposits in China and the USA constitute the largest percentage of the world's rare earth economic resources, while monazite deposits in Australia, Brazil, China, India, Malaysia, South Africa, Sri Lanka, Thailand and the USA constitute the second largest segment. Xenotime, rare earth bearing (ion adsorption) clays, loparite, phosphorites, apatite, eudialyte, secondary monazite, cheralite and spent uranium solutions make up most of the remaining resources. Undiscovered resources are thought to be very large relative to expected demand.

6.7.3. Processes

There are different types of processes, depending on the source types (hard rocks or placers), to concentrate minerals or to process them to produce various products at hydrometallurgical plants. These processes usually produce residues containing certain levels of radionuclide concentrations. Heavy minerals are extracted from mineral sand placer deposits or from hard rock using separation techniques. An example of the complex separation scheme is shown in Fig. 22. The mineral products are in turn used as raw materials in the production of mineral flour (e.g. zircon), refined metals or metal oxides.

The mining methods that can be employed are dictated by the type and location of the ore body. Placer and similar near surface deposits can be conveniently exploited by various dredging methods, while mineralizations in hard rocks require conventional deep mining techniques. Tin ore concentrates may also be recovered from re-working mineral sand that was originally mined for

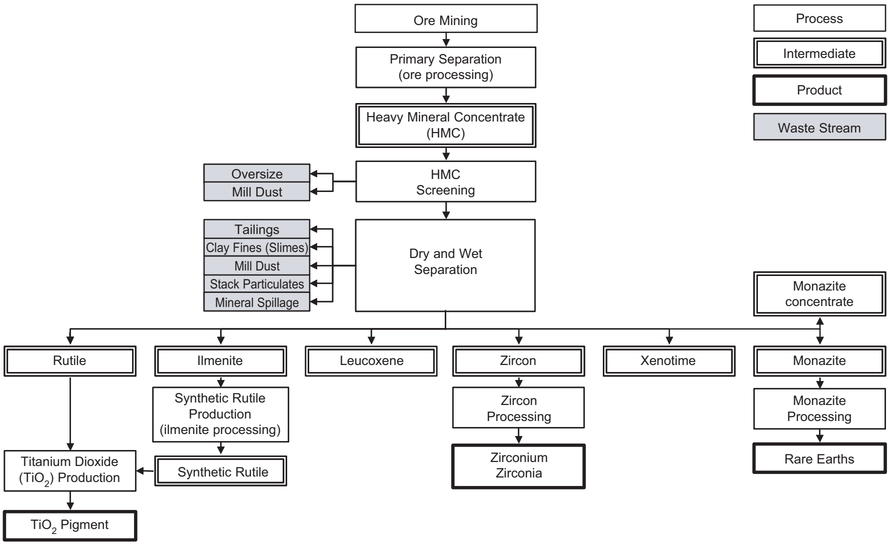

FIG. 22. Flow sheet showing separation of heavy mineral ore into pigment, zirconia/zirconium and rare earths, as well as the associated major waste streams (after Ref. [167]).

a different mineral value. Appendix A gives details of, as an example, the related mining and processing practices in Malaysia.

Dredging and hydraulic processes are mostly used to collect heavy mineral sands after the removal of topsoil. In general, mine sand residues are returned to the mined-out areas after removal of the heavy minerals, but very often the sand is stored and sold for other industrial purposes (e.g. construction).

In hard rock mining at Palabora (South Africa) baddeleyite is obtained from an open cut operation and is separated from other commercial minerals including copper and phosphate minerals. This operation may be extended to an underground operation.

In several areas of the world mining activities have developed to recover raw materials from different types of deposits and to produce marketable concentrates. Depending on the geological sources, special flowsheets for the recovery and concentration of the raw materials (hard rock, placers, by-product concentrates) have been developed. A generalized secondary separation process flowsheet for heavy minerals production is also given in Fig. 22.

Heavy minerals in sands are initially concentrated by wet gravity processes and then separated by a combination of dry magnetic and electrostatic processes into concentrates of the various minerals. Improved separation/upgrading might be required using further comminution and recycle, magnetizing, roasting and/or leaching. The mineral concentrates can then be sold or chemically processed further.

6.7.3.1. Titanium

Rutile can be sold directly into the market, but ilmenite is often further chemically processed to produce synthetic rutile [167], or pyrometallurgically treated to produce a titania slag — see Fig. 22 and Appendix II. Most of the titanium minerals (>95%) are chemically processed into pigment [167]. The titanium minerals are isolated from the raw ore by various hydrometallurgical processes, the layout of which is illustrated in Fig. 22. The further processing into the metal or purified TiO_2 takes different dry and wet chemical routes that are discussed in detail in Appendix II. The wet TiO_2 production process employs either sulphuric or hydrochloric acid.

6.7.3.2. Zircon

Zircon ($ZrSiO_4$) is predominantly sold and used as zircon without further processing, other than possibly milling to produce zircon flour. It is generally believed that there is little benefit in trying to remove radionuclides from zircon prior to downstream applications or before waste disposal because the zircon matrix is probably the most stable form in which to keep the radionuclides. A general flowsheet from heavy minerals to zircon end products is presented in Fig. 22 [167].

6.7.3.3. Tin

At tin smelting plants, tin ore concentrates from tin mining or tin tailing processing plants (see Appendix A for examples from Malaysia) are used as feed materials to produce metal tin. At the end of the process, tin slag is produced as residue. Tin smelting, dating back to prehistoric times in Britain, has resulted in millions of tonnes of glass-like slag in various parts of the country, but the hazard from this material is likely to be low [168]. Tin slag typically contains significant amounts of tantalum and niobium. It can be used as raw material to the tantalum extraction plants. Struverite is also a good raw material for tantalum extraction. However, secondary residue containing naturally occurring radionuclides is expected to be produced from the extraction plants.

6.7.3.4. Niobium and tantalum

Niobium minerals usually contain both niobium and tantalum. Since they are rather similar chemically, it is difficult to separate them. Niobium can be extracted from the ores by first fusing the ore with alkali, and then extracting the resultant mixture into hydrofluoric acid, HF. Current methodology involves the separation of tantalum from these acid solutions using a liquid–liquid extraction technique. Electrolysis of molten fluorides is also used [136].

A detailed description of the niobium production processes in Brazil, which is the most important producer in the world, is given in Appendix IV. Operational environmental impacts associated with discharges of liquid effluents into the environment can be considered negligible. In a broad way it can be said that at least 60% of each individual radionuclide total activity leaves the operational process along with the residues generated in the floatation step. In the case of ^{238}U this figure reaches 70% of the total. This finding has a very relevant bearing concerning economic recovery of uranium. However, the contamination of groundwater by ^{226}Ra and ^{228}Ra in the area of influence of the settling ponds containing barium sulphate precipitate is a relevant issue. Typical concentrations of about 5.0 Bq/L for each of the above mentioned radionuclides have been observed. In this case, the adopted management strategy/mitigation used by the company is to pump the water from the aquifer and treat it before release into the environment. The niobium industries described in Appendix IV did not show any relevant environmental radiological impact due to the release of effluents into the environment. The main concern relates to the potential use of some of the process residues as landfill for house construction sites, for which very high effective doses may be observed. The contamination of groundwater, as has been reported for one of the industries, is also of concern if people come to have direct assess (ingestion) to this water.

6.7.3.5. Rare earth elements

The main minerals used as sources of REEs are monazite, xenotime from placers and bastnaesite (China), coperite and pyrochlore from hard rock. Specific extraction processes have been developed by the local industries. An overview of the extraction processes is given in Fig. 21 and more details are available in Appendix III.

6.7.4. Residues

Dredging of surface deposits has left behind thousands of mined-out ponds worldwide. Typically such ponds have thick layers of slime. The slurries contain heavy minerals such as ilmenite, monazite, zircon and xenotime. The mined-out ponds are very large in size and most of them are quite deep. Remediation is often necessary before the former mining sites can be re-used.

Residues are produced from mining, beneficiation and chemical processing of mineral sands and minerals (Fig. 22). They are produced as tailings, fine dust, sludge (oxides, hydroxides or sulphates), scales and slag containing concentrated natural radionuclides. Residues from all types of production can cause a disposal problem because of the radioactivity content. Research and development is carried out to study the disposal impact on the environment.

The volume and the activity level of radionuclides in residues varies depending on the processing methods applied. In the mining of placer deposits, the volume of tailings generated can be very large. The tailings are usually dumped above ground in the form of huge heaps.

The main waste generated during the wet and dry processing of heavy mineral sands is the waste from the dry plant, where the activity contained can be enhanced. Slime and sand tails from wet processing are generally low in radioactivity and can be returned to the mined-out sites. The issue of whether mineral processing residues should be recombined, kept separate and/or covered needs to be assessed on a site by site basis.

Details of the management of the waste streams generated during mining and processing of heavy minerals are given in Table XXVII.

6.7.4.1. Titanium

Thorium-232 and ^{238}U could potentially go to liquid waste streams arising from further chemical processing of the ilmenite/rutile process intermediates, but expected doses are very low. Disposal of solid residues from processing is expected not to contribute significantly to pollution caused by the dissolution and migration of radionuclides. There are exceptions to this, where the sands contained significant amounts of sulphide, and acid mine drainage has subsequently mobilized pollutants such as manganese.

The nature of natural rutile, ilmenite and zircon, i.e. their chemical stability, means that for most groundwater environments there is little likelihood of radionuclides contaminating the waste disposal sites generally used as repositories for the heavy minerals, refractories containing zircon or foundry waste.

In ilmenite processing to produce synthetic rutile and titanium dioxide pigment, large amounts of hydrated iron oxide (30 000 t/a is produced in Malaysia) and gypsum residues are produced and stored in ponds or dumped above ground on the plant premises.

The dumping at sea of the waste from TiO_2 production, dilute sulphuric acid, caused serious environmental concern and has been stopped, at least in Europe. Per tonne of TiO_2 produced, 6–8 t of a dilute (20–22%) acid are generated. The acid is too dilute for further commercial use and often contaminated by heavy metals and, indeed, NORM. In a process with a relatively high energy consumption the acid is concentrated and accessory salts, mainly ferric sulphates and co-precipitates, are removed. The acid is fed back into the production process, while the ferric sulphate is used commercially as a flocculant for wastewater treatment, mainly for removal of phosphates as sparingly soluble ferric phosphates. Any ferric oxides produced are landfilled.

Fewer environmental pollution problems are encountered when pigment is produced from rutile rather than ilmenite. The chloride process, using a rutile feed, generates about 0.2 t of waste per tonne of TiO_2 product; the sulphate process, using ilmenite, generates about 3.5 t of waste per tonne of product. Producing synthetic rutile from ilmenite results in about 0.7 t of waste, mainly iron oxide, per tonne of product. Direct chlorination of ilmenite generates about 1.2 t of waste, mainly ferric chloride, per tonne of TiO_2 [125].

In the chemical processes described above, the titanium is removed from the other metals present. All radionuclides present in the feedstock (and in other process inputs, for example, petroleum coke) go to the chemical processing waste stream. The final commercial products therefore seldom have radiation issues associated with them. A range of by-product minerals produced in smaller quantities are either sold, stored or disposed of.

In some cases the radionuclides can be removed to varying degrees from the titanium intermediate products prior to being sold commercially [169]. However such processes have proved in practice to be technically difficult and costly. The waste created from this removal is similar to that produced from the downstream pigment and metal manufacturing processes. Therefore the removal of the radionuclides by the producer often serves only to move the location of the waste production, rather than to produce a waste which has less environmental risk. The early removal of radioactivity adds, possibly unnecessarily, to the overall cost of the final products. The relative quantities and the characteristics of the wastes need to be scrutinized.

The waste arising from the chemical processing of titanium feedstocks can be considered as ‘mixed industrial waste’, as it contains metals such as Pb, As, Zn, Mn, Mg, V and Nb, in addition to the radionuclides and neutralized acid effluent. This waste is kept in designated waste disposal areas, many of which

TABLE XXVII. MANAGEMENT OF WASTE STREAMS GENERATED DURING MINING AND PROCESSING OF HEAVY MINERALS ACCORDING TO THE GUIDELINES DEVELOPED FOR THE WESTERN AUSTRALIAN HEAVY MINERALS INDUSTRY [169]

	Mining and primary separation*
Oversize from screening process	**Dry mining:** Buried below sand tailings or overburden, usually at a depth of at least 3 m. Occasionally, if of a suitable quality, used for constructing on-site roads. **Dredging:** Direct discharge into the dredging pond.
Clay fines (slimes)	**Dry mining:** Pumped, either directly or as an underflow from thickeners, to shallow solar drying slime impoundments. Subsequently disposed of in mined-out pit areas. An amount may be incorporated into the subsoil as part of the rehabilitation process. **Dredging:** Portion is entrained in tailings, trapped under tailings or pumped to a thickener and combined with sand tailings. The remainder is discarded into the dredging pond.
Sand tailings	**Dry mining:** Pumped as slurry from the wet concentrator to the mined areas for disposal. **Dredging:** As above, but directly discharged in the dredging pond.
	Secondary separation
Screen oversize	Oversize from screening of minerals is transported to the spiral plant tails stockpile for subsequent disposal in designated dry plant tailings disposal sites at the mine site.
Mill dust	Dust extracted from dry mill dust extraction systems is transported pneumatically to a holding bin. From there the dampened dust is trucked to a stockpile, where it is mixed with the spiral plant tails. That mixture is transported to designated dry plant tailings disposal sites at the mine site. Dust from the monazite plant is drawn out by a dedicated dust extraction system into, for example, 2 t bulk bags. Subsequently it is transported for disposal at designated dry plant tailings disposal sites at the mine site.
Spiral plant tailings	Pumped as slurry to hydrocyclones and onto a stockpile, from where they are trucked to designated on-site disposal sites that are designed so that they can be covered by at least 3 m of sand tailings and/or overburden.
Monazite plant tailings	Separately collected, blended with mine sand tailings and disposed of into the mine pit.

For footnote see p. 81.

TABLE XXVII. (cont.)

	Secondary separation
Monazite concentrate	Either stored below ground on-site in discrete, well documented sites for subsequent reprocessing into monazite or, if treated as waste, blended with mine sand tailings before disposal in the mine pit.
Clay fines (slime)	Clay solids removed from the mill water system are pumped to shallow solar drying ponds. When partially dry, the solids are dug out and deposited on the spiral plant tails stockpile and treated as above.
Stack particulates	Atmospheric emissions are mitigated by equipping stacks with bag houses or cyclones to trap particulates. Alternatively, stack outlets may be connected to the dry mill dust extraction system. Intercepted material is disposed of with mill dust.
Mineral spillage	Collected throughout dry mills mechanically (e.g. by sweeper, ‘bobcat’ and front end loader) or by vacuuming. Dry spillage is recycled back into the process through a dry feed bin. Wet spillage can be fed back through the wet heavy mineral concentrate or feed retreatment systems. Alternatively, spillage can be taken back to the mine site as part of the dry mill tailings, or as a concentrator feed that is then spread on the mine face and re-mined.

* The radiologically benign nature of these wastes does not require delineation of the waste disposal sites other than the delineation of the former mine pit.

are lined with impermeable material to prevent leaching from the repository. In Malaysia gypsum is produced as a residue. It is estimated that about 1000 t/d of gypsum residue is produced. At the time of maintenance, small amounts of radioactive scales are also collected.

6.7.4.2. Zircon

The wastes associated with zircon processing are primarily those arising from mineral processing usually in association with the mining operation. Zircon itself along with foundry and refractory wastes are the subject of examination but generally do not represent a significant environmental problem, as they are chemically inert. Extensive studies [170] have been carried out on the landfill disposal of zircon foundry waste. Measures to minimize the inhalation of dust during handling must be adopted. When zircon is fused, in refractories or technical ceramics manufacture, the silica fume produced is collected in bag houses. The fume contains enhanced concentrations of ^{210}Pb and ^{210}Po. The production can result in the accumulation of radionuclides in off-gas systems [171], pipework and filters and in some cases in discharge to the stack gases. Protective equipment may need to be considered in some plants. The small volume of waste produced during zirconia and zirconium metal manufacture is of some concern [171] and must be stored in metal containers in special repositories. There are reports [170] that these residues are high in ^{226}Ra and that such facilities need to be licensed in the USA. Gamma radiation levels are a principal concern there, and environmental regulatory enforcement and removal actions for zircon contamination have taken place at abandoned industrial waste sites. Fused zirconia is used especially in ceramics manufacture and in glazes. About 600–700 t/a of zircon/zirconia containing bricks are produced in the UK, half for the glass industry. The bricks can contain up to 10 Bq/g of ^{238}U. A further 100 t/a of specialty zircon/zirconia technical ceramics are produced in the UK. Both bricks and ceramics are disposed of into landfill. Glazes used on fine ceramics can have concentrations of up to 6 Bq/g U and Th, which could represent a problem if present in bulk quantities. An estimate of the zircon waste in Europe [6] is about 100 000 t/a. The powder waste from filters in fused zirconia refractory plants [172] is estimated to be 20 t/a for a 2000 t refractory plant in Europe, i.e. 1 wt% of feed. This waste was assayed to contain 200 Bq/g of ^{210}Pb and 600 Bq/g of ^{210}Po. Discharge to the atmosphere is typically 150 and 50 MBq/a, respectively, for the two radionuclides. The other radionuclides remain in the refractory.

6.7.4.3. Tin

In tin ore smelting, large amounts of tin slag containing significant amounts of radionuclides are stored above ground. Tin slags are of importance owing to their niobium and tantalum content. Some tin slags are also used as landfill.

6.7.4.4. Rare earth elements

In the rare earth production industries, large volumes of thorium hydroxide have been produced. A small quantity of residue containing lead and radium is also produced through the process. The thorium hydroxide waste is normally stored awaiting the availability of final disposal facilities (inter alia Malaysia, Brazil, India and China). In Malaysia, the quantity of thorium hydroxide and lead cake wastes stored in the storage facility is estimated to be about 12 000 t. Other contaminated materials (including soils contaminated with thorium hydroxide) are estimated to amount to 11 000 t.

6.7.5. Radionuclide contents

The concentration of radionuclides in heavy minerals can vary from very low to appreciably higher depending on the sources and the extraction processes applied. Uranium and thorium are generally structurally bound into the zircon crystals with some, more accessible, radioactivity being located in weathered zones of the mineral grains, for example in ilmenite. The titanium minerals tend to contain lower concentrations of radioactivity than zircon. In most cases the titanium minerals have a low uranium content (<50 ppm) and any activity is derived from thorium and its daughters. Commercial titanium minerals generally have a total specific parent activity of less than 2 Bq/g, and many products have activities well below 1 Bq/g. The radioactivity of mineral concentrates can be significantly influenced by the presence of small amounts of other minerals, such as monazite, containing high concentrations of ^{232}Th and its progeny.

The activity concentrations of radionuclides in products and residues in the heavy mineral processing are shown in Tables XXVIII and XXIX, and in Appendix IV (Tables IL and L). It is clear that the radioactivity is related to the ^{238}U and ^{232}Th series. Thorium is the main constituent in monazite and pyrochlor but is less prominent in xenotime and zircon. Thus, processing of minerals can lead to enhancement of activity concentration of radionuclides in the residues.

Tin slags contain significant concentrations of naturally occurring radionuclides. The concentration of NORM in residues from the tin mine related processes is shown in Table XXIX.

Zircons typically can have an activity concentration of 100–10 000 Bq/kg as ^{238}U or ^{232}Th. Some Malaysian zircons [6] can contain higher concentrations of radioactivity and some South African zircons can be lower. There is some evidence that some zircon from China may contain considerably higher concentrations of radioactivity [164]. A broader range of radionuclide concentrations in zircons is given elsewhere in the literature [8, 173]

6.7.6. Remediation

The potential for dispersion of radionuclides by the reuse of old sites is recognized [6]. Guidelines [167] for modern operating mineral processing and chemical plants recommending appropriate cover of waste sites and other remediation measures, depending on the proposed land usage, might still need

TABLE XXVIII. VARIOUS HEAVY MINERAL PROCESS PRODUCTS AND THEIR RADIONUCLIDE CONTENTS [172]

Material		Th		U	
		(ppm)	(Bq/kg)	(ppm)	(Bq/kg)
Mining					
Ore		5–70	40–600	3–10	70–250
Primary separation					
Product	Heavy mineral concentration	80–800	600–6600	<10–70	<250–1700
Waste	Sand tailings	<10 ≤ 50	<100 ≤ 400	<10	<250
	Oversize	<10 ≤ 50	<100 ≤ 400	<10	<250
	Clay fines (slimes)	<30	<250	<10	<250
Secondary separation					
Products	Ilmenite	50–500	400–4100	<10–30	<250–750
	Leucoxene	80–700	600–5700	20–50	500–1200
	Rutile	<50–350	<400–2900	<10–20	<250–500
	Zircon	150–300	1200–2500	150–300	3700–7400
	Monazite	50 000–70 000	41 000–575 000	1000–3000	25 000–75 000
	Monazite concentrate (a semi-product that may either become a waste or be refined into monazite)	10 000–55 000	80 000–450 000	500–2500	12 000–60 000
	Xenotime	~15 000	~120	~4000	~100
Waste	Tailings (incl. monazite tails)	200–6000	1500–50 000	10–1000	250–25 000
	Clay fines (slimes)	~600	~5000	~30	~750
	Oversize	80–650	700–5300	50–150	1200–3700
	Mill dust	~250–2500	~2000–21 000	~10–250	~250–6200
	Stack particulates	~500–5000	~4000-41 000	~10–500	~250–12 500
Synthetic rutile production					
Product	Synthetic rutile	<50–350	<400–2900	<10–20	<250–500
Waste	Iron oxide solids	<50 ≤ 100	<400–2800	<10	<250
	Inert solids	~45	~400	~5	~100

TABLE XXVIII. (cont.)

Material		Th		U	
		(ppm)	(Bq/kg)	(ppm)	(Bq/kg)
Synthetic rutile production					
Waste	Neutralized acid effluent solids	~90–540	~700–4400	~10–60	~250–1500
	Non-magnetic fines	50–600	400–5000	10–100	250–2500
	Kiln scrubber solids	~25–270	~200–2200	~3–30	<100–750
	Kiln discharge oversize	~135	~1100	~15	~400
TiO_2 pigment production					
Product	TiO_2 pigment	Nil			
Waste	Residue slurry (wet weight)	300	2500	30	750
	Filter cake (dry weight)	230–350	1900–2900	30–40	750–1000
World mean	Soils and rocks	10	~80	3	~80

to be used for the rehabilitation of old, legacy mine sites and/or at closed processing sites. Disposal of solid wastes from the treatment of baddeleyite in Europe is considered to have the potential to give the highest doses.

Pressures for land use along coastal regions and for water use are common. These pressures are also major drivers of the waste disposal and land rehabilitation techniques selected. Potential for exposure is reduced by burying and/or covering, followed by revegetation to reduce the chance of erosion.

Coverage and revegetation (to stabilize against erosion) are required by local regulatory requirements designed to limit radon emanation and/or ingress of surface water and the formation of acid mine drainage containing pollutants not normally containing radionuclides, depending on the mineralogy of the residues.

TABLE XXIX. NATURALLY OCCURRING RADIONUCLIDE CONCENTRATIONS (Bq/kg) OF MATERIALS IN TIN MINE RELATED PROCESSES

Process	Raw materials		Product		Residues	
	^{226}Ra	^{228}Ra	^{226}Ra	^{228}Ra	^{226}Ra	^{228}Ra
Tin mining	Mining soil		Tin ore concentrates		Amang	
	—	—	50	700	100	100
Amang re-treatment	Amang		Tin ore concentrates		Monazite	
	1	1	50	700	0.2% U	5% Th
					Zircon	
					13 000	2200
					Xenotime	
					71 000	14 000
					Struverite	
					29 000	7700
					Ilmenite	
					700	500
Tin smelting	Tin ore concentrates		Tin metal		Tin slag	
	50	700	30	20	2000	3200
					$PbCl_2$	
					60	40
TiO_2 pigment production	Ilmenite		TiO_2 pigment		Gypsum	
	700	500	<10		100	60
	Ilmenite (Australia)				Scale	
	80	360			75 000	61 000
Synthetic rutile production	Ilmenite		Synthetic rutile		Iron oxide	
	700	500	300	200	700	900
Rare earth production	Monazite				Thorium hydroxide	
	0.2% U	5% Th			0.45% U	15% Th
					Lead cake	
					26 000	—
					Tri-calcium phosphate	
					100	600
Yttrium oxide production	Xenotime		Yttrium oxide		Thorium hydroxide	
	71 000	14 000	Not available		190 000	50 000

7. INDUSTRIAL MINERALS

7.1. PHOSPHATE MINING AND FERTILIZER PRODUCTION

7.1.1. Introduction

NORM containing residues are generated by the mining and processing of phosphate rock (phosphorite) that is processed to phosphoric acid or elemental phosphorus. These two intermediates are then further processed into phosphate fertilizers, detergents, animal feeds, food additives, pesticides and other phosphorous chemicals [174].

Phosphate fertilizers are primarily derived from phosphate rock mined as naturally occurring ores. The principal constituent of phosphate rock (or phosphorite) is the mineral apatite (francolite or carbonate–fluorapatite). The typical phosphate (P_2O_5) concentration of the rock is of the order of 15–30%, with clay, sand, carbonate and other impurities present in varying quantities.

Phosphate in mineable quantities is concentrated by sedimentary, igneous, weathering and biological processes (e.g. guano). Uranium may be incorporated in sedimentary phosphorite ores through ionic substitution into the carbonate–fluorapatitic crystals or by adsorption. Igneous phosphorite contains less uranium, but more thorium. High phosphate contents usually correspond to high uranium contents (50–300 ppm).

Approximately 30 countries produce phosphate rock for use in domestic markets or for exports. The principal countries supplying phosphate rock are North African countries, the USA, China and the Former Soviet Union (FSU). Sedimentary rocks are mostly found in North and West Africa, the USA, China and Australia (amounting to approximately 90% of world production); igneous rocks are found in the Kola Peninsula, FSU, South Africa, China, Finland and South America (notably Brazil). Almost all phosphate rock is mined in open pit mines. Figure 23 shows P_2O_5 production and consumption figures for different regions. Table XXX presents worldwide reserves and annual production of phosphate rock. Radionuclide concentrations in ores are given in Table XXXI.

7.1.2. Basic processes in the phosphate industry

7.1.2.1. Beneficiation

Generally, the starting material for the production of phosphoric acid (see below) is beneficiated phosphate ore, referred to as marketable phosphate rock. During beneficiation, phosphate particles are separated from the rest of

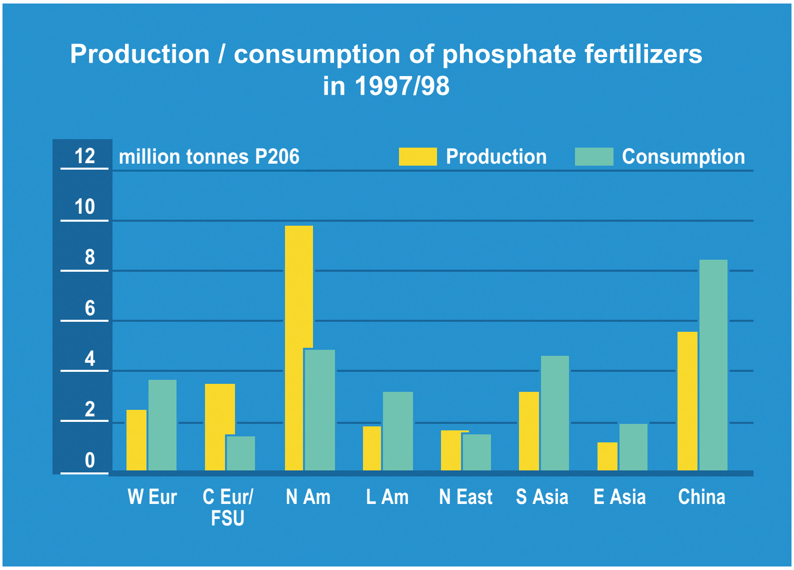

FIG. 23. World annual phosphate (P_2O_5) production and consumption [175].

the ore. Beneficiation can be very simple, just screening or sieving the material and the overburden can be piled up or returned to the mine, or very elaborate, including washing, flotation steps, producing phosphatic clay tailings (clay slime from washer stages of beneficiation) and sand tailings (from flotation stages of beneficiation). Phosphatic clay tailings are stored in large settling areas. Sand tailings are either returned to the mine and used as a backfill in mined-out areas, used for construction of clay tailings retention dams or mixed with clay tailings to increase the solids content of the clay tailings and reduce settling times. In general, beneficiation does not reduce the radionuclide concentration in the ore. Figure 24 provides a flow diagram with an example of the radionuclide balances during beneficiation [181].

7.1.3. Processing of phosphate ore: Acid leaching

Processing of phosphate ore in order to produce intermediate products can be done by acid leaching of the ore resulting in phosphoric acid or by the production of elemental phosphorus by furnacing.

The commercial production of phosphoric acid and of phosphatic application products proceeds from natural phosphatic ore deposits (above all, apatite, $3Ca_3(PO_4)_2{\cdot}CaF_2$, and phosphorite, $3Ca_3(PO_4)_2{\cdot}Ca(OH,F,Cl)_2$),

TABLE XXX. WORLDWIDE RESOURCE BASE AND ANNUAL PRODUCTION OF PHOSPHATE ROCK

(data from IFA 2000)

Country	Annual production (10^3 t P_2O_5)	Geological resource estimates (10^6 t P_2O_5)
Finland	277	42
Total Europe	277	69
FSU (70% from Kola)	4 405	3 197
Canada	125	64
USA	11 419	8 026
Total North America	11 544	8 090
Brazil	1 687	235
Columbia	12	156
Mexico	301	195
Peru	2	450
Venezuela	105	—
Total South America	2 107	1 036
Algeria	262	120
Morocco	6 902	15 750
Senegal	462	57
South Africa	1 051	1 080
Togo	493	36
Tunisia	2 491	480
Zimbabwe	42	—
Total Africa	11 703	17 649
Egypt	317	660
Iraq	90	891
Israel	1 305	260
Jordan	1 811	441
Saudi Arabia	0	1 576
Syrian Arab Republic	656	—
Turkey	0	42
Total Middle East	4 179	4 090
China	5 470	2 850
India	336	3 378
Korea, Rep. of	46	—
Sri Lanka	10	—
Vietnam	243	—
Total Asia	6 447	—

TABLE XXX. (cont.)

Country	Annual production (10^3 t P_2O_5)	Geological resource estimates (10^6 t P_2O_5)
Australia	225	615
Christmas Islands	197	—
Nauru Islands	194	—
New Zealand	0	22
Total Oceania	616	715
Total World	41 278	26 095

TABLE XXXI. CONCENTRATIONS OF MAJOR RADIONUCLIDES FOR DIFFERENT PHOSPHATE ROCKS

Country	Uranium		Thorium		^{226}Ra	^{228}Ra
	(Bq/kg)	(ppm)	(Bq/kg)	(ppm)	(Bq/kg)	(Bq/kg)
FSU	44–90	—	78–92	—	30–70	—
USA [176, 177]	259–3700	62–156	3.7–22.2	—	1540	—
Florida [178]	1500–1900	—	16–59	—	1800	—
South and central Florida [173]	847–1980	—	—	—	881–1980	—
North Florida [173]	241–981	—	—	—	229–884	—
Idaho [178]	1850	—	30	—	300	—
Wyoming [178]	2300	—	10	—	1200	—
Brazil	114–880	27–71	204–753	55–185	330–700	350–1550
Chile [178]	40	—	30	—	40	—
Algeria [178]	1295	—	56	—	1150	—
Morocco [178]	1500–1700	—	10–200	—	1500–1700	—
Senegal [178]	1332	—	67	—	1370	—
South Africa [178]	163–180	—	483–564	—	—	—
South Africa [179]	100–200	—	—	—	—	300–500
Tanzania [178]	5000	—	—	—	5000	—
Togo [178]	1360	—	110	—	1200	—
Tunisia [178]	590	—	92	—	520	—
Egypt [178]	1520	—	26	—	1370	—
Israel [178]	1500–1700	—	—	—	—	—
Jordan [178]	1300–1850	—	—	—	—	—
Australia [180]	15–900	—	5–47	—	28–900	—

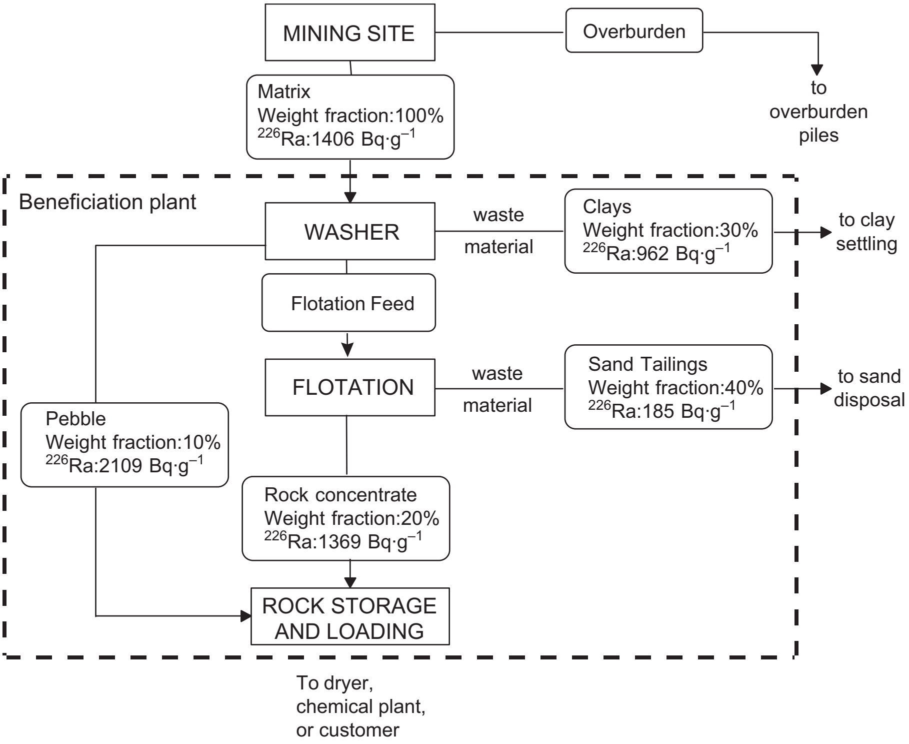

FIG. 24. Simplified flow diagram of mass and radioactivity balances for phosphate mining and beneficiation (after Ref. [181]).

by means of a treatment with strong acids (sulphuric acids, hydrochloric acid and nitric acid). The conversion of apatite takes place according to the following general equation (X = acid):

$$3Ca_3(PO_4)_2{\cdot}CaF_2 + 20\ HX \Leftrightarrow 10\ CaX_2 + 6\ H_3PO_4 + 2HF$$

The phosphoric acid (H_3PO_4) arising from the treatment with acid (HX) is separated and further processed.

7.1.3.1. Material and radionuclide flows following sulphuric acid acidulation

Most acidulation (95%) is done with sulphuric acid, which leads to the formation of gypsum ($CaSO_4{\cdot}xH_2O$), which is not very soluble in the resulting reaction mixture. Solid gypsum crystals precipitate and can be easily separated from the raw phosphoric acid by filtration following a washing step. A neutralization step may be included. Unless the phosphoric acid is used for fertilizer production, it is further purified to the extent needed for its further use. Purification of the phosphoric acid is achieved by solvent extraction. In terms of rounded figures, the production of 1 t of phosphate (P_2O_5) results in the generation of 4–5 t of phosphogypsum [6, 18, 182–185]. A number of variations on this scheme (Fig. 25) are in use, mainly to reach higher yields and cleaner and more concentrated phosphoric acid.

In general, about 80% of the ^{226}Ra, 30% of the ^{232}Th and 14% of the ^{238}U is left in the phosphogypsum. Uranium and thorium become enriched in the fertilizer to about 150% of their original value (70% of the total U and Th budget) [6, 182, 183, 186]. However, the amounts of radioactivity that are fractionated to gypsum vary significantly.

7.1.3.2. Residues from the sulphuric acid process

Phosphogypsum (calcium sulphate) is the principal waste product generated by wet process phosphoric acid production. The phosphogypsum arises as slurry and is typically either deposited in piles or discharged into rivers or the sea [187, 188]. These disposal areas, which are referred to as phosphogypsum stacks, are generally constructed directly on virgin or mined-out land, with little or no prior preparation of the land surface [191]. Each phosphoric

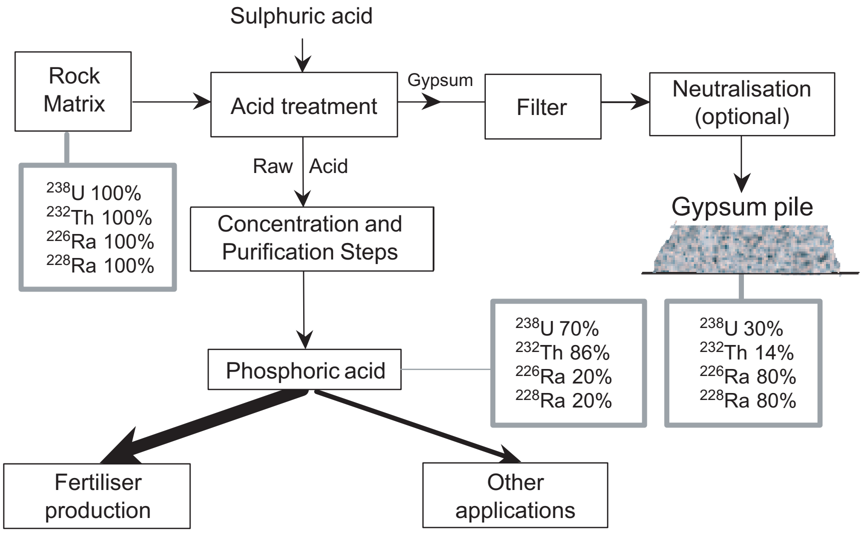

FIG. 25. Flow diagram of the sulphuric acid phosphorus extraction process.

TABLE XXXII. EXAMPLES OF RADIONUCLIDE CONCENTRATIONS IN PHOSPHOGYPSUM (Bq/kg)

Country (source)	^{226}Ra	^{238}U	^{210}Pb	^{210}Po	^{232}Th
USA [183]					
South and central Florida	507–1358	41–366	577–1853	437–1765	11
North Florida	270–599	22–451	348–551	355–566	
Europe [191, 192, 178]	15–1700	500	1300	900	10
South Africa [193]					
Local rock	45–48	64–73	76–132		205–284
Togo rock		17			61
Australia [180]	280–350	10–24	320–440	150–360	4–7

acid production facility may have one or more phosphogypsum stacks. Stacks may be covered, with water covering up to 60% of the top section of the stack [190]. Radium activity concentrations in the gypsum range from background to 1700 Bq/kg (Table XXXII).

Additional waste streams in the phosphoric acid process arise from scale deposited in small quantities in process piping and in filtration receiving tanks, and from filter cloths used to filter the solid gypsum from the acid liquid, which have to be removed regularly because of wear. Worn out pipes and other parts containing scales are also to be removed from the plant. Even though these wastes do not add much compared with the volume of phosphogypsum produced, they involve concentrations up to 1000 times higher (Table XXXIII).

TABLE XXXIII. EXAMPLES OF RADIONUCLIDE CONCENTRATIONS IN SCALES (Bq/kg)

Country (source)	^{226}Ra	^{238}U	^{210}Pb	^{210}Po	^{232}Th
South Africa [195]					
Local rock	70–4470	66–3510			918–11 000
Togo rock	129 000–139 000	0.62			470–2730
USA [191]	333 000				
Europe [182]	370 000–3700 000				
Australia [180]	3–3900		30–1300	5–112	
Brazil [194, 195]	52–889	52–132	45–186		24–189

Wastes of this kind are presently disposed of on gypsum piles along with the gypsum, or in normal landfills in cases where the gypsum is discharged into the sea or rivers [191]. Table XXXIV shows that for a given facility most of the ^{226}Ra originates in the phosphogypsum (98.9% of the total ^{226}Ra inventory). Filter cloths and filter parts contribute only 0.083 and 0.0006%, respectively, to the total ^{226}Ra. Hence, the disposal of these odd wastes does not contribute significantly to the resulting radiation exposure via releases of radioactive materials to surface or groundwaters and/or via atmospheric releases of radon, as long as they are not removed from the pile. In the long term, radiological problems may arise due an unauthorized removal of wastes.

Some countries have elaborate systems to manage these wastes. In the Netherlands, for instance, scales are packed into containers for long term storage at the COVRA facility [35].

7.1.3.3. Material and radionuclide flows in and residues from the hydrochloric acid process

The hydrochloric acid process was developed by the Israel Mining Institute (IMI) from the 1950s onwards [196, 197]. It is generally not in use in the EU, nor in the USA. In this process the ore is treated with hydrochloric acid, bringing both phosphoric acid and calcium chloride into solution. The CaF_2 solids are disposed of together with the $CaCl_2$ waste stream as waste. The material flowsheet for the hydrochloric acid leaching process is comparable to the flowsheet presented in Fig. 24 except for the types of wastes produced and the partitioning of the radionuclides. All the ^{226}Ra in the raw phosphate is released with the $CaCl/F_2$ effluent and most of it will leave the site.

At Tessenderlo Chemie in Belgium, the hydrochloric acid process is used for the production of dicalciumphosphate, which is predominantly used as an

TABLE XXXIV. DISTRIBUTION OF ^{26}Ra IN MATERIALS DEPOSITED IN PHOSPHOGYPSUM PILES AT IMC FERTILIZER, INC., MULBERRY, FLORIDA [191]

Material	Annual quantity	^{226}Ra flux (GBq/a)	^{226}Ra concentration (Bq/g)
Phosphogypsum	8 250 000 t	7922	0.96
Miscellaneous scale	2500 t	83.2	33.3
Filter cloths	1170 cloths	6.7	—
Pipe/equipment	20 t	0.67	33.3
Filter parts	3 pans	0.044	—

additive in animal fodder [198]. During the phosphorus extraction, $CaCl_2$ and CaF_2 salts are produced as by-products, containing most of the radionuclides. In the first phase of the phosphorus extraction process, the formation of monocalciumphosphate, CaF_2 is formed, which is then separated, filtered and disposed of as waste. About 0.5 t CaF_2 is formed per tonne of treated P_2O_5. In the second phase of the process the monocalciumphosphate is precipitated as dicalciumphosphate and filtered off.

The $CaCl_2$ solution (the filtrate) is always discharged in the surface water because dried calcium chloride cannot be disposed of due to its high efficiency in attracting water from air. Contact with even the smallest amount of rainwater would have the effect of washing away the pile very quickly. The most commonly applied treatment step prior to discharge of the calcium chloride solution is its neutralization by addition of calcium carbonate. In that step most of the toxic metals in the ore form insoluble hydroxide/carbonate sludges that can be removed by allowing the solution to settle. This waste stream has to be disposed of as toxic solid waste with a high solubility under acidic conditions. The slurry containing heavy metals and 2000–4000 Bq/kg of ^{226}Ra is disposed of. Also the ^{238}U released from the ore will be precipitated by the addition of lime and buildup of CaF_2 on the dump site, resulting in uranium levels ranging from 3600 to 6000 Bq/kg in fluorophosphate precipitates [182]. The wastewater contains after this neutralization step predominantly $CaCl_2$, which remains in solution and is disposed of in the surface water. The $CaCl_2$ solution still contains ≈20 Bq/L of ^{226}Ra. Another possible step prior to discharge is the specific removal of radium. The addition of barium sulphate or barium chloride to the calcium chloride solution allows for the separation of a co-precipitate $(Ra,Ba)SO_4$. This can be removed by filtering, and the separate crystals can be disposed of. Owing to the fact that the radium mass in solution is very low (less than 1 ppb) the process is very effective but leaves a relatively low volume waste with a very high radon production requiring safe long term disposal [182].

Some additional wastes are to be expected from the extraction liquids used in the extraction process, the elevated metal corrosion due to the use of hydrochloric acid and contaminated piping and metal service parts. Scales are to be expected in relatively low amounts compared with the sulphuric acid process. For the other waste streams from this process no detailed description of the volumes and masses involved is available, nor information on the distribution of radioactivity among the different streams.

The CaF_2 is disposed of in stacks which are generally constructed directly on virgin or mined-out land with little or no prior preparation of the land surface.

7.1.3.4. Material and radionuclide flows in and residues from the nitric acid process

The nitric acid process was developed by BASF (Ludwigshafen, Germany) and has only very recently come into operation. The ore is treated with nitric acid and minor soluble salts are washed out. Further treatment with ammonia for neutralization and carbon dioxide to precipitate calcium carbonate yields ammonium phosphate and ammonium nitrate, which are directly used for fertilizer production. The solid carbonate formed can be used to again produce the carbon dioxide used in the process, leaving calcium oxide as residue. This can principally be used in cement production.

The main waste stream in the nitric acid process is the calcium oxide leaving the process. No detailed analysis of the radioactivity is available so far, as the process has only come into use very recently. Uranium is also expected in the remaining calcium oxide, whereas other contaminants can be expected to precipitate during the neutralization step with ammonia (as insoluble hydroxides). If there is no special removal of radium, all the radium entering the process is to be expected in the calcium oxide residue leaving the process.

Calcium oxide is not deposited due to its solubility in water (after it has reacted with water) and due to the extremely high pH value (11–12) of the resulting leachate solution, nor can it be discharged into rivers or seas. It has to be reused, either for mine backfilling purposes or for the production of cement. If the carbon dioxide used in the process is not made from calcium carbonate but comes from other sources, the remaining calcium carbonate can be easily disposed of. Calcium carbonate is even more stable and less leached under normal environmental conditions than gypsum.

7.1.4. Elemental phosphorus production with the thermal process

7.1.4.1. Material flows during the thermal process

Phosphate rock can be melted in a furnace (1400°C) with sand, iron oxide and coal for the production of elemental phosphorus. Phosphorus and carbon monoxide, driven off as gases, are vented out of the furnace via a stack. Furnace off-gases pass through dust collectors and then through water spray condensers, where the phosphorus is condensed and trapped in water. The residual solids in the furnace contain ferrophosphorus and calcium silicate ($CaSiO_3$), also known as slag. Slag is the principal waste by-product of elemental phosphorus, and ferrophosphorus is considered to be a by-product. The latter is used in specialty markets, such as speciality steels, cast alloys and paint pigments [199]. The slag holds nearly all the uranium, thorium and radium and some of the lead isotopes, while part of the lead and polonium isotopes leave the oven in the gas stream due to the elevated temperatures. The electric dust filter separates part of the radionuclide content from the gas stream, while polonium leaves the process mainly with the off-gas. A flow scheme of the furnace process is given in Fig. 26.

7.1.4.2. Radionuclide flows and contents in residues from the thermal process

The quantity of the slag amounts to 85% of the raw phosphate ore and contains the major fraction (93%) of ^{238}U and ^{226}Ra [6, 182, 183, 186]. The slags find use as substitutes for valuable raw materials, for instance as road building aggregate, ballast in concrete and in wallboards.

The quantity of dust produced with thermal processing is less than 1% of the raw phosphate ore and contains levels of ^{210}Pb and ^{210}Po of the order of 1000 Bq/kg [200]. Lead-210 and ^{210}Po can be immobilized with cement.

According to Baetslé [182], the ^{226}Ra concentration in the slag is 1300–1500 Bq/kg and the ^{238}U concentration is 1100 Bq/kg. Higher concentrations were found in slag at a US plant. A flow diagram on the radioactivity balances in that plant is shown in Fig. 27. Table XXXV presents some data on radionuclide concentrations in different residues from the furnace process.

7.1.4.3. Residue management

The general management practice for slag at an elemental phosphorus production plant is storage or disposal in waste piles. The separated stack dust

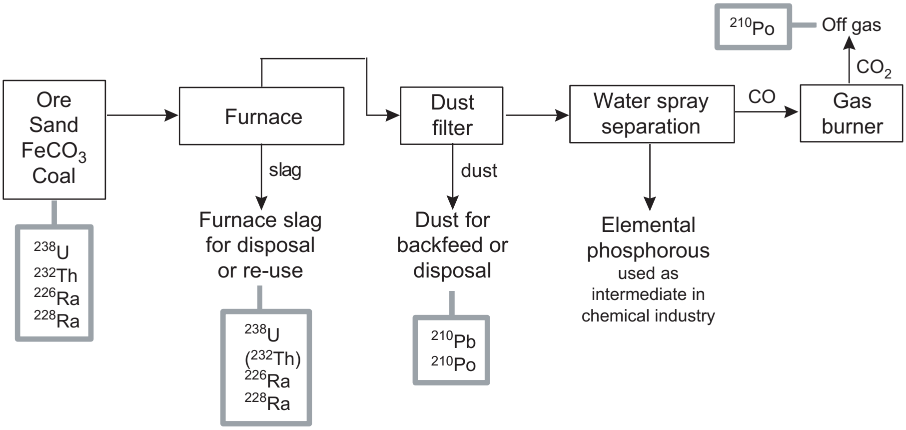

FIG. 26. Flow diagram of thermal production of elemental phosphorus.

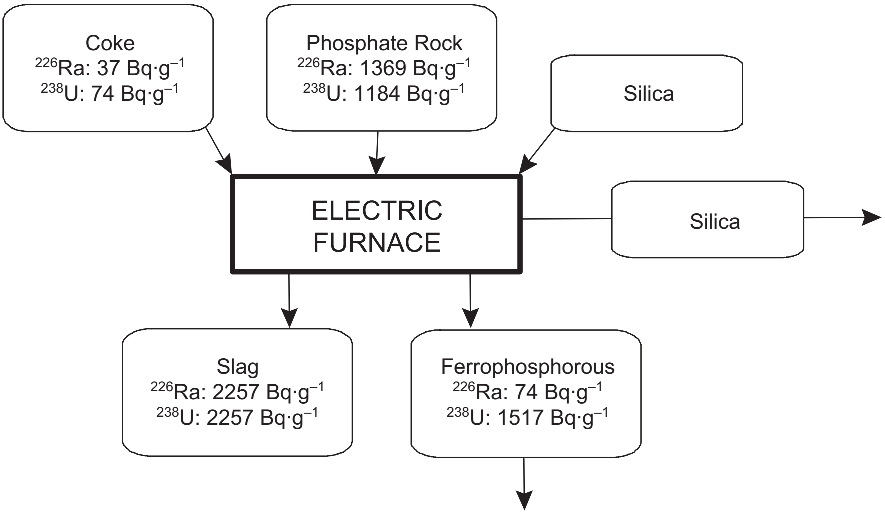

FIG. 27. Flow diagram and radioactivity balances for elemental phosphorous production in a US plant [186].

may be backfed into the oven (re-feed of the contaminants to the slag) or has to be disposed of with the slag. The ^{210}Po and ^{210}Pb in the dust can be immobilized with cement.

7.1.5. Environmental impact of residues

The environmental impact from (phosphate) fertilizer production has recently been summarized in a UNEP/UNIDO report [207]. The release of

TABLE XXXV. RADIONUCLIDE CONCENTRATION IN VARIOUS RESIDUES FROM THE FURNACE PROCESS (Bq/kg)

Country (source)	^{226}Ra	^{238}U	^{210}Pb	^{210}Po	^{232}Th
Furnace slag:					
USA [201–206]	407–1517	444–2072	56	31	9–41
Europe [191, 192, 200]	1000	1000–1500	—	—	—
Ferrophosphorus:					
USA [201–206]	7–44	359–407	27–144	37	3
Calcined dust:					
Europe [200]	—	—	1000	—	—

radium ions into leachate and surface run-off from phosphogypsum piles is mainly determined by the solubility of the gypsum, which is generally very low. The dose resulting from ingestion of contaminated drinking water or food products after contamination of surface or groundwater is generally of no concern even for a member of a critical group living close to the pile. Exposure due to dust inhalation and radon is generally low due to the predominantly liquid stage of the gypsum stack. Furthermore, gypsum hardens out when dry. Intrusion may result in high exposure, predominantly due to radon, and external radiation (gamma radiation levels up to 0.3 μSv/h at the stack [178, 181]).

The river dumping of gypsum as was formerly carried out in Huelva, Spain [188], may result in the contamination of the river borders and increased exposure to the people living close by, mainly due to external exposure and exposure to radon.

The problems linked with the disposal of CaF_2 sludge are comparable with the potential exposure problems linked with the gypsum deposits, except that the former also contains uranium and may contain higher levels of radium if $BaSO_4$ is used for radium recovery from $CaCl_2$ liquid waste. Doses to a critical group living close to a CaF_2 dump reached a maximum of 0.5 mSv/a and were mainly due to radon [6]. The surface water disposal of the $CaCl_2$, if no $BaSO_4$ radium recovery step is included, may result in an important contamination of river sediments and borders or alluvial areas.

The furnace slag is a glassy material containing the radioactivity in a vitrified matrix, resulting in limited leaching of radionuclides. In addition, the slag has a high carbonate content, which also reduces radionuclide solubility [208]. However, USEPA's Report to Congress on Special Wastes from Mineral Processing [209] documents groundwater contamination at several phosphate slag waste management sites. Therefore, despite its properties, slag may still be susceptible to significant leaching, depending on weathering conditions.

Following land pressure, former phosphate mines and historic phosphogypsum dumps and CaF_2 heaps may be reallocated for industrial or domestic use [200].

7.1.6. Concentrations of radionuclides in fertilizers

The most traditional phosphate fertilizer, the production of which is gradually being phased out, consists of adding H_2SO_4 or H_3PO_4 to phosphate rock until the formation of $Ca(H_2PO_4)_2$. This product is then used as such as fertilizer. This type of fertilizer contains variable amounts of U–Ra–Th depending on the acid used for acidulating the natural phosphate ore. For the production of normal superphosphate, high grade phosphate ore (≈33% P_2O_5)

is required for the production of a product containing 20% available P_2O_5 after acidulation of the rock with sulphuric acid. In triple superphosphate (TSP), phosphoric acid is used as an acidulating agent. The uranium content in the TSP compared with that in the initial phosphate rock depends on the uranium concentration in the acid used to produce the TSP. Dicalcium phosphate and diammonium phosphate are produced from wet phosphoric acid by neutralization with lime and ammoniac.

The radioactivity content of fertilizers is highly variable and depends on the radionuclide content of the marketable phosphate ore and on the method of production. Table XXXVI shows some values reported by Baetslé [182], extracted from a number of references. Moreover, several liquid extraction processes have been developed to recover uranium from H_3PO_4, resulting in uranium free fertilizer [182].

Fertilizers are generally depleted in ^{226}Ra content and display a uranium concentration pattern which is the result of either the dilution or the concentration effects of the fertilizer production process on the initial phosphate concentration in the ore. Being a natural product at the onset, fertilizers are not considered radioactive materials.

Although most of the uranium and thorium remain in the fertilizer, fertilizer application does not contribute to the general dose (max. 2 μSv/a for an individual [182]). Commercial calcium phosphates from furnace grade phosphoric acid do not contain any of the radionuclides present in natural phosphate.

Work carried out in Finland has shown that the annual contribution of ^{238}U contained in NPK fertilizers was about 0.25% of the total uranium

TABLE XXXVI. RADIOACTIVITY LEVELS (Bq/kg) IN PHOSPHATE PRODUCTS [178, 182]

Product	^{238}U	^{226}Ra	^{232}Th
Phosphoric acid	1200–1500	300	—
Normal superphosphate	520–1100	110*–960	15–44
Triple superphosphate	800–2160	230*–800	44–48
Mono-ammonium phosphate	2000	20	63
Diammonium phosphate	2300	210	<15
Dicalcium phosphate	—	740	<37
PK fertilizers	410	370	<15
NP fertilizers	920	310	<30
NPK fertilizers	440–470	210–270	<15

* Low value for FSU countries.

naturally occurring in the upper 10 cm of the soil. Similar results have been reported in Belgium, where it was found that ^{226}Ra contamination in phosphate fertilizers added to the soil represents only 0.25% for 14 kg/ha of phosphorus and 0.96% for 54 kg/ha of phosphorus of the total ^{226}Ra in the upper 20 cm layer of the soil. The long term effects of phosphate fertilization on radioactivity have been studied in the USA. TSP produced from Florida phosphate rock and applied at the rate of 30 kg/ha of phosphorus during more than 50 years did not change the concentrations of U, Th and Ra in corn leaves and grain, wheat grain and straw, or soybean leaves and beans, in comparison with non-fertilized plots [210].

7.1.7. Utilization of by-products and residues

7.1.7.1. Utilization of phosphogypsum

Since there are large quantities of phosphogypsum, the industry encourages its use in order to minimize the disposal problem. Phosphogypsum is used as a fertilizer, soil conditioner and for backfill for road construction, and as a construction material, for example for the production of plasterboard and as a cement aggregate and as road construction material.

In the USA, the primary use of phosphogypsum is in agricultural applications (1–2% of phosphogypsum generation), while other uses of phosphogypsum in construction and building materials are now banned [209] due to the resulting radon emanation. Other applications being considered include sulphur recovery from gypsum, introduction of gypsum into municipal landfills to increase biological decomposition, construction of artificial reefs, and conversion of phosphogypsum to calcium carbonate and ammonium sulphate [185].

Radon exhalation (reuse as building material) and the direct radiation (reuse for road construction) may be radiologically relevant. Phosphogypsum is also used in road construction. However, radiation survey measurements made on finished road sections have shown that exposure rates were not discernible from background levels [209, 211].

Some additional information on the use of phosphogypsum in building materials is given in Section 7.2.

7.1.7.2. Slag utilization

Phosphate slag is used as an aggregate in asphalt manufacture and as an aggregate for Portland cement concrete in making construction blocks and driveways. Slag has been incorporated in roofing shingles, septic tank fields and in the manufacture of rock wool insulation. Phosphate slag is also used as railroad ballast and as stabilization material for stockyards [209, 211]. Forty percent of the phosphate slag produced in the USA is used in construction materials.

$CaSiO_3$ slags from the furnace process might be used for road construction. Exposure increases above that of asphalt containing slag are of no concern.

For some additional information on the use of slag in building materials the reader is referred to Section 7.2.

7.1.8. Measures for improvement and remediation

In most EU countries the phosphate industry is not subject to the radiation protection measures common for a number of other industries handling materials with comparable hazards [191]:

- The operation of the facilities does not require radiation protection licenses;
- Effluents from those facilities and the associated radiation doses are not limited to common individual protection levels, and no monitoring requirements are in place;
- Wastes and disposal sites do not require radiation protection licensing for handling and disposal;
- Handling, repair and disposal of equipment used in the phosphate industry does not require licenses, even though doses may be of regulatory concern.

Three major environmental hazards from phosphogypsum waste have to be considered:

(1) The potential for releasing radium and non-radioactive contaminants into the environment;
(2) The radon exhalation into the atmosphere;
(3) The potential reuse of materials due to the loss of institutional control, see, for example, Refs [212, 213].

There are several properties of these wastes that long term management options have to cope with:

- The large volumes of the wastes;

— The long time persistence of the potential hazards due to the long half-lives of the relevant radionuclides;
— The solubility of the wastes in water.

Options for remedial action on phosphogypsum piles can be guided by the state of the art in remediation of wastes from uranium mining and milling [214–217], because the various hazards and requirements are quite similar.

There are processes, involving the concentration of the phosphoric acid and sedimentation, for the production of uranium in the form of yellowcake. However, theses processes are not economically viable at the present price of uranium [218].

7.2. CLAY, CERAMICS AND BUILDING MATERIALS

7.2.1. Introduction

Building materials can contain NORM. This can be of natural origin, that is contained in the raw material (e.g. natural stone), or it can be due to the addition of industrial products (e.g. zircon sand), intermediates or by-products (e.g. coal ash, phosphogypsum and furnace slags). Recycling of industrial residues and wastes has been encouraged for reasons of minimizing resource use and is often favoured by industry since it adds value to a material for which otherwise disposal costs might arise. Concern has arisen, however, over the potential content of radiological and other contaminants in the recycled materials. Owing to the wide variety of raw and recycled materials and their possible NORM content, no general data are presented here. The NORM content in natural resources may also vary considerably over short distances, for example in granite quarries.

The abstraction of sediments, such as sands and gravel, which typically have low radioactivity content, per se does not lead to elevated concentrations in the product or waste stream. However, it could be envisaged that slimes and fines from aggregate plants may be relatively enriched, as typically the NORM resides in these fractions. Quarried rocks, such as granite or tuff, which may be rich in natural radionuclides, can either directly expose the population to NORM, or increase the exposure due to radon release when applied indoors. It is unlikely that quarrying and stone cutting residues are of significant radiological concern. However, very little information is available on this type of scenario. Indeed, the majority of radiological investigations published in the international literature on building materials is concerned with scenarios of residential exposure, i.e. gamma, radon and eventually thoron exposure of the inhabitants. Some studies and estimates are available on the occupational exposures in some particular industrial working environments [171, 219] and for construction work [175], which are, however, outside the scope of the present report.

In the international literature, activity concentrations are not readily available for the raw material used for the production of building material, whereas they are abundant for building material in use, i.e. the final product. As regards recycling of by-products (e.g. coal ash, phosphogypsum and furnace slag) their production is discussed elsewhere in this report, but their possible activity concentrations are also summarized in Table XXXVII and Fig. 28.

Table XXXVII gives an idea of natural radionuclide concentrations in building materials as final products. The table was constructed using a recent compilation of data from European Union member and non-member states [253] as well as a range of European and international papers. This range can give an idea of the 'normal' distribution of these values. It is worth noting that values of activity concentration of the ^{238}U chain in building materials are published generally as ^{226}Ra concentration, but sometimes also as ^{238}U. It is

TABLE XXXVII. ACTIVITY CONCENTRATIONS (Bq/kg) OF NATURAL RADIONUCLIDES IN BUILDING MATERIALS
(compiled from Refs [213, 219–252])

	^{226}Ra	^{232}Th	^{40}K
Concrete	1–250 (5000 ^{238}U)[a]	1–190	5–1570
Aerated and lightweight concrete	9–2200	<1–220	180–1600
Clay (red) bricks	1–200 (590 ^{238}U)[b]	1–200	60–2000
Sand–lime bricks and limestone	6–50 (90 ^{238}U)[c]	1–30 (60)[c]	5–700
Natural building stones	1–500 (870)[d]	1–310 (540)[e]	1–4000
Natural gypsum	<1–70	<1–100	7–280
Cement	7–180	7–240	24–850
Tiles (glazed and unglazed)	30–200	20–200	160–1410
Phosphogypsum (plasterboard)	4–700	1–53	25–120
Blast furnace slag stone and cement	30–120	30–220	—

Notes: Some extreme values have been reported in the literature and are marked: (a), [220]; (b), [236]; (c), [233]; (d), [239]; (e), [242].

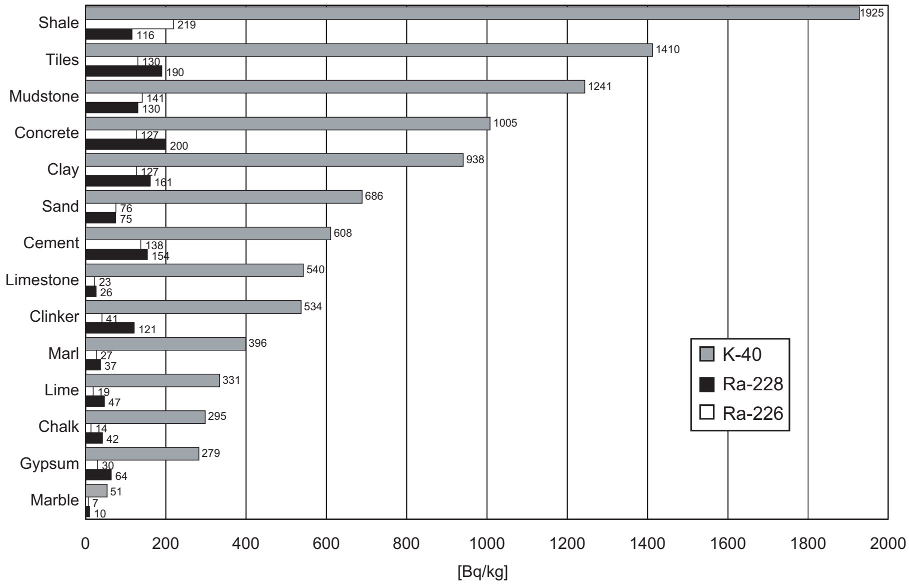

FIG. 28. Radionuclide contents in various Polish building materials (courtesy of J. Skowronek).

worth noting that some materials have higher ^{232}Th than ^{238}U activity concentration [242]. When buildings or other structures such as roads are being demolished, i.e. when the material is converted into rubble, the given activity concentrations can be considered the concentrations in the residues. Table XXXVII does not claim to provide a statistically representative picture of the situation for two reasons: firstly, because very few national representative investigations have been made in different countries; secondly, because analytical efforts are typically focused on samples and materials that are expected to show high activity concentrations.

Aerated or lightweight concretes are produced mainly with the same raw materials as ordinary concrete, but a small quantity of aluminium powder is added to create cell structure in the final product. It is mainly used for non-supporting walls in residential construction. Lime bricks (called also white bricks) are used as building material in house construction only in some European countries [253]. Tuff is consolidated volcanic ash composed largely of fragments (less than 4 mm size) resulting directly from volcanic eruptions [241]. The production and use of alum shale concrete was banned in Sweden in 1975 [220] because its disposal and possible residues of past processing could constitute a serious concern. Use of phosphogypsum in building materials was also banned in some countries, for example Belgium and Romania [224].

More than 280 Mt of coal ash (fly ash and bottom ash combined) are produced annually. About 40 Mt of these are used in the production of bricks and cement [254]. Since most of the process residues further processed into building materials do not meet the required technical specifications, they are typically mixed with pristine raw materials. The net effect is a dilution of the NORM content relative to the process residues.

In 1996 it has been estimated that up to 15% of phosphogypsum was recycled and that within the European Union some 2 Mt were recycled annually [6]. Its activity concentration depends on the origin and the chemical treatment of the raw material: for example, phosphogypsum from phosphate rocks generally contains considerably higher concentrations of ^{226}Ra than gypsum from carbonate rocks. In any case, not only the radium concentration but also the radon exhalation from it can be higher than normal [253].

Zircon sand extraction and milling are discussed above, where an estimate of their activity concentration is given. Zircon flour is used in the ceramic and refractory materials industry, particularly for glazing tiles.

7.2.2. Processing

Very little information is available on the processing of building materials with respect to the possible radiological significance of their residues or of the wastes generated. However, this does not mean that the procedures need not be examined for possible exposure scenarios. For instance, processing of clays (where uranium and other nuclides can be present bound by sorption to clay mineral surfaces) into a variety of ceramics, bricks and similar products can lead to concentration of radionuclides in certain products or residues. Moreover, technical processes, such as grain size separation, floatation and other purification steps, can lead to the accumulation of radioactivity in either wastes, in certain fractions of the feedstocks or in products. Finally, firing and other types of heat treatment typically lead to the volatilization of for example ^{210}Po [219], which then accumulates in dust extraction systems, precipitates on chimney walls or is dispersed around the facilities. Otherwise, concentrations in products and wastes do not differ significantly from those in feed materials.

7.2.3. Residues

Residues can arise as waste from processing (solid and liquid), including substandard products, and the actual building materials in the form of rubble or debris from demolished buildings or dug-up pavements and similar structures.

Operators are frequently unaware of the presence of any radiation hazards and, hence, manage residues without any special precautions. In addition to the relevant necessary occupational or residential radiation protection, which is treated in other IAEA reports, care has to be taken that the NORM cannot be further dispersed in the environment. However, due to the ceramic or glassy nature of the products and residues from many thermal processes, radionuclides bound in them typically have very low release rates for the aquatic exposure pathways. Upswept dust, radon exhalation and direct exposure may be still of concern. On the other hand, materials that do not originate from thermal processes, such as phosphogypsum, tend to have higher release rates for aqueous exposure pathways.

Blast furnace slags are used mainly as aggregate in concrete (crushed) as well as an additive in cement (finely ground). The activity concentration in them depends on the ore type, the origin of the raw material and the metallurgic processes. The use of (coal) fly ash and slags in concrete is a well recognized source of gamma exposure that is due to the presence of activity concentrations of ^{226}Ra, ^{232}Th and, to a lesser extent, ^{40}K, while its effect via radon exhalation is controversial, due to the low emanation coefficient from the ash [255]. Phosphopgypsum and gypsum from flue gas desulphurization used, for example, in the production of plasterboard may give rise to similar concerns.

On the other hand, the use of zircon sand as an opacifier in glazes for tiles does not seem to significantly increase the exposure of the general public to gamma radiation [256, 257] or to exhaled radon, owing to the typically relatively low ^{226}Ra concentrations [257] and low emanation factors from the glaze. However, measurements performed by some Chinese researchers in rooms with and without glazed tiles [256] showed that the average area density of total beta radionuclides in tile glaze was 12–13 times as much as that in ordinary building material. Similar results were obtained in a very recent study from China [258]. This causes an increase in external beta radiation dose received by the public in buildings and, therefore, a possible exposure pathway to the public from residues due to processing or possible waste material.

8. USE OF VARIOUS RADIO-ELEMENTS

8.1. URANIUM

8.1.1. Introduction

For obvious reasons the radiological implications of uranium mining and milling are well established, and in most of the countries concerned some form of regulatory control exists. Therefore, there is no need to discuss this in the present context. However, the working practices in this industry and the handling of the respective residues might be of interest so that attention is drawn to this.

A self-evident source for NORM are the mining and milling activities for the radionuclides uranium and thorium. Owing to the incomplete removal of the target nuclides and due to the selectivity of the milling process for these nuclides rather than their daughter products, milling residues may contain considerable amounts of radioactivity. Below-grade ore and sometimes 'barren' rock may also contain significant amounts of radioactivity and heavy metals, and may have an acid generation potential adding further to the hazard.

The production situation for uranium has been analysed in great detail in the so-called Red Book, jointly produced by the OECD Nuclear Energy Agency and the IAEA [218]. An assessment of the environmental activities around uranium mining and milling on a worldwide basis is provided by two joint OECD-NEA/IAEA publications [120, 259]. The situation is summarized here in Section 8.

The major uses of uranium are fuel for nuclear reactors and in nuclear weapons. There have been other, minor, uses of uranium mainly historical, for example as colouring agents in glasses [260] and glazes for tiles. Owing to the small scale of related operations they are not treated separately here, the major concern being the mining and milling activities.

World uranium resources found in 49 countries of the world have been classified by the IAEA [140] into fifteen major geological deposit categories. Of the 582 entries in an IAEA database used to create a world uranium distribution map, 159 (25.1%) were in Europe, 167 (29%) in North America, 111 (19.2%) in Asia, (of which 78 (12%) were in Central Asia (primarily Kazakhstan and Uzbekistan)), 34 in Australia (6%), 61 (10%) in Africa, 18 (3%) in South America, and 32 (5%) in the Russian Federation.

Estimated cumulative world production of uranium up to 1999, based on information provided in Refs [140, 261], amounted to approximately 1 966 900 t. With these data, it is estimated that 33.1% had come from North

America, 28.2% from Europe, 13.5% from Asia (including the Central Asian states), 17.1% from Africa, 4.8% from the Russian Federation, 3.1% from Australia and 0.2% from South America.

8.1.2. Mining processes

The majority of all past and present world uranium mining has taken place using conventional (underground and open pit) mining methods. The use of in situ teaching (ISL), phosphate or metal ore by-product and mine water recovery combined has averaged since 1994 less than 21% of all production, whereas the once predominant underground mining method has been decreasing relative to open pit mining over the same period [261].

In milling, the ore from the mine is crushed and is then subject to a leaching process. The solvent is usually H_2SO_4, or NaOH for carbonate-rich rocks. After the uranium minerals (typically pitchblende, $UO_{2(s)}$) have been dissolved, the pregnant solution is separated from the residual solids (tails). Typically the solids are washed in several steps with fresh solvent until the desired level of metal value recovery is attained. The dissolved uranium is recovered from the pregnant solution by neutralization, followed by precipitation, non-aqueous solvent extraction or ion exchange. The final marketable product normally is yellowcake [262]. The stripped solution may be replenished and recycled for use within the leaching circuit or as the liquid component in the crushing/grinding operation. Ultimately, the residual solids are neutralized or washed with water prior to being pumped to the tailings pond. This wash serves to recover any remaining solvents and reduce the quantity of acid being placed in the tailings impoundment and thus to stop further leaching reactions. The wash water may be recycled.

ISL combines the mining and part of the milling process. The solvent is injected underground and the pregnant solution is extracted in a specially designed array of boreholes. The array is designed such that outward migration of process solutions into adjacent aquifers is prevented. In some countries, namely Germany and the Czech Republic, a combination of underground mining and ISL was also applied.

8.1.3. Residues

Residues from uranium mining consist of overburden and barren rock materials excavated from underground and open pit mines to provide access to economic deposits of U_3O_8 ore. The spoil material, usually deposited in piles adjacent to the mine, may also consist of gangue and ore that was considered uneconomic (of little or no likely value), sub-economic (ore of lower grade than what was economic to extract at time of mining, often called ‘protore’), as well as drill cuttings from exploration or production drill holes. Many of these materials contain radium, uranium and thorium, and their radioactive decay products, and emit radon gas. The uranium bearing ore is processed at a mill and the depleted ore residue is placed in a mill tailings impoundment. Heap leaching as a milling process will result in a similar class of materials. These materials generally still contain various radionuclides and often (toxic) heavy metals.

ISL does not lead to significant amounts of radionuclide bearing spoils and tailings, but there is a risk of incomplete control over the hydraulics of systems that may lead to the spread of contaminated solutions into adjacent aquifers. Some Member States have separate regulatory classifications for the residues generated by ISL processes.

Given the diversity of depositional environments of uranium ores described above, it is quite likely that the levels of radiation and radionuclide distributions at national sites will be highly variable. Therefore, it is difficult to state the radionuclide concentrations in residues.

On the international level there are a large number of both reclaimed and unreclaimed uranium mining sites [259]. The possibility exists, and has been documented, that members of the public will ignore advice and planning regulations and locate houses on or immediately adjacent to such a mine or mill tailings site. Mill tailings present a hazard in that the sand sized material appears perfect for use as an aggregate for construction. Similarly, their ready availability, without much need for crushing, etc., makes mining residues an attractive building material. When used in road construction the radiological impact may not be of concern, but this changes when the material is used for building purposes. Examples exist from around the world where private homes have been built from such material and then had to be demolished when this was discovered. In cases where mining and milling sites have been redeveloped for industrial or residential purposes, radon emissions from below may be of concern. Section 11 discusses the various radiation exposure pathways that may possibly be found at such settings. The relevant safety aspects are laid out in detail in Safety Guide WS-G-1.2 [263].

The management of uranium mining and milling residues typically involves isolation by capping in order to reduce radon exhalation and to minimize acid generation by infiltration of atmospheric water. Relocation into underground mines or mined-out pits is typically a good option for current operations, but often difficult and expensive in retrospect.

8.2. RADIUM

Certain natural radionuclides, such as radium, have been in use for decades outside the nuclear industry for their special properties. The use of sealed radium sources, which found widespread use in clinical chemotherapy and materials sciences, is of particular significance, though outside the scope of this report. The IAEA has developed an extensive programme for making such sources safe through conditioning and proper disposal, which is still ongoing [264].

People have also deliberately exposed themselves to radium in, for example, spas over several decades (see Ref. [265] for a selection of links). A selection of data and a discussion of exposure scenarios is given in Ref. [92]. However, not much information appears to be available on the downstream fate of the radoactivity discharged from spas.

In the context of the present report only contamination of the environment by the production and disposal of either production wastes or of the final products after the end of their useful life is considered.

For several decades, until the late 1960s, radium was used as a luminizing agent on watch and clock dials and arms, and on car, aircraft and shipboard instruments. Of concern here are obsolete operations to extract radium from ores and its further processing, factories and other establishments performing the luminizing operations, and scrap yards handling large numbers of instruments. The latter became a localized problem after both world wars at defence establishments that decommissioned military hardware. Both during and after World War II, there were many examples of informal in situ disposal on defence sites of defective or surplus equipment, including luminized components [266]. At various defence establishments in the UK, for instance, dials were burnt in the open to recover the metal scrap by processes loosely termed ‘bash, burn and bury’. The residual ashes, containing the radium, were spread on fields, requiring remediation later [267]. However, it is believed that, as a contaminant, buried radium from luminizing processes is largely immobile [266].

About half of all radium extracted worldwide was handled at a factory in Olen, Belgium. The resulting contamination and remedial activities following the closure of the factory and its waste management facilities has already been discussed in detail elsewhere, for example, Ref. [268].

8.3. THORIUM

The thorium content of monazite produced, around 1500 t/a, greatly exceeds the demand for thorium. Large stocks of thorium bearing residues exist. The largest reserves of thorium are in monazite in India, the USA, Australia, Norway and the Russian Federation, and also, not yet exploited, in Canada in the form of uranothorianite.

A sample of its current applications are [269]:

- Combined with magnesium alloys and in tungsten filaments for light bulbs and electronic tubes.
- Adding thorium makes ceramic items, such as laboratory crucibles, more heat resistant.
- Makes carbon arc light lamps burn brighter.
- Added to bulb filaments to help sunlamps mimic the light emitted from the sun.
- Makes tungsten welding electrodes burn hotter.
- Forms strong but less brittle (magnesium) alloys.
- Added as thoria (thorium oxide (ThO_2)) to help gas mantles burn hotter and brighter.
- Added to make high refractive index glass, camera and binocular lenses [270].
- Used as a catalyst for the oxidation of ammonia to nitric acid and other industrial chemical reactions.
- Used as a breeder reactor fuel.

Possible future growth can be seen in the two areas of gas mantles and nuclear energy. Biogas production in developing countries is growing slowly. Extensive use for lighting as well as cooking could lead to significant new demand for mantles and for thorium. On the other hand, yttrium can be substituted for this use, leading to less radioactive waste to be managed. Thorium has become an important alloy for aluminium in high-tech applications, such as structural parts in aircraft. The cutting, turning, milling and drilling processes produce swarf that has to be either recycled or disposed of.

Thorium can also be used as a nuclear fuel in breeder reactors and in closed fuel cycles. Although research had been conducted for over 30 years, these systems have not yet reached full commercialization. This fuel cycle, however, may provide the only practicable way of disposing of the large stocks of ^{239}Pu. In the longer term, therefore, thorium reactors may be built for this purpose, even though they may be less attractive economically as electricity producers.

9. VARIOUS INDUSTRIAL PROCESSES

9.1. SULPHURIC ACID PRODUCTION BY BURNING PYRITES

9.1.1. Introduction

Radiological problems in sulphuric acid production originate from NORM in the raw materials from which the SO_2 is produced. This can be either pyrites specially recovered for acid production or from off-gases generated during the melting of sulphide minerals (e.g. sulphide copper concentrate). Only pyrites as a raw material will be treated here. Other metal sulphides are discussed in the sections on the respective metals.

Pyrites is present in many mineral deposits worldwide. Generally it cannot be recovered economically on its own and is mostly extracted as a by-product or for local use. Pyrites burning is globally used for the production of sulphuric acid. Radiological problems only occur when the pyrites contains enhanced concentrations of NORM. The problem is, for instance, considered to be minor in Sweden [271] due to the selection of low activity raw materials. On the other hand, when the extraction of uranium from the gold bearing ore deposits of the Witwatersrand Basin in South Africa started in the 1950s, the presence of pyrites in the same ore bodies made this method an attractive option for the production of sulphuric acid to leach the uranium. The discussion below mainly relates to radiological problems associated with the latter deposits. This material generally contains nuclide concentrations around 3000–8000 Bq/kg.

Because pyrites is not an internationally traded mineral, information on global production is not available. In South Africa, pyrites burning sulphuric acid production decreased with the uranium production with only two 250 t/a plants and one 500 t/a plant still operational.

9.1.2. Processes

NORM containing pyrites would probably not be mined as a primary product. If extracted as a by-product, the mining method will rather relate to the primary product. Pyrites is extracted from milled ore through a flotation process, which can be either alkaline or acidic. The latter process may, for instance, follow an acidic uranium leaching process on the South African gold ore. After being repulped, the pyrites is injected into fluidized bed roasters, where the sulphur is burned off as SO_2, also producing calcine (Fe_2O_3). The SO_2 gas is next cleaned from the calcine flue dust through dry cyclones, wet scrubbers and electrostatic precipitators. Eventually the clean SO_2 is converted first to SO_3 in a converter with V_2O_5 as a catalyst and subsequently to H_2SO_4 by the progressive concentration of sulphuric acid in absorption towers.

9.1.3. Residues

9.1.3.1. Form and concentrations

The residues from a pyrites extraction process will mainly be the tailings of the primary extraction process. Residues from sulphuric acid production mainly consist of calcine. Pyrites flotation and sulphuric acid production from pyrites may also cause the formation of highly active scales on the walls of vessels and pipes, and on ceramics, plastics and rubber linings of the flotation and sulphuric acid plants. These scales are generated mainly due to the decreasing solubility of radium sulphates with decreasing temperatures and the presence of co-precipitation agents in the process water.

The masses of the pyrites input and the calcine output are normally about the same and around 15–20% higher than the mass of the acid produced.

While alkaline pyrites flotation processes do not cause any enhancement of radionuclides, the acidic process creates scales with enhanced levels of radioactivity (especially ^{226}Ra) on the walls of pipes, vessels and especially flotation cells. While the pyrites from Witwatersrand ore has specific alpha activity concentrations of around (24–64) × 10^3 Bq/kg, alpha activity concentrations within the range (36–86) × 10^4 Bq/kg have been observed in scales collected in flotation plants [272]. Much higher alpha activity concentrations, generally within the range (1–100) × 10^6 Bq/kg, have been observed in scale on the walls of pipes and vessels of, in particular, the gas cleaning sections of sulphuric acid plants. Similar levels, but with some impregnation, were found on rubber linings, polypropylene and ceramic saddles and spirals used to enhance the gas cleaning process and on refractory linings used to isolate surfaces from the effect of high temperatures. While ^{226}Ra dominated in those samples analysed for various nuclides, all the subsequent progeny analyses were around 40–80% of the ^{226}Ra activity concentrations [272]. Calcine should have similar concentrations to pyrites, but enhancement, up to alpha activity concentrations of (5–50) × 10^4 Bq/kg, has been observed in calcine both in some plant sections and in calcine sediments [272].

While pyrites and calcine consist of milled material, contaminated materials can be in a variety of physical forms corresponding to the items mentioned above.

9.1.3.2. Environmental impacts

High levels of occupational exposure were observed in South African sulphuric acid plants, especially during maintenance operations [273]. This requires careful planning and control of such operations. Some plants in South Africa have also been modified to replace scrubber tower walls, containing refractory linings, with stainless steel walls while changing to glass fibre stripper towers. Ceramic inserts, to improve contact surface areas in scrubbers, have also been replaced with polypropylene inserts. With these in place and with improved handling and control measures, occupational exposures were drastically reduced. Detached scales have also caused the contamination of recycling and refurbishing sites.

Lower impacts have been observed where SO_2 was recovered from copper sulphide smelting furnaces.

9.1.3.3. Management of residues

The calcine may be recycled to extract gold or other minerals, which normally become more accessible through the roasting process. The calcine is subsequently mainly discarded with the tailings of the primary extraction process, from which the pyrites was extracted as a by-product, or with the tailings of the mineral extracted with the sulphuric acid produced. Where this is not the case, calcine impoundments or piles are created, which require maintenance similar to that of other tailings impoundments containing elevated levels of NORM. The composition of the material will normally not allow normal vegetation to grow easily on the impoundments. On the other hand, the oxidized state will restrict acid drainage. Limited reuse as a colorant in paving bricks and paints may be possible but needs further investigation when elevated levels of NORM are present.

The scale, refractory material, plastic, rubber and concrete containing high activity concentrations also need special management. In South Africa high activity concentration scales on contaminated surfaces of objects from sulphuric acid plants need aggressive decontamination to allow unrestricted release. Otherwise such objects may be reused at other licensed facilities or are stored. Volume contaminated residues below 10^6 Bq/kg can be diluted into licensed tailings impoundments following steps to improve amenability if required (e.g. shredding of rubber). Residues above 10^6 Bq/kg are presently retained in interim storage areas (Section 6.6.3.5).

9.2. BURNING OF SHALES TO EXTRACT OIL

In about a hundred places in Sweden there are deposits of burnt alum shale. Alum shale has been used for the extraction of alum, for burning limestone in the field and, for a short period during World War II, as a raw material for oil production. These deposits are in some places very large, several hundred square metres and up to 20 m thick. The gamma radiation above these deposits is about 0.5–1.2 mSv/h. The radon levels in the soil air in these deposits can be very high, up to 2000 kBq/m^3. In some places in Sweden large numbers of residential buildings have been erected on such deposits [274].

9.3. RIVER AND HARBOUR DREDGING

Rivers and estuaries are the recipients of discharges and effluents from industry and geological materials eroded by natural processes. Owing to their geochemical behaviour, many metals, including heavy metals and radionuclides, accumulate in river sediments, typically in the fine grained or organic-rich fractions. In order to ensure unobstructed shipping, sediments are periodically dredged from shipping lanes, harbour basins and canals. The dredged materials are used for civil engineering or land reclamation purposes (e.g. dykes), are landfilled or are disposed of at sea.

The amount of dredged material annually dumped in waters covered by the London Convention (LC) [34] varies between 150 and 400 Mt. Dumping of these materials in internal waters adds another 100–150 Mt annually. Annual fluctuations occur due to the variation in maintenance dredging and new works associated with shipping activities, or with exceptional projects such as the huge dredging activities in recent years in connection with the extension of Hong Kong airport. Probably two thirds of the material is connected with maintenance operations to prevent silting of harbours, rivers and other waterways. Approximately 10% of the dredged material is moderately to heavily contaminated from a variety of sources including shipping, industrial and municipal discharges, and natural erosion of soils and rocks. Dredged material has always had a special position under the LC. About 70% of all dumping permits notified to the Office for the LC in 1972 concerned dredged material. Although the LC prohibits dumping of material exceeding exemption levels, no numerical criteria are available and permits are based on individual risk assessments and expert opinion [274].

Development of sites that have received such dredged material can lead to residential or intrusion exposure scenarios. In addition, dredging bottom sediments, which are typically ‘reduced’, and disposing of them may oxidize ferrous oxides and sulphides, leading to acidification of the material and, hence, release and enhanced migration of radionuclides.

9.4. WASTES FROM THE DECOMMISSIONING OF INDUSTRIAL FACILITIES

Building and other structural materials, for example steel, may have become contaminated during their use with NORM containing materials, residues (e.g. scales, ^{210}Pb/^{210}Po containing soot in exhaust stacks and chimneys) and products or may contain a priori elevated levels of radioactivity. The relevant scenarios leading to the presence of radioactivity have already been discussed in the preceding sections. A common problem is the joint occurrence of hazardous substances (e.g. asbestos, polychlorinated biphenyls (PCBs) and heavy metals) and NORM in decommissioning wastes.

The wastes arising from the decommissioning of industrial and housing structures require separate attention as their contamination may pass undetected. A recent report for the European Commission summarizes the hazardous materials associated with decommissioning and demolition wastes [278]. The implications for the decommissioning of mining and milling facilities and the close-out of respective waste management facilities have been treated extensively in an IAEA technical report [215] and a Safety Standard [276]. The disposal of wastes from the decommissioning of the numerous offshore installations for oil and gas production around the world has already been mentioned. The problem of radiologically contaminated wastes from the decommissioning of fertilizer plants has also been highlighted by a recent UNEP/UNIDO report [207].

Further information on decommissioning and the handling of decommissioning wastes can be found in Refs [277–281].

10. REGULATORY ASPECTS

This section is intended to describe the current situation and state of the debate concerning the regulation of NORM containing materials and to highlight some of the problems encountered.

Differences in national and international NORM regulations and standards appear to be the result of differences in risk management policy rather than differences in underlying scientific information. Radiation originating in NORM containing materials has effects on humans and the environment that are no different from those of any other source of radiation. There exists, however, the problem of distinguishing and accounting for background radiation from the same radionuclides present in the NORM material and in the surrounding environment.

Waste management methods and approved disposal sites appear to vary widely for specific industries, waste forms (sludge, scales, liquids and dusts) and Member States. Statutory and regulatory controls in Member States also vary widely, with some countries having complete schemes of NORM radiation protection and waste management, and other countries having no separate regulatory arrangements whatsoever. One underlying problem is the different national approaches to what constitutes NORM, and consequently which industries, wastes and products are covered by relevant guides and regulations. In other words, there is at present no international consensus on the scope of regulations.

Details of the generic principles and criteria for exclusion, exemption and clearance have been given in a Safety Standard [282].

There is general concern and uncertainty over potential impacts of regulations on international trade and industry. While some believe that a ‘clearance’ of materials would open up the markets for these materials, others fear that an explicit statement that a material contains radioactivity, if only below a specified level, would brand it by implication as radioactive. It is also feared that specification of one or the other numerical value could be used as a political and economic instrument to influence market positions. On the other hand, there is a variety of national and international regulations concerning toxic and hazardous substances, such as heavy metals and certain organic compounds, in commodities and other materials.

Notwithstanding the applicability of general radiation protection principles, owing to the variety in occurrence and possible exposure scenarios for members of the public, deriving allowable doses from radionuclide concentrations appears to be a difficult task. Related to this problem is the question of multiple exposures of members of the public to different sources that in themselves may not pose a concern.

Concerning workplaces, the situation is much clearer. The general principles as laid out in Ref. [282] are applicable and often national regulations do exist. In addition, international guidelines on radiation protection in certain industries, such as oil and gas and uranium and thorium mining, are currently being developed.

11. IMPACTS OF NORM CONTAINING RESIDUES

Abatement and remediation strategies and technologies are usually developed with a view to eliminating certain or all exposure scenarios for the site under consideration. Exposures to radiation originating from NORM may occur at several points during domestic or industrial activities. Direct exposure at the workplace and similar scenarios, however, are not the subject of this report. The main target of remediation measures are liquid and airborne releases that may end up in the environment. Table XXXVIII illustrates the consideration given to the residues from uranium mining and milling. Uranium mining and mill tailings tend to have a very advanced regulatory regime and relatively strict regulatory requirements when compared with other NORM, on both the national and the international level [276]. Owing to concerns regarding potential exposures, it is quite common to have long term land use restrictions applied to uranium mining and milling sites.

Relevant pathways of exposure to radiation originating from NORM in process residues and wastes depend on the mode of disposal, the local environment, the demographics of the population in the region and any institutional measures that may be applied to a given site. Disposal practices may be industry specific and may also depend on the state of the development in the industry and the specific socioeconomic circumstances. The vast amounts of NORM which are generated, however, tend to limit the choices for disposal options. Landfilling or landspreading, surface impoundments and surface mounding (waste piles) are the most common forms of waste management for NORM. Sometimes the wastes are covered, but in many cases the NORM containing wastes are disposed of without cover. The disposal method chosen determines the potential for direct exposure to the NORM and also the potential for surface water and groundwater contamination arising from it.

Table IXL provides a perspective on the relationship between different disposal practices and releases. The table emphasizes the level of isolation provided by different disposal methods. Once a radionuclide is released into the environment, it can also migrate via multiple pathways, thus leading to other types of exposure. The potential exposure pathways also depend on the local environment and demographics. For example, the rural population in Europe has different habits and living conditions from the rural population in sub-Saharan Africa. For instance, the prevalence of manual labour, subsistence farming and homes made of local soil may lead to exposure scenarios quite different from those in industrialized countries. Such differences are important considerations when developing abatement and remediation solutions.

For instance, increased exposures could be expected in tropical developing countries because more time is spent near the home. It is interesting to note that

TABLE XXXVIII. EXPOSURE PATHWAYS FOR MEMBERS OF THE PUBLIC ON OR ADJACENT TO A URANIUM MINE OR A URANIUM MILL TAILINGS SITE

Exposure pathway	Environmental pathway/subpathway	Description	Comments
External radiation	Direct exposure from ground and surface water used for bathing/recreation	Agricultural field adjacent to house, and/or home constructed with mine wastes; open pits collect water used by residents.	Radioactive materials may serve as construction material, located under and adjacent to house. Gamma and alpha radiation
Inhalation	Resuspended dust	Dust entering house or breathed by inhabitants	Alpha exposure to lungs and skin
	Indoor radon		From walls of the basement and the foundation
	Outdoor radon		Ambient levels entering windows
Ingestion, water	Groundwater	Wells may be contaminated by uranium mine waste site plumes	
	Surface water (runoff or pit water)	Bathing and cooking	
Ingestion, plants	Water	Plants watered from pit or wells	
	Soil	Plants grown on waste soils	
Ingestion, meat/milk	Water	Animals drink water from wells or pit lake.	
	Plants Water (fodder)	From well or pit lake	
Ingestion, fish	Surface water (contamination flowing to pond, lake or river)		
Ingestion, soil	Direct contamination		

Note: Many of the these pathways would also be applicable to other types of mining.

accumulation of radon in tropical homes built on or near NORM containing residues may be less of a concern, because the homes are often open and allow free air circulation. Table XL provides some examples of considerations related to exposure scenarios in different regions.

Misuse of NORM containing residues and waste materials, or in other words failure of institutional control, may constitute one of the major causes of human exposure. Owing to their accessibility and sometimes favourable properties, mining and milling residues are often used for building or fill materials. Unless access restrictions are placed on a contaminated area, such human activities are likely to occur and thus warrant consideration when specifying appropriate exposure scenarios. Likewise, partially decommissioned facilities, such as facilities that have been stripped of valuable materials (steel, copper wiring, etc.) but have not been decontaminated, may lead to exposure in a variety of ways: buildings may be re-occupied or contaminated materials may be reused.

Certain waste materials, such as fly ash, slag or phosphogypsum, are being used for civil engineering purposes. While this use may not lead to appreciable exposure, decommissioning or rebuilding such structures and the ensuing dust generation or landfilling of secondary wastes may lead to exposure. The same waste materials are used to make a variety of mainly lightweight construction materials. Lightweight building blocks and plasterboard are typical examples with a potential to result in external exposures.

The failure of institutional control over disposal sites resulting in a failure of the containment may lead to a release of NORM into groundwaters or as airborne dusts. Again, in settings where the nearby population obtains drinking water and foodstuffs predominantly from local sources, this may lead to exposure. Given the long half-lives associated with NORM, institutional controls may need to be maintained for a long time [276].

12. SELECTED MANAGEMENT AND REMEDIATION STRATEGIES AND TECHNOLOGIES

12.1. PRINCIPLES

Two main strategies to mitigate adverse effects on human health and the environment from NORM containing residues can be envisaged:

(1) An ‘end of pipe’ strategy, whereby residues, wastes and effluents are treated or whereby contaminated sites are remediated.

TABLE IXL. CORRELATION BETWEEN DISPOSAL PRACTICES, WASTE FORMS, RELEASE MECHANISMS AND NATURAL ENVIRONMENT

Disposal practice[b]	Waste form	Release mechanism from waste/disposal facility into natural environment[a]							Waste exposed on surface (surface soil)
		Leaching to			Overland runoff to		Volatilization to air	Suspension to air	
		Deep groundwater geosphere	Ground- water	Surface water	Soil	Surface water			
Surface disposal (without cover)									
Waste piles, open mines, farms, uncovered landfills and disposal on ground surface (landspreading)	Solid, stabilized or unstabilized		✓[c]	[d]	✓	✓	✓	✓	✓
Impoundments, slurry lagoons, wastewater ponds, mud pits and evaporation ponds	Aqueous and aqueous slurries		✓[c]	[d]			✓		
Near surface disposal (with cover)[e]									
Landfills and shallow land burial	Unstabilized or stabilized solids		✓[c]				[f]		
Above ground or near surface concrete vaults, lined landfills, near surface tunnels and near surface mined cavities	Special containers and/or enclosed in engineered barriers		[c,g]				[f]		

For footnotes see next page.

TABLE IXL. (cont.)

Disposal practice[b]	Waste form	Release mechanism from waste/disposal facility into natural environment[a]							Waste exposed on surface (surface soil)
		Leaching to			Overland runoff to		Volatilization to air	Suspension to air	
		Deep groundwater geosphere	Ground-water	Surface water	Soil	Surface water			
Geological disposal									
Deep well injection and disposal at abandoned mines	Unstabilized/stabilized solids, aqueous and aqueous slurries	✓							
Mined cavity disposal and deep tunnel disposal	Special containers and/or engineered barriers	[g]							

Notes: These recommendations should be appropriate in most cases; however, some special cases may exist that would require special consideration.

✓ Release mechanism requires consideration in an assessment, if appropriate.

[a] These are the first media to be considered when identifying transport pathways through the natural environment (Table VIII).

[b] Condition at the time of assessment (note, for example, that a covered landfill would need to be treated as uncovered surface disposal after erosion of the cover).

[c] Releases to vadose zone when waste is disposed of above the water table and to aquifer when waste is disposed of below the water table.

[d] Direct release only if waste is in contact with surface water body.

[e] 'With cover' implies that the waste is not exposed at the surface.

[f] Volatilization through a cover may occur in some cases (persistent substances).

[g] Leaching may occur after failure of container and/or engineered barriers; some limited releases may occur while barrier is intact.

TABLE XL. EXAMPLES OF LIFESTYLE DIFFERENCES AND RESULTING EXPOSURES FOR HOMES BUILT ON NORM WASTES

Urban/suburban, temperate developed country	Rural, tropical developing country
Less time at home (work away from home) results in reduced external radiation exposure.	Majority of time near the home, which maximizes external exposure.
More robust home which provides some shielding while inside the home; less air circulation, however, so that radon can accumulate.	Home constructed of soil (may actually enhance external exposure rather than providing shielding). Open air design of home, so that indoor radon concentrations are not significantly enhanced.
Food purchased away from home, only a fraction may be contaminated with NORM.	Subsistence farmer can obtain essentially all food and milk from around the home.

(2) A ‘change the process’ strategy, whereby (industrial) processes and practices are re-engineered so as to avoid or minimize contaminated wastes, effluents, emissions, etc.

Or, in other words, cleanup technology versus clean(er) technology.

The major challenge associated with many NORM containing residues or NORM contaminated sites is the large volumes of material and the typically relatively low specific activities. These properties make conditioning of residues according to practices in nuclear or hazardous waste management costly, difficult or inefficient. Many remediation technologies typically applied to environmental contamination [10] would also be inefficient.

For ongoing operations, residue management methods may need to be adjusted to remove or reduce exposure risks. Retrospective conditioning of historical wastes, in particular those from mining or milling, appears to be not practical in many circumstances, unless an immediate threat to the surrounding population occurs and hence a protective action is justified. However, resources may need to be, and often can be, made available to treat historical wastes associated with ongoing operations.

The main objectives of proper residue management and remedial activities are to protect the environment and the public by preventing dispersal of radionuclides, either through water or as gaseous or dust emissions [9]. Shielding against direct irradiation may also be required. Owing to their physical properties as loose materials and because they are apparently ‘free’, many mining and milling residues may be attractive local sources of building material. Unauthorized reuse of such residues has been observed in both developing and developed countries. Thus, effective passive institutional control to prevent or discourage access to and use of the NORM containing materials may be an important objective of remediation measures.

Remediation is typically preceded by a proper assessment of the site or of the process in question [283]. This assessment begins with the identification and consultation of records, if available. The historical assessment is then confirmed by a scoping physical site characterization, for example by walk-over gamma ray surveys. Detailed sampling and analysis may follow to more clearly identify hot spots and to delineate materials or areas that do not require further attention. In recent years strategies and technologies for efficient site characterization have been developed [284, 285].

Similarly, residue and waste characterization is a basic step in developing effective management techniques. Their physical, chemical and radiological characteristics are determined. Of particular importance is the form in which the radionuclides are present, i.e. whether they form a separate mineral phase, are sorbed, or bound in precipitates or slags, etc. This is assessed carefully in order to evaluate the associated risk and to decide on appropriate management measures. For instance, radionuclides bound in a vitreous matrix, such as slag, have a low probability to become solubilized, hence migration and subsequent uptake in organisms is an unlikely scenario. Similarly, radionuclides may be bound in vitreous components of fly ashes, making water mediated migration unlikely but inhalation of unconsolidated dusts possible.

The concurrent presence of other contaminants, such as heavy metals, arsenic, acid generating minerals and toxic organics, is taken into account when developing a remediation plan (Table XLI). The abatement and remediation

TABLE XLI. NON-RADIOACTIVE HAZARDOUS COMPONENTS IN NORM WASTES
(as quoted in a questionnaire sent to relevant industries in the EU and adjacent countries [134])

Industry/users	Component
Oil and gas	Hg, heavy metals, hydrocarbons
Phosphate	Cd, Zn, Pb, F
Iron and steel	Pb, Zn, Cr, Cd, Cu, As, Hg, Ni
Institute/regulator	Hg, heavy metals, sulphides, organic compounds
Others	Hg, PCBs, asbestos, tungsten

technology is chosen such as to prevent impacts from these contaminants as well as from NORM. That is, it is ascertained that the chosen management strategy and technology is compatible with all the contaminants present and that no detrimental interaction between different contaminants occurs. The presence of such contaminants often imposes additional legal constraints on the acceptable remediation methods and related waste disposal routes. An available disposal site may not be licensed to accept ‘mixed’ wastes.

In addition to technical constraints, there may be a wide range of regulatory and socioeconomic constraints on choosing the appropriate remediation or residue management strategy [286]. Regulations in Member States may prescribe certain techniques and prohibit or discourage others. International agreements may also preclude or restrict other strategies, for instance ‘dilute and disperse’ see, for example, Refs [34, 36]. As has been discussed in great detail elsewhere [6], public reassurance and acceptability can be a major factor for selecting a particular remediation or residue management technique (Table XLII).

TABLE XLII. REMEDIATION TECHNOLOGY AND PUBLIC REASSURANCE
(from Ref. [6])

Remediation technology	Public reassurance
Removal of waste	
Soil excavation	High
Soil scraping	High
Turf harvesting	High
Separation and removal of waste	
Soil washing	Moderate–high
Flotation	Moderate–high
Chemical/solvent extraction	Moderate
Containment	
Capping	Low
Slurry walls	Low
Grout curtains	Low
Immobilization	
Cement based solidification	
Ex situ	Low
In situ	Low
Chemical immobilization	
Ex situ	Low
In situ	Low

Technically feasible remediation and management techniques and strategies have to be evaluated in any one case with respect to their implementability in a given socioeconomic context. As has been discussed in Ref. [28], a variety of non-technical factors control the implementability of remediation measures in practice. For instance, the technique preferred from a technological point of view may not be available in a given Member State, because of lack of infrastructure, skilled personnel or equipment.

The siting of management and disposal facilities for NORM containing residues requires careful assessment of the geological and other environmental properties. The siting and design process is usually controlled by a variety of legislation in Member States and guidance on an international basis [287, 288].

12.2. CLEANUP TECHNOLOGIES

In the following sections a selection of remediation and residue management techniques are presented (Fig. 29) and their applicability to current practices and in a remediation context is discussed. It should be pointed out that these strategies and techniques are not to be seen as mutually exclusive alternatives, but they may need to be applied in support of each other.

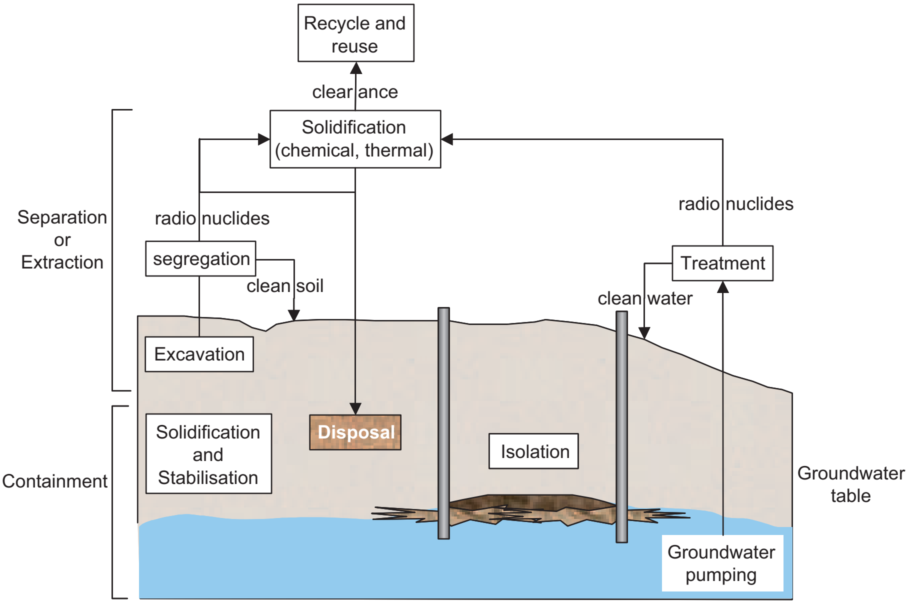

FIG. 29. Classification of remediation techniques by function.

12.2.1. Containment

The objective of containment is to prevent the outward migration of contamination, into either water or air, and to prevent or hinder the inward migration of the transporting agent, i.e. water or air (Fig. 30(b)). Reducing the infiltration of water or air reduces the mass turnover and, hence, release rates and may also reduce the availability of fresh reactant, such as oxygen.

Preventing or discouraging human intrusion can also be an important objective of containment. The objective is typically achieved by providing a low permeability boundary around the waste or the contaminated material, by reducing the permeability of the contaminated material itself (Fig. 30(a)). In addition, groundwater flow can be diverted around the emplaced material by artificially constructed high permeability zones around it (pervious surround). The effectiveness of such measures largely depends on the permeability contrast between the waste material and the surrounding host rock or engineered barriers.

Infiltration of atmospheric precipitation and exhalation of airborne radionuclides, namely radon, are reduced by the application of surface cappings. By hindering the outgassing of radon, it will decay in situ and remove the associated potential exposure. Preventing atmospheric precipitation from infiltrating the body of waste reduces the potential for waterborne releases and migration of contaminants.

In addition to physical barriers, use can be made of chemical or geochemical barriers. Many of the low permeability materials used in the construction of physical barriers, such as clays, also retain contaminants owing to their sorptive properties. Other chemical reactions, such as precipitation,

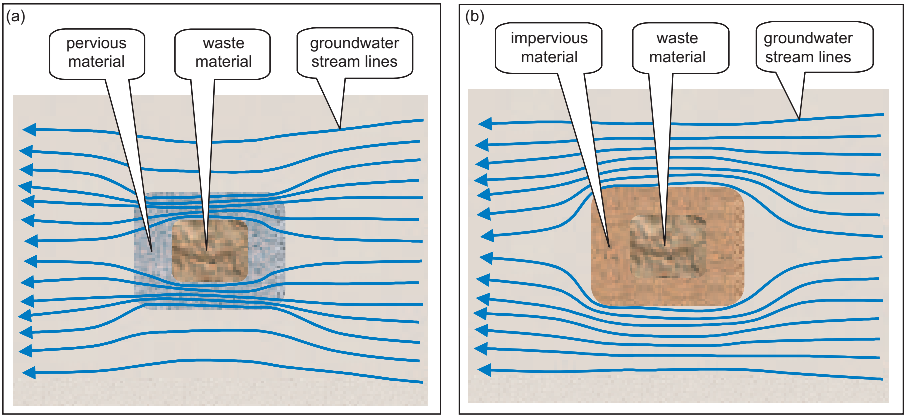

FIG. 30. The concept of (a) pervious surround and (b) containment with impervious barrier.

may also occur. In recent years, reactive chemical barriers have been developed that affect, for example, precipitation of contaminants while being consumed. Certain hydraulic properties and layouts are required to direct the groundwater movement towards the barriers and to bring the contaminants into contact with the reactive material.

Containment can be planned for at the design stage of new waste management facilities or can be put into place retrospectively, that is to say in a remediation context. For backfitting facilities still in operation, there may not be such a clear distinction between the two situations.

Engineered disposal facilities may be constructed above or below ground level and typically consist of a bottom liner with drainage and some form of engineered cap. Bottom liners, which would extend up to the side walls in below ground facilities, are constructed in one or more layers of low permeability geological material, such as clays, and/or synthetic geomembranes. Layers of geotextiles to improve the stress resilience of the liner may be added. The drainage systems typically consist of a layer of coarser geological material and/or synthetic drainage mats and a collecting system of pipes or similar.

The engineered cap may also be made up of several layers: a low permeability layer to prevent infiltration of atmospheric precipitation and the exhalation of radon gas, a (coarser) drainage layer to aid surface run-off, geotextiles to improve the mechanical stability of the capping and, depending on the climatic conditions, a substrate for (re-)vegetation. Intermediate coarse layers may be introduced to hinder burrowing by animals and to discourage human access.

Rehabilitation of underground impoundments may be undertaken along similar engineering lines. However, putting in bottom liners may not be feasible for technical and cost reasons in most NORM related situations. Confining wastes and contamination by slurry walls, injection screens or sheet piling, on the other hand, is common practice. The reverse is also possible: installing a high permeability zone, for example sand filled trenches, around the impoundment will effect a diversion of groundwater flow.

Backfilling of mined-out materials into mine voids may have a number of advantages. First of all, it would remove the material from the surface and thus reduce the amount of land used, i.e. the footprint of the facility. Secondly, if the geochemical conditions of the mined-out material and the host rock can be brought back to near pre-mining conditions, a stable system should arise. Mining, however, severely disturbs the hydrogeology of the surrounding area, creating additional pathways and changing the hydrochemistry, for instance through aeration. Before backfilling any reactive material, an assessment of the likely consequences is required. Certain additional measures, such as sealing of mine openings and fractures, may be needed. The additional measures may also extend to confining the mine by slurry walls or injection screens. These confinement measures may be combined with reactive barriers to condition any drainage waters. Alternatively, special drainage systems to divert groundwaters around the mine may be put in place.

12.2.2. Immobilization

12.2.2.1. Concept

Immobilization, as opposed to containment, is intended to affect the waste material itself. Two basic options can be distinguished: in situ and ex situ treatment. In fact, a number of treatment technologies can be used for both in situ and ex situ treatment, the method of application varying in each case. In situ treatment leads to immobilization without the contaminated material being removed, while ex situ treatment proceeds in some sort of plant, either on-site or off-site. After treatment, the material is either returned to the place of its origin or disposed of in an engineered repository.

The underlying objective of immobilization is to change the waste form into one that is less likely to release any radionuclides contained and/or to reduce the (reactive) surface area.

12.2.2.2. In situ treatment

Two major methods to implement in situ immobilization are available, based on chemical and heat treatment, respectively. Chemical immobilization is based on the injection of a variety of grouts. These grouts can be based inter alia on ordinary Portland cement (OPC), water glass (sodium silicate), gypsum or organic polymers, for example acrylic or epoxy resins. Prices probably increase in this order, but this may vary and depend on the availability of the agent as a waste product from some industrial processes.

The applicability of immobilizing agents via injection depends largely on the hydraulic properties of the contaminated material. OPC and epoxy resins typically have a high viscosity, while water glass and gypsum solutions, or acrylic acid suspensions, can be made up with a viscosity close to that of water.

Heat treatment aims at in situ vitrification, whereby loose sand is fused into a lump containing the contaminants, see for example Ref. [10]. Resistance or inductive heating methods are available. However, the logistics and cost involved make this method less applicable to NORM contamination. Another problem with heat treatment may be the volatilization of ^{210}Po and Hg. This can be overcome, however at additional cost, with the installation of abstraction hoods and exhaust gas scrubbing. The wastes from the scrubbing will require special treatment and proper disposal.

12.2.2.3. Ex situ treatment

A wide variety of agents have been used, or proposed, for the solidification of excavated waste materials. Often the objective is not only to immobilize the contaminants, but to add value to the waste material by converting it into a useful product, for example for construction purposes. The use in general construction as a substitute for valuable raw materials requires special testing and licensing procedures to ensure environmental compatibility and compliance with quality criteria, such as compressive strength, freeze–thaw cycle stability and leacheability. Solidified wastes may also be used in the construction of, for example, cappings for (hazardous waste) landfills, or road bases. If only small volumes arise, the material may be combined with material from other waste streams requiring a similar immobilization treatment. Combining waste streams can make the process more viable economically as products in marketable quantities are produced.

The treatment may be undertaken on-site or off-site at dedicated facilities. For off-site treatment, the material has to comply with the applicable transport regulations and has to meet the appropriate safety criteria when being handled.

As for in situ immobilization, possible (cold bonding) solidification agents include OPC and other hydraulic binders, such as certain fly ashes, gypsum, silicate gels (water glass) and organic polymers, particularly acrylic and epoxy resins. The overall cost of treatment obviously depends on the amounts or volumes requiring treatment. Some binders are more economic in larger quantities than others. Organic polymers, in most cases, appear to be applicable only to smaller volumes of material. It should also be noted that the long term stability of organic binders is largely unknown.

The conversion of contaminated gypsum ($CaSO_4{\cdot}2H_2O$) slurries, for example from flue gas desulphurization, into semi-hydrate/plaster of Paris ($CaSO_4{\cdot}\frac{1}{2}H_2O$) requires heat treatment in a kiln. It is only the plaster that exhibits the necessary curing properties. It must be remembered that heat treatment of contaminated gypsum may lead to the volatilization of ^{210}Pb or ^{210}Po, if present, as can be the case in flue gas desulphurization residues.

An important conclusion from a recent report [133] on the European perspective of NORM waste treatment was that “immobilization is not widely used or accepted as a treatment. Many companies regard this type of technology as less feasible for the NORM waste material itself. Hence, they do not pursue the development of immobilization techniques as a waste treatment process of its own. However, for treating the radioactive remainder of a separation step, immobilization is widely seen as a treatment with a high potential.”.

12.2.3. Dilution/dispersion

Many NORM containing geological materials have been dispersed in the environment since the formation of our planet, and continue to be by the natural processes of erosion. Therefore, release of residues with unaltered concentrations of NORM and unaltered forms of binding is unlikely to have an environmental impact. Increased erosion rates and hence sediment loads and sedimentation rates, however, do have an environmental impact, but this is not related to the NORM content as such. Changed binding forms of NORM that facilitate its release from the mineral matrix can have an impact.

Dilution and dispersion into surface waters has been a waste disposal option commonly practiced for centuries. Its effectiveness depends on the assurance that the contaminants remain dispersed. It has been observed, however, for a number of contaminants, notably lipophilic organic compounds, mercury and certain metal–organic compounds, that they reconcentrate and accumulate in some environmental compartments, including body tissue. The problem is aggravated by accumulating effects along the food chain. Another uncertainty that to date remains unresolved is the potential effect of prolonged exposure to very low concentrations. In the light of these concerns, discharges and releases have been prohibited (declared radioactive waste) or significantly curtailed in some regions of the world through international agreements, for example the London convention [34] and agreements under OSPARCOM [36]. The dumping of 1.5–7 Mt annually of inert geological materials such as mine tailings is reported to the International Maritime Organization [289].

A generic study on the environmental impact of well injection of oil and gas industry derived NORM wastes concluded that this impact would be below regulatory concern in the USA [290], and there are commercial companies offering such services. Injection of radioactive liquids into deep boreholes has been practised in the FSU countries for many years.

The re-use of conditioned residuals can be considered under some circumstances as ‘dilute and disperse’. Down-blending to reduce the specific activity in combination with immobilization would reduce the potential radiological risk from direct exposure or ingestion. However, in some Member States down-blending to achieve clearance or exemption levels is forbidden.

Landspreading of sewage treatment sludges had been practised for many decades around the world. However, the resulting accumulation of heavy metals in soils became a concern in some countries and this practice was terminated. It can be expected that radionuclides would behave similarly under such conditions. However, landspreading of scales and sludges originating in the oil and gas industry is a practice that is allowed in the USA in a few states and in some other countries, but is not generally accepted. The main objective is to dilute the material and to foster degradation of hydrocarbons. A recent study [290] addressed the radiological impact depending on the radon concentrations for a variety of industrial, residential and agricultural scenarios. The main recommendation from the study was for institutional controls to limit exposures over the long term.

12.2.4. Natural attenuation

Natural attenuation is akin to the dilute/disperse concept in that it relies on geochemical mechanisms to retain the radionuclides in a particular environmental compartment. The concept of natural attenuation or monitored natural attenuation has received quite some attention in recent years in the context of environmental remediation, but is by no means new. It forms an integral feature of the current design criteria for geological and other repositories that rely on the geosphere to prevent or delay radionuclides from reaching the biosphere.

Owing to the changes with time in any natural system and to our incomplete understanding of natural systems in most cases, the use of natural attenuation requires monitoring of its effectiveness. Hence, the general reference to monitored natural attenuation.

The concept of natural attenuation relies on the incorporation of the contaminant within the environment, rather than isolation from it. The mechanisms involved are sorption, precipitation and dilution/dispersion. While dilution/dispersion in ground and surface waters may not be a (politically) acceptable method as such, its macroscopic effects are often difficult to distinguish from sorption or precipitation. (Deep) plowing or soil mixing can be an efficient method for dilution in soils and in particular for reduction of direct surface gamma exposures [177]. It would also reduce the potential for unwanted plant uptake if the soil concentrations are reduced in the root zone of agricultural plants. In all cases the concentrations arriving at a given point in space at a given time are less than those that would be observed without such mechanisms working. In the case of radionuclides the radioactive decay has an additional effect. Often the arrival of a radionuclide can be delayed for so long that it will have decayed to insignificant activity levels. The decay into daughters that are of concern, however, has to be taken into account. Table XLIII indicates the time frame for various decay chains of potential interest.

TABLE XLIII. RADIONUCLIDE DECAY CHAINS TO BE CONSIDERED IN ASSESSMENTS OF NATURAL ATTENUATION

^{238}U series			^{235}U series		
Nuclide	Half-life	Radiation	Nuclide	Half-life	Radiation
^{238}U	**4.468×10^9 a**	**Alpha**	**^{235}U**	**703.8×10^6 a**	**Alpha**
^{234}Th	24.1 d	Beta	^{231}Th	25.52 h	Beta
^{234m}Pa	1.17 min	Beta	**^{231}Pa**	**32 760 a**	**Alpha**
^{234}U	**244 500 a**	**Alpha**	**^{227}Ac**	**21.773 a**	**Beta**
^{230}Th	**77 000 a**	**Alpha**	^{227}Th	18.718 d	Alpha
^{26}Ra	**1600 a**	**Alpha**	^{223}Ra	11.434 d	Alpha
^{222}Rn	3.8235 d	Alpha	^{219}Rn	3.96 s	Alpha
^{218}Po	3.05 min	Alpha	^{215}Po	778 µs	Alpha
^{214}Pb	26.8 min	Beta	^{211}Pb	36.1 min	Beta
^{214}Bi	19.9 min	Beta	^{211}Bi	2.13 min	Alpha
^{214}Po	63.7 µs	Alpha	^{207}Tl	4.77 min	Beta
^{210}Pb	**22.26 a**	**Beta**	**^{207}Pb**	**Stable**	**—**
^{210}Bi	5.013 d	Beta			
^{210}Po	138.378 d	Alpha			
^{206}Pb	**Stable**	**—**			

Note: Nuclides with half-lives of more than a year are marked in bold.

The decision on the application of monitored natural attenuation as management strategy depends on an array of scientific, economic and political criteria and should be based on a risk analysis for the specific site [291]. To evaluate the potential applicability of monitored natural attenuation as an environmentally sound solution, it is necessary to make use of a technical protocol to confirm and quantify its efficacy. In particular, methods such as deep plowing have to be evaluated carefully in order to assess potential migration pathways for the radionuclides concerned. Before a backdrop of scarce resources, various initiatives to promote the acceptance of natural attenuation as part of a cost effective and environmentally sound solution for contaminated sites on the basis of a risk based application of monitored natural attenuation are being undertaken (see, e.g., Ref. [292]).

The concept of (monitored) natural attenuation and its applicability in a remediation context is discussed in more detail in a forthcoming IAEA technical publication [13].

12.2.5. Separation

The objective of separating NORM from wastes is to minimize the volume of waste requiring management and disposal in an engineered facility or a similar option. This applies to current practices as well as an intervention in the remediation context. In this sense, separation is both an ‘end of pipe’ and a ‘change of process’ strategy. Reworking of legacy residues can have the added incentive of recovering mineral value, or conversely the residue volume reduction is a beneficial side effect of processes to recover mineral value [164].

On the basis of a workshop and of a questionnaire sent to the EU and other countries, it was reported [137] that “separation is being used or studied by almost all companies which are already dealing with NORM wastes. There is a need for more development of immobilization techniques for the remainder of separation. Furthermore, several new separation techniques are currently under development by industries that were recently confronted with the NORM problem.”.

Melting of scrap metal can be used to separate the radionuclides [134], which accumulate either in the slags or in the flue gas dusts.

In essence, the treatment of effluents and other waters amounts to separation, and a lower volume of waste to be managed is achieved. Water treatment issues are discussed in Section 12.2.6.

12.2.6. Treatment of waters and effluents

The main effect from any water treatment is the concentration of radionuclides into a residue. These residues have to be brought into a waste form that is amenable to further handling and eventual disposal. The increase in specific activity may lead to increases in doses to operators and other personnel. Hence appropriate precautions may have to be taken.

The implications from the treatment of drinking and industrial process waters, including those used for geothermal energy production, have already been discussed in Section 5.

Drainage waters from waste management facilities and contaminated sites may require treatment. Frequently such drainage waters are low in pH, in particular those from mining residues, where acid generation due to sulphide oxidation is a problem. For active management facilities and in cases where active remediation takes place, conventional treatment techniques are likely to be applicable [293]. In certain cases, conventional water treatment techniques would be inefficient. This would be particularly the case where radionuclide concentrations are low and the treatment plant would be required to run for extended times to prevent accumulation of radionuclides in certain environmental compartments. In such cases, artificial wetlands, phyto-remediation and similar passive treatment methods might hold the answer. Reactive barriers might also be used to prevent further dispersal of contaminated seepage waters. Such methods and their applicability in a remediation context will be discussed in more detail in a forthcoming IAEA technical publication [13].

12.3. CLEANER TECHNOLOGIES

12.3.1. Tools

The basic step in changing (industrial) processes towards lower resources use and hence less waste generation or to improve residue properties is an assessment of the substance flows involved. The concepts of materials flow analysis (MFA) and substance flow analysis (SFA) were developed to this end and consist of assessing where a substance enters or leaves an environmental compartment in a (industrial) process, and where it appears in products, intermediates, residues and wastes [294]. In the present context this requires, of course, that the respective materials be analysed for relevant radionuclides. Such MFAs can be undertaken at various levels, ranging from single plants or mines to whole countries (as illustrated in Fig. 31 for example). As a matter of fact, many industrial operations routinely apply MFA techniques to manage and control their materials requirements during the production process. This can form a good starting point for environment related MFAs.

While MFA and SFA are substance oriented, life cycle analysis (LCA) focuses on a product or a service and is a method for evaluating the impacts this product or service might have on the environment and natural resources. This evaluation method takes into account the impacts that go from the extraction of natural resources to waste disposal, including the end of life of a product (cradle to grave). LCA is a decision making tool that is promoted inter alia by the International Organization for Standardization (ISO 14000 series [295]). LCA was developed in response to enterprises for which environmental protection was a prime concern in the production, improvement and development of their products or their processes. Environmental certification according to the ISO 14000 series is seen by many companies as a means to exhibit an environmentally friendly image and thus increase their competitiveness.

Typically an LCA consists of four elements:

(1) Goal definition (ISO 14040) — the basis and scope of the evaluation,

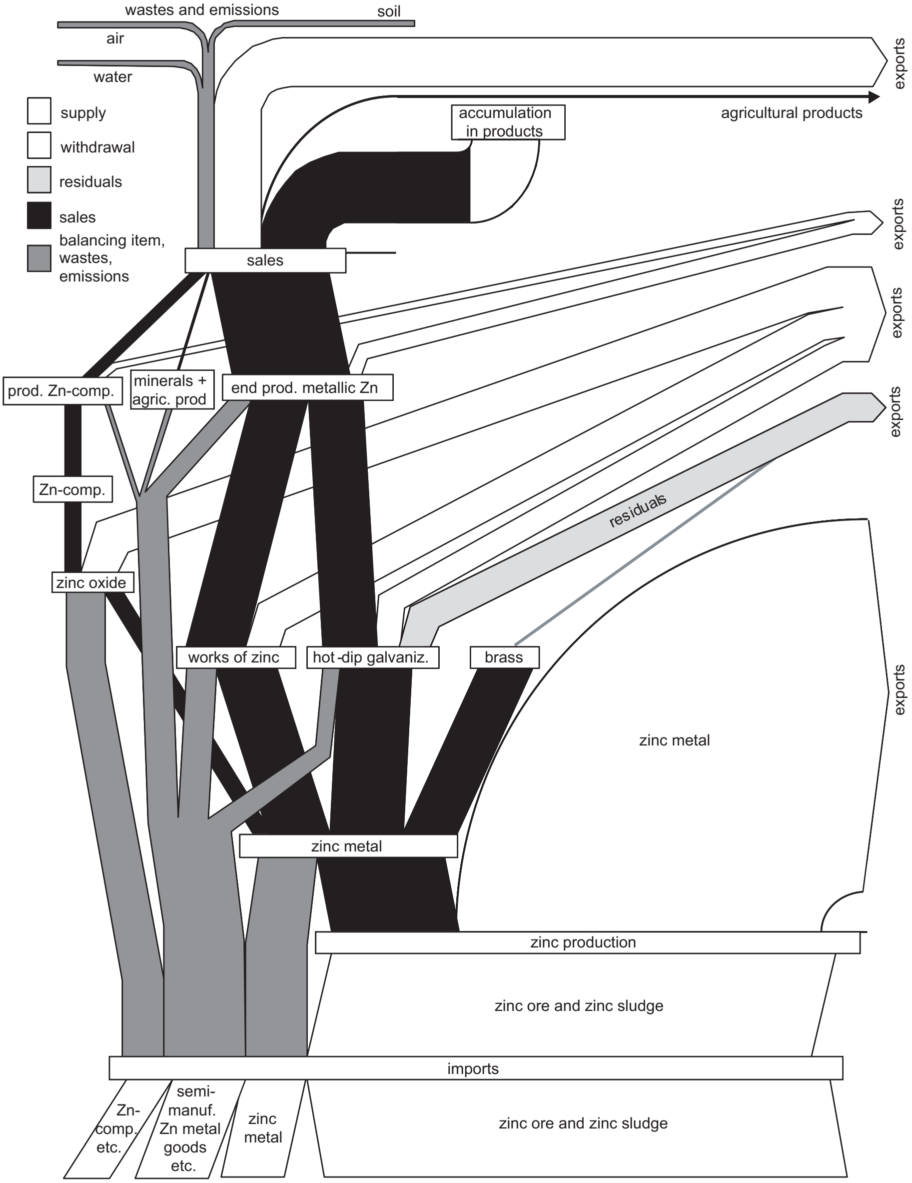

FIG. 31. Example for MFA: flow chart for zinc in the Netherlands in 1990 (simplified after Ref. [296]).

(2) Inventory analysis (ISO 14041) — process trees and material balances,
(3) Impact assessment (ISO 14042) — resources use and emissions as related to impacts,
(4) Improvement assessment/interpretation (ISO 14043) — identification of corrective actions.

On the basis of the results from such analyses, the potential for changing the (industrial) process to direct the NORM into those waste streams where it can be managed best can be assessed. There may be scope for adjusting the process so that the NORM mainly ends up in an inert form, or, for instance, digestion procedures may be altered such that NORM is not released or not converted into a mobile form. Concentrating the NORM into a single residue stream, or at least a small number of residue streams, would also make its management more efficient. Depending on the type of radionuclide and available disposal options, either increasing or decreasing the specific NORM concentrations may be of advantage. The first would reduce the volumes of waste to be treated, while the second might be desirable in a dilute and disperse scenario, or where disposal in a conventional landfill may then become possible.

Frequently raw material abstraction and processing takes place at different locations, even in different parts of the world. This results in residues and wastes being generated away from the source of the raw materials, and thus precludes such options as backfilling into the mined-out voids. Provided that adequate waste conditioning and waste management facilities are available to prevent the generation of environmental contamination, processing at or near the mining location might be of advantage. While until the recent past many developing countries relied on exports of raw materials, they now generate added value by exporting refined materials or semi-products. This development is frequently driven by the tightening environmental legislation in developed countries, which often makes the management of process residues expensive. Therefore, these changes have to be closely monitored for their environmental implications. Again, MFA would be a useful tool on a more global basis.

It should be pointed out that MFA does not require sophisticated computer simulation models, although they would be useful. Simple bookkeeping of masses or volumes would be sufficient, together with radiological and chemical analyses of the materials concerned.

12.3.2. Reuse/recycle

Reuse and recycling of NORM containing geological material has been already discussed to some extent in the context of immobilization/solidification. Residues typically require some sort of binder to give them useful mechanical properties. Application is restricted to situations where direct exposure and radon release are not of concern, for instance in road construction or for backfilling in mines. Recycling in nuclear waste management related applications, for example as structural materials or liners in repositories, might also be considered. From a logistic point of view, this might also be an option for various types of landfills, but national legislation typically prevents the mixing of hazardous and radioactive materials.

It has been indicated in the industry and material specific sections that at least nowadays the mining industry has become very resources conscious and screens its residues for potential value and reuse. Often residues are stockpiled for later reworking, when market conditions at a given time make processing not commercially viable. It should be noted that such stockpiles must not give rise to environmental contamination or human exposure, for example due to acid drainage formation or radon exhalation.

An example of re-using residues (iron oxides) from titanium oxide production is given in Appendix III.

12.3.3. Avoidance at source

In certain instances, the generation of NORM contaminated residues can be avoided at source, by selecting appropriate raw materials. This is probably the most desirable option, as it avoids exposures and the need to manage residues. Its economic viability, however, has to be evaluated and the resulting environmental benefits carefully balanced against any detriments.

In mining, NORM is often associated with particular parts of the geological strata. If these can be avoided during the mining operation by careful excavation, no contamination will be brought to the surface. For instance, in coal mining, seams high in sulphides such as pyrites can be avoided, and they are typically avoided to minimize the amount of sulphur in flue gases. Another example is peat, used as bio-fuel in Sweden, albeit on a rather small scale. Peat excavation is controlled by the Swedish Geological Survey and sampling for uranium is required before a concession is granted. Therefore the peat that is used in Sweden usually has low levels of uranium [271]. Similarly, in Sweden low activity materials are chosen for the production of sulphuric acid.

Since NORM is frequently associated with these sulphidic minerals, the side effect would be less NORM in flue gas desulphurization residues. Users can also make choices on the suppliers of raw materials. Market forces will thus disfavour suppliers with commodities of high NORM content. In addition to this, governments have taken in some instances measures to phase out hazardous substances, such as lead, cadmium or asbestos, from non-essential uses. MFA provided an essential tool to this end.

13. SOCIOECONOMIC IMPACT OF MANAGEMENT AND REMEDIATION MEASURES

Notwithstanding the criteria for the justification of remedial action as stipulated in the relevant safety guides [263], the benefit of changing industrial processes and of remediating NORM contaminated sites must be judged against the associated socioeconomic costs and other impacts. This probably needs to be undertaken on a case by case basis, rather than for industries in general. Costs and benefits will vary considerably, depending on the socioeconomic circumstances in any one country and for any one operation. Defining the scope of the cost–benefit analysis will significantly determine its outcome. It is important not to restrict it to purely radiation, health or environmental protection benefits, but to include the wider local and societal context. The implications of remedial actions, in particular when they affect ongoing practices, have to be analysed carefully. Remediation, which may entail removal of homes or closure of industries, will have a significant impact on the quality of life of the local population. The net benefit from such remediation has to be carefully weighed against the benefits from other ways of spending the same money, for instance better health care or education. On the national level the allocation of resources thus might follow other priorities. The resolution of this debate between remediation and other actions may depend heavily on the availability of national resources as well as of international aid.

The factors to be considered in remediation projects and relevant decision aiding techniques have been discussed in a dedicated IAEA publication [286]. There it was pointed out that decisions relating to the priority of environmental remediation of NORM contaminated sites will have to accommodate the realities of budget constraints and the relative importance of remediation compared with other (national, regional, etc.) needs. Land use considerations can be very important. Availability and allocation of other resources, such as technical or workforce investments, are other important considerations. Tools have been developed to predict and analyse the societal and economic outcome of decisions made on a technological, institutional or regulatory basis. Some of these tools have also been discussed in Ref. [291] and further references can be found therein.

A recent UNEP sponsored conference on abandoned mines, although not addressing explicitly the radiation problems, explored possible funding mechanisms for remediating orphaned contaminations [297]. In cases where remediation costs cannot be absorbed into operating cost, i.e. mainly in cases of orphan and historical wastes, a cost–benefit analysis is particularly useful to minimize and target public expenditure. As has been discussed in more detail in Ref. [286], the relative benefit derived from public expenditure on alternative projects has to be carefully judged. In many cases, however, it may not be clear who has the responsibility to remediate historical waste sites with the originator having ceased to exist, as is the case for example with many industries in Eastern Europe or the FSU countries. Acknowledging that such wastes and associated contamination may have arisen from activities in the public interest, for example defence related activities, governments have assumed responsibility even if the contamination was caused by a private company. The idea is that the government, i.e. the society that received the benefit from exploiting a natural resource, would retrospectively bear the cost. Other mechanisms discussed at the UNEP workshop [297] include levying a tax on current raw material use to pay for legacy wastes. This surcharge could be paid into a remediation fund, managed either nationally or internationally. As an additional form of insurance, consortia may be formed in a particular industry to pay for remediation.

In many Member States at present the granting of operation licenses or other authorizations is tied to proper waste disposal and closure plans. Financial provisions have to be arranged for the disposal of wastes and eventual cleanup of sites. These provisions may take the form of bonds or other securities that will ensure adequate cleanup, even if a company ceases to operate prematurely. In addition, governments may require the treatment and remediation of historical wastes originating from an ongoing concern, for example in the context of renewal of their licenses, and these activities may have to be costed in, i.e. all (environmental) costs have to be internalized.

Given the widespread and century long occurrence of mining, the potential need to address the problem in any one Member State may be enormous. The potential size of the task requires a careful assessment and site characterization in order to be able to target resources and to prioritize the problem sites. Such cadastral surveys of mining residues have been developed, for instance, in Germany [298].

As can be seen in the previous sections, many NORM related problems are associated with the mining industry. The mining industry has typical socio-economic patterns associated with it, such as the dependence of local communities on a single industry, i.e. the mine. Very often the industry operates under a ‘boom and bust’ scenario that is controlled by the availability of the resource and the competitiveness of the commodity prices. Often the profit margins are rather low and any additional cost incurred, for example by more stringent environmental or radiation protection standards, can price a company out of the market. Closure of the sole primary source of income usually has disastrous effects on the local communities affected, as has been observed in many primary resource based industries around the world.

MFAs, together with cost and economic analyses, may help to select an optimized strategy for abating the NORM problem. While full remediation and adjustment of waste streams to zero emission are desirable in the context of environmental sustainability, the socioeconomic feasibility of such undertakings must be carefully assessed. Objective assessment, however, is difficult and the judgment of what is feasible and what not is often rather subjective. The controlling variable is the profitability of the business concerned. The expected return on investments vary over time and between different cultures and different parts of the world. Among other factors, the economic viability of a treatment facility or a remediation project depends on the size of the operation.

The specific costs of various treatment and remediation technologies have been investigated in a recent IAEA publication [286]. It was concluded that very little detailed information on the various cost elements is available. The major source of data is still the Historical Cost Assessment System (HCAS) in the USA [299].

14. SUMMARY AND CONCLUSIONS

A picture of the worldwide scale of potential problems with NORM in a variety of industries has been developed on the basis of literature surveys and the contributions by experts from various Member States (Table LXIV). A comprehensive survey has been undertaken for Europe [6] and data are also available for North America. The knowledge base, however, for most less developed countries remains scarce. For several reasons the following circumstances in less developed countries are of particular concern:

(a) A large proportion of the world mining operations and to a lesser degree also milling operations are located in these countries.

(b) Environmental and radiation protection standards may be less stringent, or their enforcement may be less strict.

(c) Artisanal mining and milling and other artisanal industries with less stringent occupational health and safety precautions are widespread. As opposed to developed countries, such activities are still more integrated with private and family life, potentially leading to exposure of the public

TABLE LXIV. ACTIVITIES THAT MAY LEAD TO NORM CONTAMINATED RESIDUES AND SITES

Mineral ores and extracted materials		Other processing/manufacturing
Aluminium (bauxite)	Rare earths	Water treatment
Copper	Tin	Sewage treatment
Fluorospar	Titanium	Spas
Gypsum	Tungsten	Paper and pulp
Iron	Vanadium	Ceramics manufacture
Molybdenum	Zircon	Paint and pigment manufacture
Phosphate	Coal (and coal ash)	Metal foundry facilities
Phosphorus	Oil and gas	Optics
Potassium (potash)	Geothermal energy	Incandescent gas mantles
Precious metals (gold, silver)	Uranium and thorium	Refractory and abrasive sands
		Electronics manufacture
		Building materials

(e.g. residential/garden plots on or adjacent to 'industrial' sites and re-use of contaminated materials to optimize resource use).

(d) Limited or no resources are available to deal with legacy wastes and for upgrading plants and waste management infrastructure.

(e) Responsibilities for legacy wastes and contamination are unclear.

Regardless of the socioeconomic circumstances, NORM containing materials, residues and wastes pose particular technological challenges owing to:

(a) Their often large volumes;
(b) The long half-lives of the radionuclides concerned;
(c) Their widespread occurrence in many industrial and societal areas;
(d) The vast number of legacy sites.

Changing the processes to avoid NORM containing raw materials, to generate better waste forms or to reduce workplace exposure typically is not so much a technological challenge as an economic one. Sometimes it can also be a societal or socio-psychological one, as it may for instance involve replacing manual labour by machinery. Several studies and conferences have concluded that by proper workplace protection measures and good industrial practice, the majority of exposure scenarios can be eliminated.

For (geo)chemical reasons NORM related problems are mainly associated with the abstraction and processing of raw materials, including water, and in particular with the early stages of their processing. The processing of ores and other geological materials aims at arriving at the pure product, which typically means that most radionuclides end up in slags, fly ashes, scales, slurries and other residues. As a result the oil and gas industry, coal mining and burning, many metal ore and industrial mineral industries, as well as water users were identified as major producers of NORM containing residues and wastes.

Technologies to condition and dispose of NORM residues and wastes do exist in principle, but their (economic) applicability largely depends on the volumes of material arising. This report provides a broad overview of suitable technologies and strategies. While such technologies do exist in principle, and their application to legacy wastes and NORM contaminated sites is possible, the necessary resources are often not available in regions where there is a major need for them. The report briefly describes also low-tech and low maintenance solutions. More details on such technical solutions will be found in a forthcoming IAEA publication [13].

Given the long half-lives of the relevant radionuclides (mainly the uranium series) and the limited resources and technical options to deal with legacy wastes, the question of long term stewardship and monitoring arises. Hence the concept of monitored natural attenuation and its implications is increasingly being discussed.

No simple solution can be found for funding the remediation of legacy sites. In some developed countries a decisive effort has been made over the last two decades or so. The limited gross national products of many other Member States has prevented them carrying out similar activities. The multinational character of many mining operators has often made it difficult to hold them accountable for cleanup of historical wastes in particular. The situation is slowly changing, however, as these operators try to promote a green image of themselves. On the other hand, the contamination legacy from small scale artisanal mining and other artisanal industrial activities remains a difficult problem to solve.

Appendix I

TIN MINING AND RELATED PROCESSES IN MALAYSIA

I.1. INTRODUCTION

Malaysia has long been known as one of the world's leading tin producers. However, the overall trend of tin ore concentrate production has been declining (from 10 000 t in 1993 to 6000 t in 1998). This is partly due to price decreases, depletion of high grade reserves and rising production costs. The number of operating tin mines decreased to 38 in 1998. This has left behind more than 2000 mined-out ponds. The lengths of the ponds are up to 500 and 1000 m for gravel pump and dredge mines, respectively. Most of the ponds have thick layers of slurry slime at the bottom. The slurry contains heavy minerals such as ilmenite, monazite, zircon and xenotime.

I.2. MINING AND EXTRACTION METHODS

I.2.1. Dredging

Depending on the type of dredger (bucket or suction) continuous digging at water depths of up to 400 m is possible. The material brought to the surface is spread on revolving or oscillating screens and disaggregated using high pressure water jets. Large stones and rubble are retained by these screens and discarded. The tin bearing material (the undersize portion) passes to a separating plant consisting of primary, secondary and tertiary jigs, where tin ore concentrates are separated from tailings containing sand, mud, clay and gravel. The usual content of cassiterite in the dredge concentrate at this stage is of the order of 5–25% [300]. The average mineral composition of the dredge concentrate is shown in Table XLV. The flow chart of the dredging process [301] is shown in Fig. 32. The dredge concentrate is transported to a treatment plant commonly known as a tin shed, where the marketable ore containing at least 76% tin is separated out, leaving tin tailings, locally known as amang (Fig. 33). At the tin shed, unrefined zircon, monazite, columbite and ilmenite ores are also separated out.

I.2.2. Gravel pumps

In gravel pump mining, powerful jets of water are used to wash out gravel containing tin ore from the exposed face of a quarry or hillside. The slurry is

TABLE XLV. MINERAL COMPOSITION OF THE DREDGE CONCENTRATE BY wt (%)

Mineral	Values from Ref. [302]	Values from Ref. [301]
Cassiterite	9.7	8.4
Ilmenite	41.5	53.1
Monazite	2.0	0.5
Zircon	2.1	0.7
Tourmaline	0.8	—
Rutile	0.5	—
Pyrite	—	1.3
Silica (sand)	43.4	36.0

collected in a sump at the base of the slope and is then pumped by a gravel pump to a long sloping sluice box (locally called a palong) for gravity separation of heavy minerals. Stones are extracted and ejected from the feeder palong, while the remaining material is evenly distributed to the different compartments of the palong. The concentrate accumulating behind the wooden bars on the palong is collected and taken to the tin shed for further treatment to separate out marketable tin concentrate leaving behind tailings. The unwanted matter — sand, mud, clay and gravel — flows to the lower end of the palong and away along the channel to the tailings area [300].

I.2.3. Open cast

In this method, the tin bearing material is excavated and conveyed as it is by mechanical means to the treatment plant. Water is then added to enable the heavier tin concentrate to be separated from the remainder of the material.

I.2.4. Underground mining

In underground mines, tin bearing rock is obtained after blasting and is then transported to a mine head mill. There the rock is crushed and tin ore concentrate is extracted.

I.2.5. Re-working of residues

Amang contains tin ores, quartz sand, monazite, zircon, xenotime ilmenite, struverite and other minerals, including those containing uranium and thorium (table XLVI). Amang processing plants started operations in the 1950s.

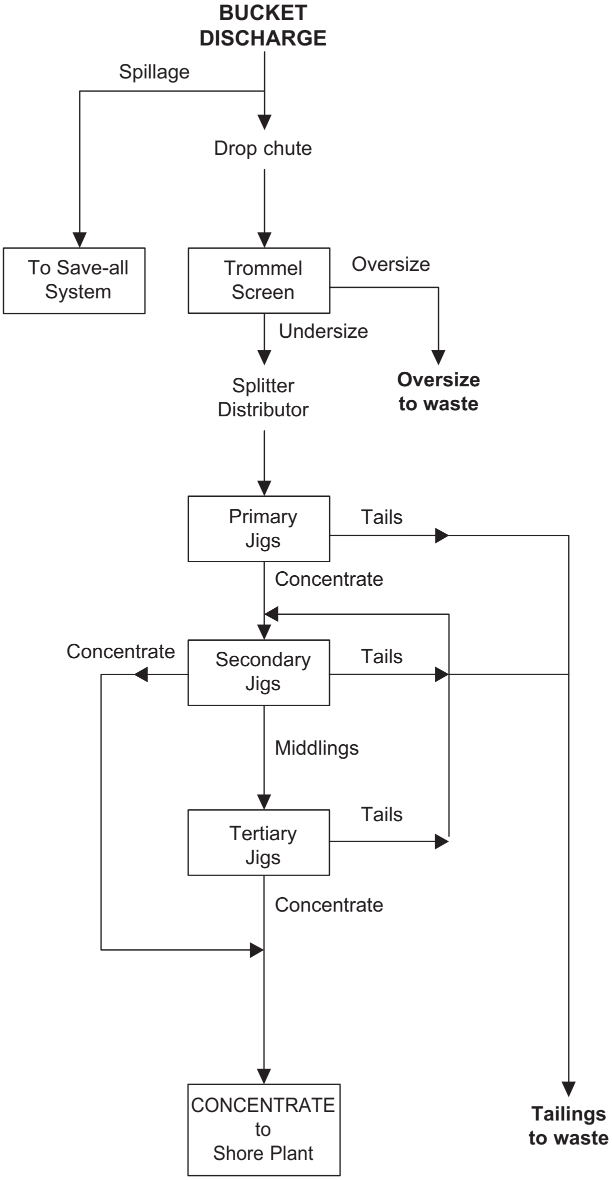

FIG. 32. Flow chart of the dredging process [301].

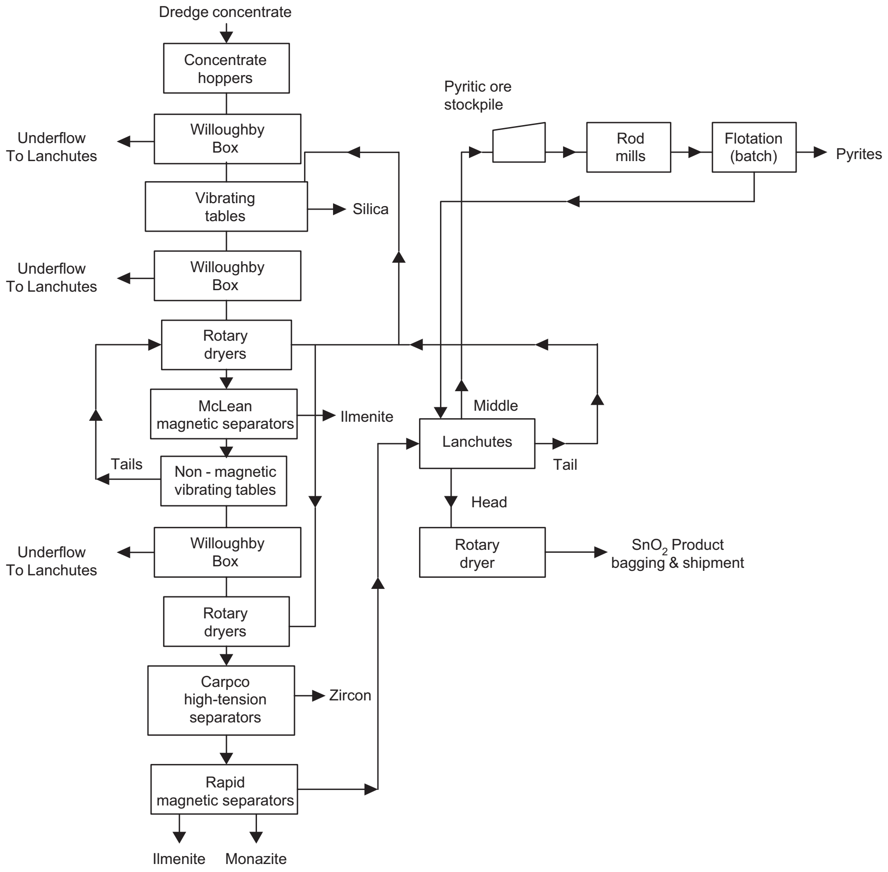

FIG. 33. The tin shed process [302].

In 1989, there were 66 amang plants operating in Malaysia and these produced 3% of the total tin concentrate in the country. However, the number of amang plants is decreasing due to the decline of the tin mining industry.

In these plants, tin ores are separated from other minerals using magnetic separators, a classifier, shaking tables for gravitational separation, dryers and high tension electrostatic separators (Fig. 34). After drying in the sun, the amang is processed by using magnetic separators to separate ilmenite (magnetic) from other non-magnetic materials (monazite, zircon, xenotime and cassiterite). Using a Willoughby classifier, the materials are separated into an overflow and an underflow. Both material streams are further treated in the same manner. Alternatively, rapid magnetic separators discriminate between magnetic (xenotime) and non-magnetic (monazite, zircon and cassiterite)

TABLE XLVI. MINERAL COMPOSITION (%) OF AMANG [304]

Mineral	Jelapang, Perak	Petaling, Selangor	Pucong, Selangor
Zircon	<0.5	3.5	5.5
Monazite	0.5	0.5	<0.5
Xenotime	4.0	<0.5	<0.5
Ilmenite	85.5	76.5	60.5
Rutile	<0.5	0.5	1.5
Quartz	<0.5	4.5	19.0
Pyrite	<0.5	0.5	—

minerals. On shaking tables the heavy minerals are separated out into a concentrate and the remainder discarded as tailings. After drying, the concentrate is treated in a high tension electrostatic separator to separate conducting minerals (cassiterite) from non-conducting minerals (monazite, xenotime, zircon and quartz). The electrostatic separator effluents are then treated in a rapid magnetic separator to separate (non-magnetic) cassiterite and zircon from (magnetic) ilmenite and monazite.

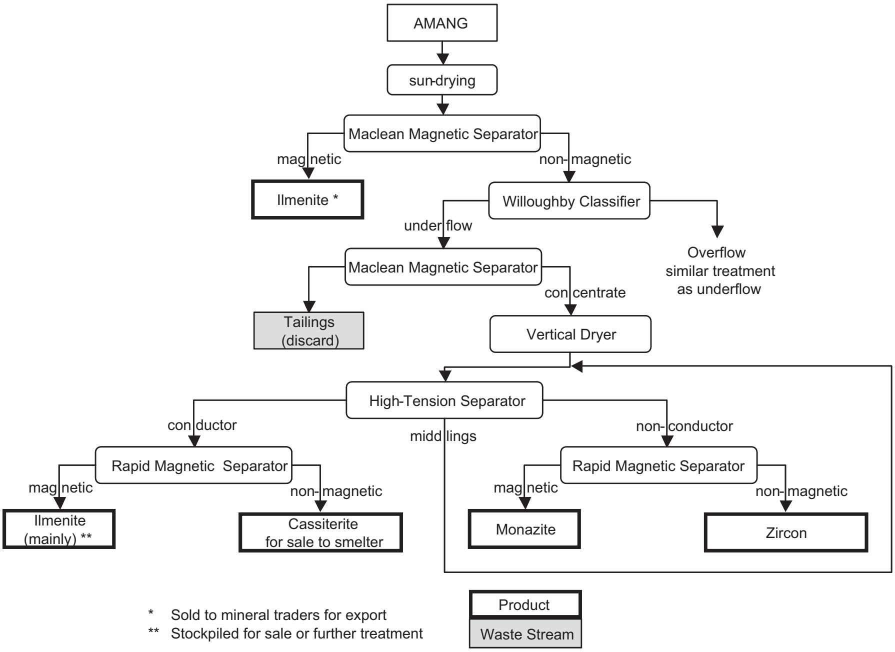

FIG. 34. Flow chart for the recovery of cassiterite and heavy minerals from amang [306].

I.3. TIN SMELTING

Malaysia did have two tin smelters. At their peak both plants were capable of processing 60 000 t/a of tin ore concentrates. However, one of the plants ceased operation in 1999. The tin ores recovered as described above form the feedstock for the smelters producing tin metal. The residue produced is slag. The total accumulated tin slag in 2000 was estimated to be 120 000 t.

I.4. DISPOSAL OF RESIDUES

Prior to the enforcement of the Atomic Energy Licensing Act 1984 (Act 304) in 1985, minerals and slag were not considered as radioactive. The residues were disposed of in landfills that were subsequently redeveloped. A study [305] showed that a public park has been contaminated with radioactive minerals down to a depth of more than 30 cm. The study also showed that, to some extent, the soil outside the tin mine perimeter was contaminated with NORM from the mining activities. Following the establishment of the Malaysian Atomic Energy Licensing Board (AELB) in February 1985, the disposal of tin slag in landfills has required a radiological impact assessment prior to the AELB's approval.

I.5. REMEDIATION ACTIVITIES

The dredged ponds can be extensive in size and most of them are quite deep. Remediation is necessary before the former mining sites can be re-used. Some of the mining sites have been used for the development of residential areas after landfilling. A feasibility study on remediation of mined-out land in Malaysia is planned, but it has not yet started.

Tin slags and contaminated soils from the former tin smelter site were removed and transported to another site. The work was monitored by the national regulatory authority. The site was then covered with local red soil and can now be used for other purposes.

Appendix II

TITANIUM PROCESSING

II.1. INTRODUCTION

The mining methods and resources of titanium have already been discussed in the main part of this report. In the following the different processing methods for titanium metals and pigment will discussed, together with their radiological implications.

II.2. PRODUCTION OF TITANIUM METAL

The Kroll method is used on large scales for titanium production and involves the action of chlorine and carbon upon ilmenite ($TiFeO_3$) or rutile (TiO_2). The resultant titanium tetrachloride, $TiCl_4$, is separated from the ferric chloride, $FeCl_3$, by fractional distillation. Finally $TiCl_4$ is reduced to metallic titanium by reduction with magnesium. Air is excluded so as to prevent contamination of the product with oxygen or nitrogen,

$$2\ TiFeO_3 + 7\ Cl_2 + 6\ C\ (900°C) \rightarrow 2\ TiCl_4 + 2\ FeCl_3 + 6\ CO^{\uparrow}$$

$$TiCl_4 + 2\ Mg\ (1100°C) \rightarrow 2\ MgCl_2 + Ti^0$$

Excess magnesium and magnesium dichloride is removed from the product by treatment with water and hydrochloric acid to leave a titanium 'sponge'. This can be melted under a helium or argon atmosphere to allow casting as bars.

II.3. PRODUCTION OF TITANIUM DIOXIDE PIGMENTS

In a first step the ore is crushed to a grain size of about 12 mm and then ground to a fine powder. Sulphides and magnetite as ore accessories are separated by floatation and in magnetic separators, respectively, leading to an increase of ore concentration from 18 to 45% (Fig. 35).

Two methods are used to produce TiO_2 from the ore, employing either sulphuric or hydrochloric acid.

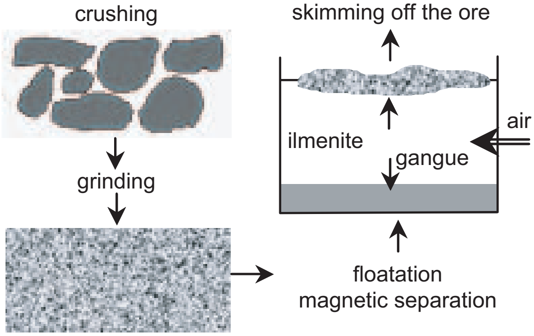

FIG. 35. Generic scheme of titanium ore dressing.

The sulphate process (Fig. 36) was developed in 1915 by the Norwegians Farup and Jebsen and has been used on an industrial scale since 1919. The finely ground and pre-concentrated ore is digested with concentrated sulphuric acid, converting the ferric and titanium oxides into sulphates. The large quantities of sulphur dioxide generated are neutralized by adding sodium hydroxide. Ferric and titanium sulphate are separated by re-crystallization. Titanium sulphate can be converted into an oxyhydrate by boiling in water, and is then dehydrated by roasting at 800–1000°C in a rotating furnace. The resulting TiO_2 may undergo further purification steps, depending on the intended application.

Sea dumping of the waste from the boiling step, dilute sulphuric acid, caused serious environmental concern and has been stopped, at least in Europe. Per tonne of TiO_2 produced, 6–8 t of a dilute (20–22%) acid are generated. The acid is too dilute for further commercial use and often contaminated by heavy metals and, indeed, NORM. In a relatively energy consuming process the acid is concentrated and accessory salts, mainly ferric sulphates and co-precipitates, are removed. The acid is fed back into the production process, while the ferric sulphate is used commercially as a flocculant for waste water treatment, mainly to remove phosphates as weakly soluble ferric phosphates. Any ferric oxides produced are used as landfill.

In the chloride process (Fig. 37) the pre-concentrated titanium ore or rutile is mixed with coke and heated to 1000°C in the presence of chlorine gas. The chlorine reacts with the ore to give titanium tetrachloride and carbon dioxide:

$$TiO_2 + C + 2\ Cl_{2(g)}\ (1000°C) \rightarrow TiCl_{4(g)} + CO_{2(g)}\uparrow$$

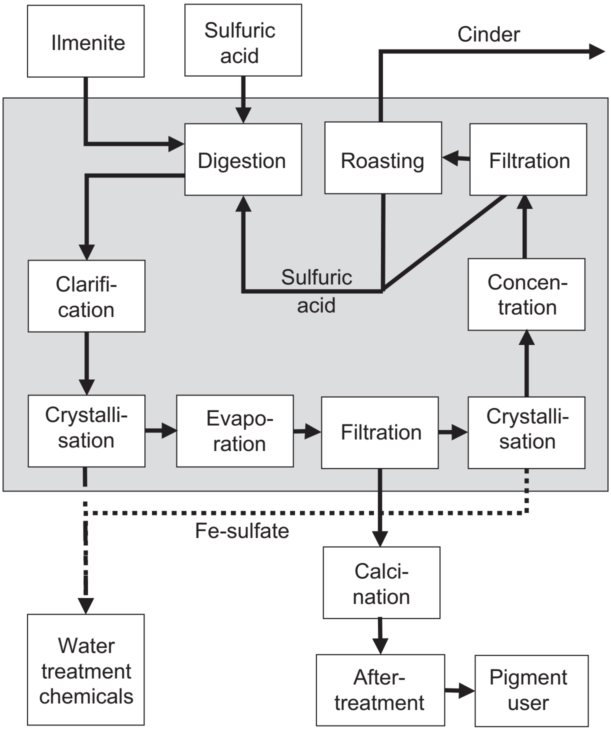

FIG. 36. The sulphate process for TiO_2 *production [306].*

Iron (II) chloride formed during the chloride process is dissolved in water and separated. Any hydrochloric acid formed by the dissolution of chlorine in the residual cinders is separated and sold. The gaseous $TiCl_4$ is condensed and purified by re-destillation, before being re-oxidized to give pure TiO_2. The freed chlorine is fed back into the production process.

Fewer environmental pollution problems are encountered when pigment is produced from rutile rather than ilmenite. The chloride process, using a rutile feed, generates about 0.2 t of waste per tonne of TiO_2 product; the sulphate process, using ilmenite, generates about 3.5 t of waste per tonne of product. Producing synthetic rutile from ilmenite results in about 0.7 t of waste, mainly iron oxide, per tonne of product. Direct chlorination of ilmenite generates about 1.2 t of waste, mainly ferric chloride, per tonne of TiO_2.

For a number of technological reasons related to pigment application, both processes, which generate different mineralogies of TiO_2, have retained their validity.

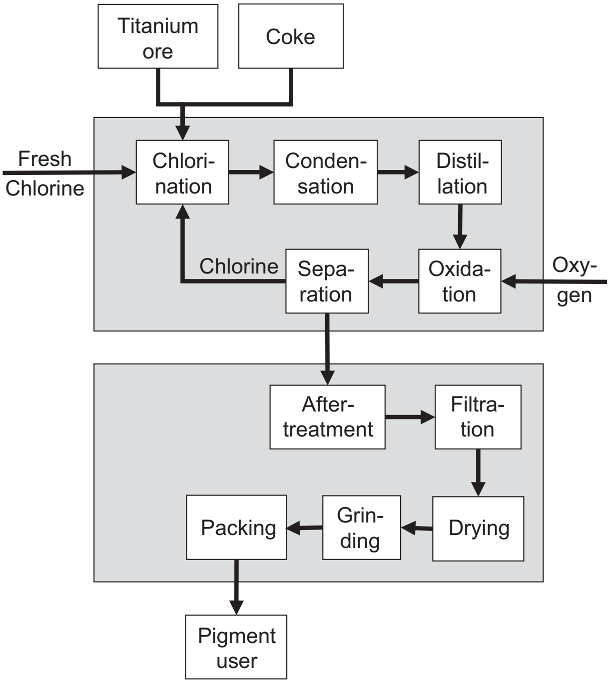

FIG. 37. The chloride process for TiO_2 production [306].

II.4. SYNTHETIC RUTILE PRODUCTION

Ilmenite is a mixed oxide containing both titanium and iron. As an intermediate on the way to titanium pigment, synthetic rutile is produced, resulting in a fivefold increase in the value per ton of material [307]. While Australia is probably the world's biggest producer, in Malaysia synthetic rutile is produced from the ilmenite separated out in the amang plants.

The Becher process is generally used, in which the iron oxide contained in the ilmenite is reduced to metallic iron by adding coal and then re-oxidized to iron oxide. In the process, the titanium dioxide separates out as synthetic rutile of about 91–93% purity. The process involves a high temperature kiln to heat the ilmenite with coal and sulphur. After screening, the mixed slurry of metallic iron and titanium dioxide in water is oxidized with air and separated in settling ponds. The resultant synthetic rutile is acid washed to remove other metal oxides and then dried and sold for conversion to the pigment.

Some plants, for example in Malaysia, convert ilmenite directly with chlorine and dispose of the resulting ferric chloride in landfill sites. In countries where electricity is cheap (such as Canada, South Africa or Norway) electric furnaces are used to produce a titanium dioxide rich slag, and the iron, instead of being returned to the mine as waste as in the Becher process, is sold as pig iron. The electric furnace route is very competitive and is seen as depressing synthetic rutile prices, but also provides incentives to add value to residues otherwise ending up in waste streams. Proprietary methods for process improvement have been developed, for example the process shown in Fig. 38 [308].

The management of waste streams from the production of synthetic rutile in Australia is illustrated in Table XLVII. The low value iron oxide (which represented at least 40% of the ilmenite) is returned to the mine site as waste or landfill. It is estimated that 30 000 t/a of iron oxide residue is produced in Malaysia.

Briquetting of fine iron oxide dust residues allows their use in blast furnaces to produce pig iron, rather than an environmentally sensitive disposal [307]. The heat roasting process as employed in Australia is illustrated in Fig. 39. Limited amounts of iron oxide can also be used as an ingredient for making paving bricks or as soil additives, as in the case of red muds from aluminium production.

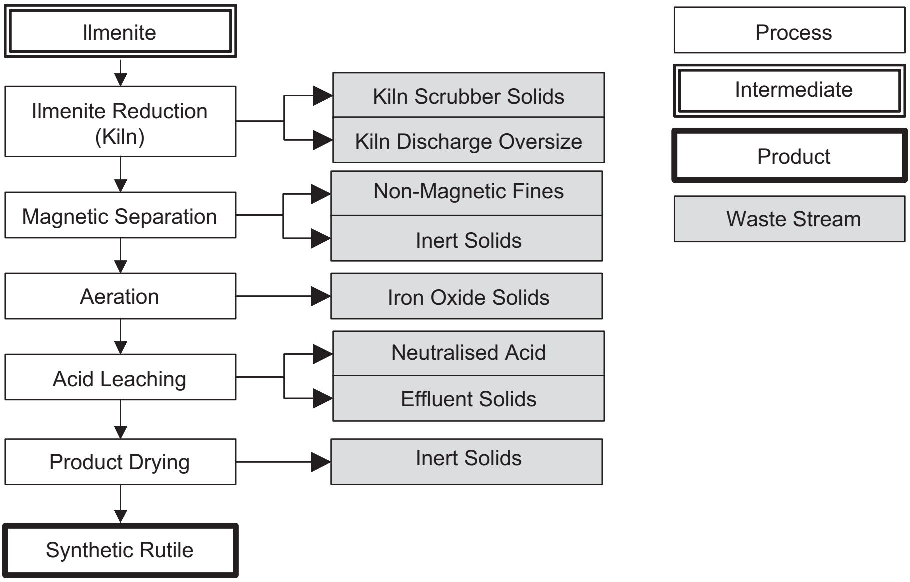

FIG. 38. Generalized synthetic rutile process flow chart [166].

TABLE XLVII. MANAGEMENT OF WASTE STREAMS GENERATED DURING SYNTHETIC RUTILE PRODUCTION [167]

Waste stream	Management
Kiln scrubber solids	Wet scrubbers: pumped as slurry to a lined pond. Subsequently, either rehabilitation in situ or, still wet, dug out and trucked to final disposal site. Dry scrubbers: directly trucked to final disposal site. Wetted prior to trucking to prevent dusting during transport. In any case, solids must be allowed to settle and dry before capping and rehabilitation.
Kiln discharge oversize	Collected into bounded paddock from where trucked to lined dry waste disposal site. Alternatively, it can be crushed and recycled, or used as a road base.
Non-magnetic fines	Collected into plant storage bins, loaded into trucks, wetted and transported to final disposal site. Alternatively, transported as slurry through pipeline to lined pond at the disposal site. They must be allowed to settle and dry before capping and rehabilitation.
Inert solids	Collected from plant dust extraction system via wet or dry scrubbers, followed either by intermediate storage in ponds or by final disposal in lined sites. Dry scrubber solids are wetted prior to trucking to prevent dusting during transport.
Iron oxide solids	Stored in lined ponds to allow settling and drying. Dry material can either be rehabilitated in situ or trucked for final disposal to repository site. Alternatively, they may be transported as a slurry through a pipeline directly to the lined ponds of the final disposal site. Thus dust generation from handling the dry material is eliminated. They must be allowed to settle and dry before capping and rehabilitation. A promising commercial scale technology of processing the iron oxide solids to produce iron has been under development. If successful, it could result in practical elimination of this largest solid waste stream.
Neutralized acid effluent solids	Pumped to solar drying ponds, from where, still wet, they are dug out and trucked to final disposal site. Alternatively, transported as slurry through pipeline to lined pond at the disposal site. They must be allowed to settle and dry before capping and rehabilitation. A recently developed alternative is the option to utilize the solids as a slow release sulphur fertilizer.

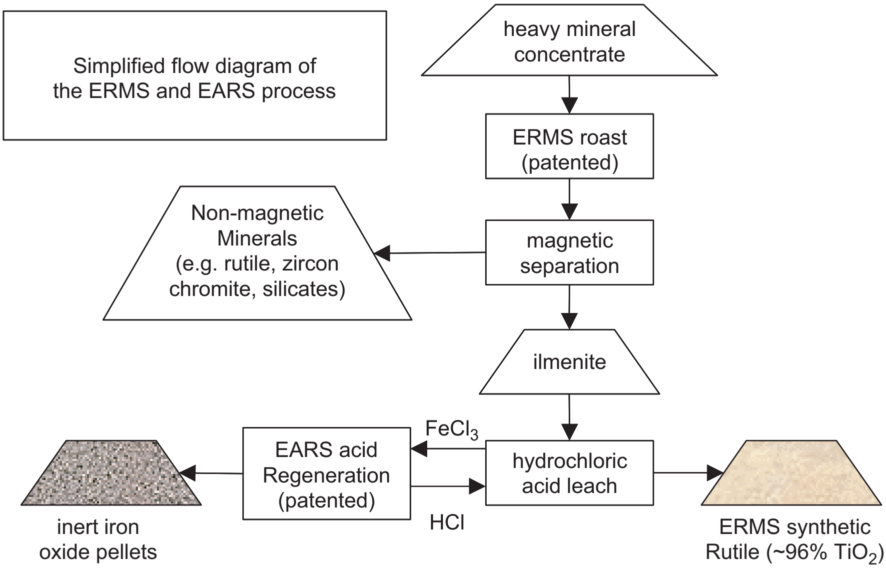

FIG. 39. A proprietary process for ilmenite separation and conversion into rutile [308] (ERMS, enhanced coasting and magnetic separation; EARS, enhanced acid regeneration system).

II.5. DOSES DURING PROCESSES

The exposure risk of producing and using titanium feedstocks is also low. The highest potential exposure occurs during the maintenance of plant or the removal of process scales containing high concentrations of radium.

Appendix III

RARE EARTH ELEMENT PROCESSING IN MALAYSIA

III.1. INTRODUCTION

Monazites are processed for REEs such as cerium, lanthanum and yttrium. Xenotime, on the other hand, is used to extract yttrium. The rare earth plant in Malaysia was capable of processing 4000 t/a of monazite. This enabled it to produce 4200 t of rare earth chloride and 550 t of rare earth carbonate [309]. Xenotime processing in Malaysia started in 1976. Thorium hydroxide was produced as residue. The operation was terminated in 1986. Operation of the monazite processing plant started in 1982 and terminated in 1992.

III.2. EXTRACTION METHODS

III.2.1. Mixed rare earths

Mixed rare earth carbonates and chlorides are mainly produced from the digestion of monazite. About 24 000 t of monazite (70% local) were processed in Malaysia. The process uses acidic and alkaline digestion, filtration, precipitation and solvent extraction. After alkaline digestion and filtration, the phosphate solution is separated from solid hydroxide cake containing elements such as thorium, uranium, radium, lead and REEs. The phosphate solution undergoes crystallization and filtration processes to produce tri-calcium phosphate (used for the manufacture of fertilizer) and wastewater. The hydroxide cake undergoes acid digestion and filtration to produce a chloride solution of REEs, lead and radium and a solid thorium hydroxide residue containing uranium, thorium and REEs. Precipitation of the chloride solution removes lead sulphide and barium radium sulphates as lead cake residue. Solvent extraction followed by precipitation of the solution produces rare earth carbonates and chlorides separately. The process flow chart [310] is shown in Fig. 40. Thorium hydroxide and lead cake are radioactive residues that need to be managed safely. A typical composition of thorium hydroxide from a monazite and xenotime cracking plant is shown in Table XLVIII.

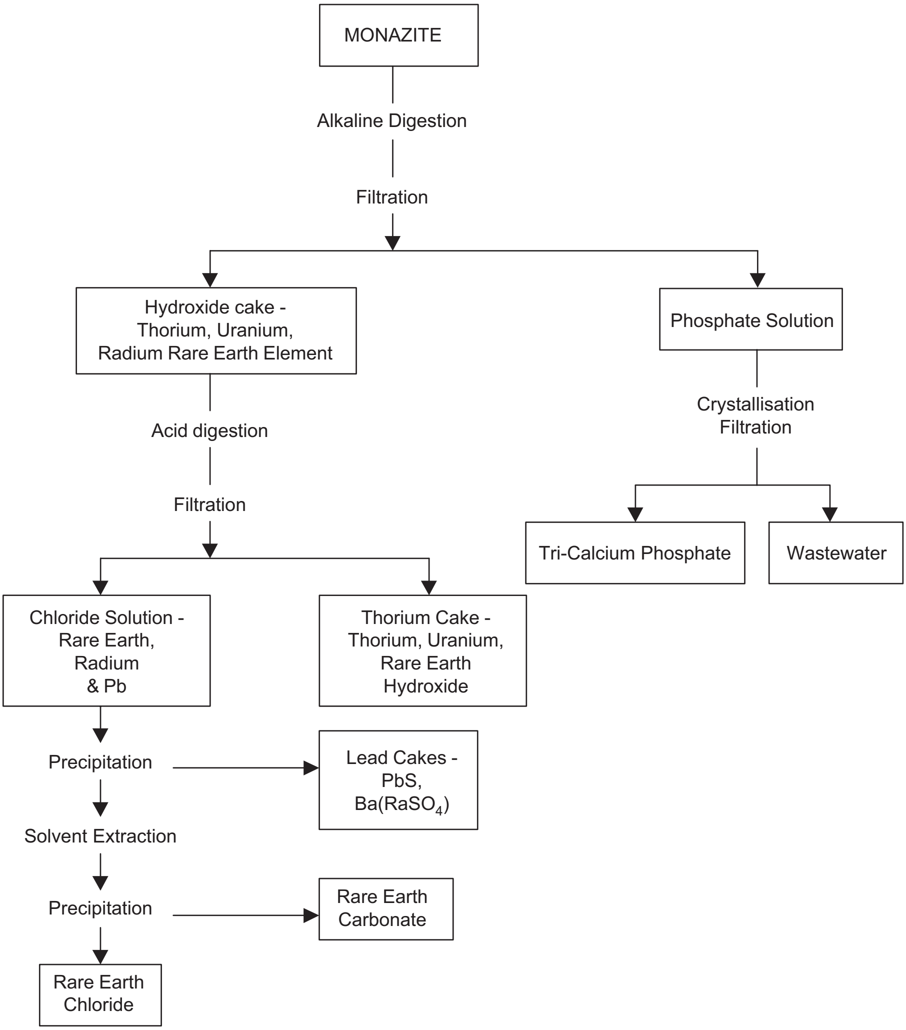

FIG. 40. Flow chart for the recovery of rare earths from amang derived monazite [303].

III.2.2. Yttrium

Yttrium oxide is obtained from the processing of xenotime (YPO_4). About 2000 t of xenotime have been processed in Malaysia. The process involves milling, roasting, digestion, leaching, filtration, precipitation and calcination. Thorium hydroxide is produced as residue. The process flow chart for the recovery of yttrium oxide [304] is shown in Fig. 41.

TABLE XLVIII. TYPICAL COMPOSITION (%) OF THORIUM HYDROXIDE WASTES

Content	From monazite	From xenotime
Insoluble residue	21	45
Rare earth chloride	8	15
Thorium hydroxide	14	1.6
Uranium hydroxide	0.6	1.6
Moisture	54	22

III.3. WASTE MANAGEMENT

Initially the residues from the xenotime and monazite processing plants were dumped in a trench on the plant premises. Later, the dumped residues were dug out and drummed. The residues from later operation were stored in drums. Finally, the drummed residues were stored in an above ground concrete

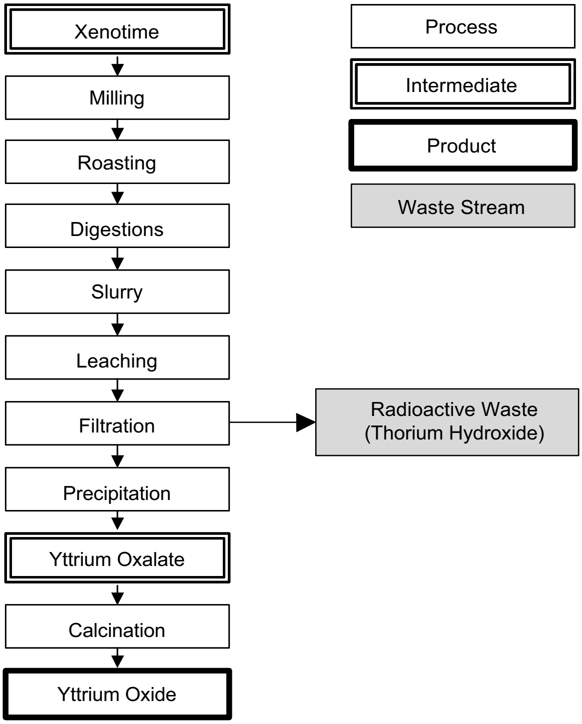

FIG. 41. Process flow chart for the recovery of yttrium oxide [304].

bunker, known as the long term storage facility (LTSF). The quantity of thorium hydroxide and lead cake wastes is estimated to be about 12 000 t.

At the end of its operation, the rare earth plant left 32 m^3 diethylhexyl phosphoric acid (DEHPA) residues, an organic solvent used to extract rare earths from the monazite. The residue was transported to the National Radioactive Waste Management Centre at the Malaysian Institute for Nuclear Technology (MINT) in Bangi for treatment and disposal.

III.4. DECOMMISSIONING AND REMEDIATION

As the operation of the xenotime and monazite processing plants has been terminated, the plants need to be decommissioned. Decommissioning will result in large amounts of slightly contaminated plant equipment, building materials and soils. The decommissioning waste is planned to be disposed of in an engineered cell built next to the existing LTSF.

Remediation of the plant site would be carried out together with the decommissioning process. After the decommissioning of the plant equipment and buildings, the contaminated soils within the plant vicinity would be excavated to an approved acceptable limit by the regulatory authority. The plant site would then be covered with local soils.

With regard to the stored thorium hydroxide residues, no decision has been made as to what to do with them yet. Studies have been carried out to extract residual REEs as well as separating uranium and thorium from them [311]. The process flow chart is shown in Fig. 42. A method to extract uranium (95%) and thorium (98%) quantitatively from the thorium hydroxide residue has been developed [309]. In addition, a study to use ordinary Portland cement for the conditioning of the waste was carried out [312] in preparation for the disposal of the thorium hydroxide wastes. Furthermore, studies on the possibility of using local Malaysian soils as radionuclide adsorbent for uranium and thorium from thorium hydroxide wastes have been undertaken [313, 314].

Radioactively contaminated DEHPA used in the solvent extraction of rare earths in the monazite processing plant was left over as waste at the end of the operation. The waste was sent to the National Radioactive Waste Management Center at MINT for treatment and disposal. Pre-treatment of DEHPA waste prior to incineration has been studied [315]. The flow chart for the pre-treatment of DEHPA waste is shown in Fig. 43. Another study on the cleaning and recycling of the contaminated DEHPA was also carried out [301]. DEHPA was found to be contaminated with significant amounts of U (990 ppm). Interestingly, it was also found that the DEHPA residues still contain 9600 ppm Sm, 7320 ppm Nd, 680 ppm Y and 290 ppm Dy.

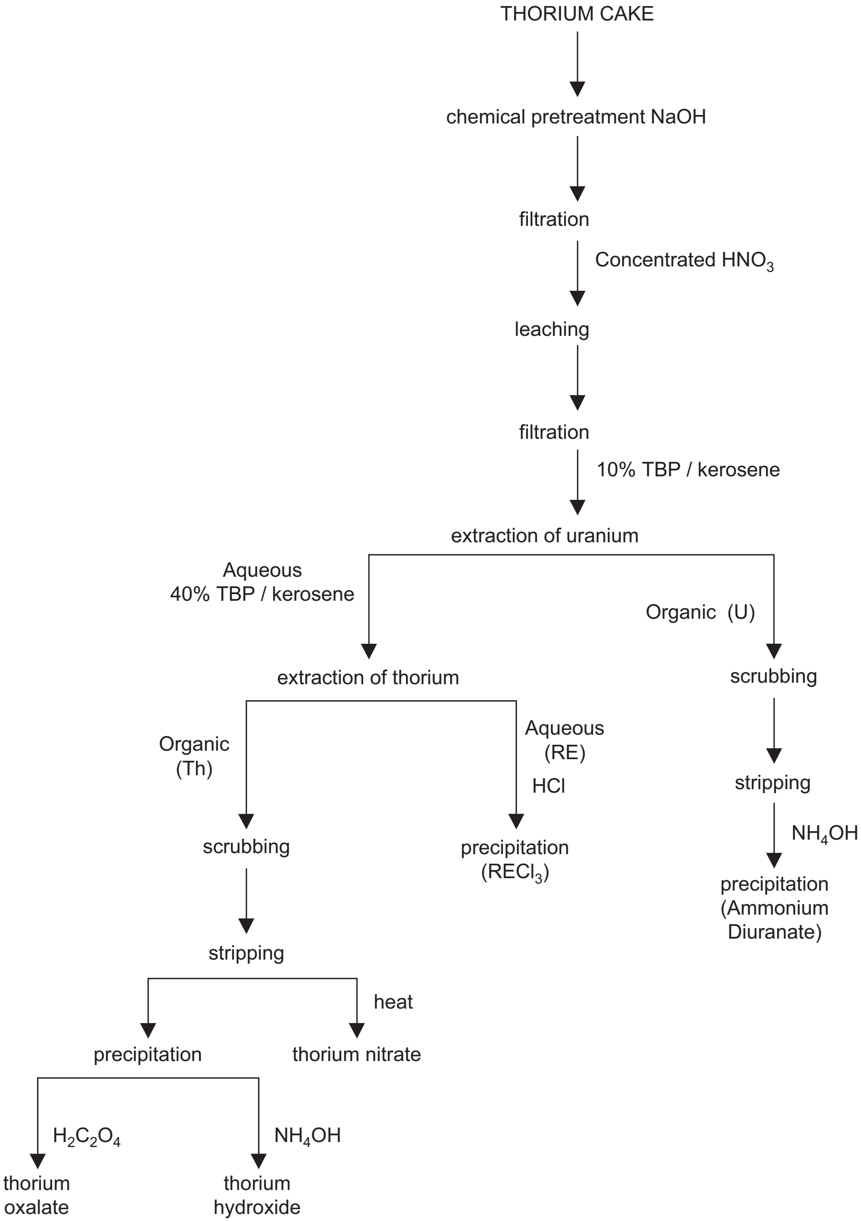

FIG. 42. Experimental extraction scheme for U, Th and REEs from thorium cake ($Th(OH)_4$) [309].

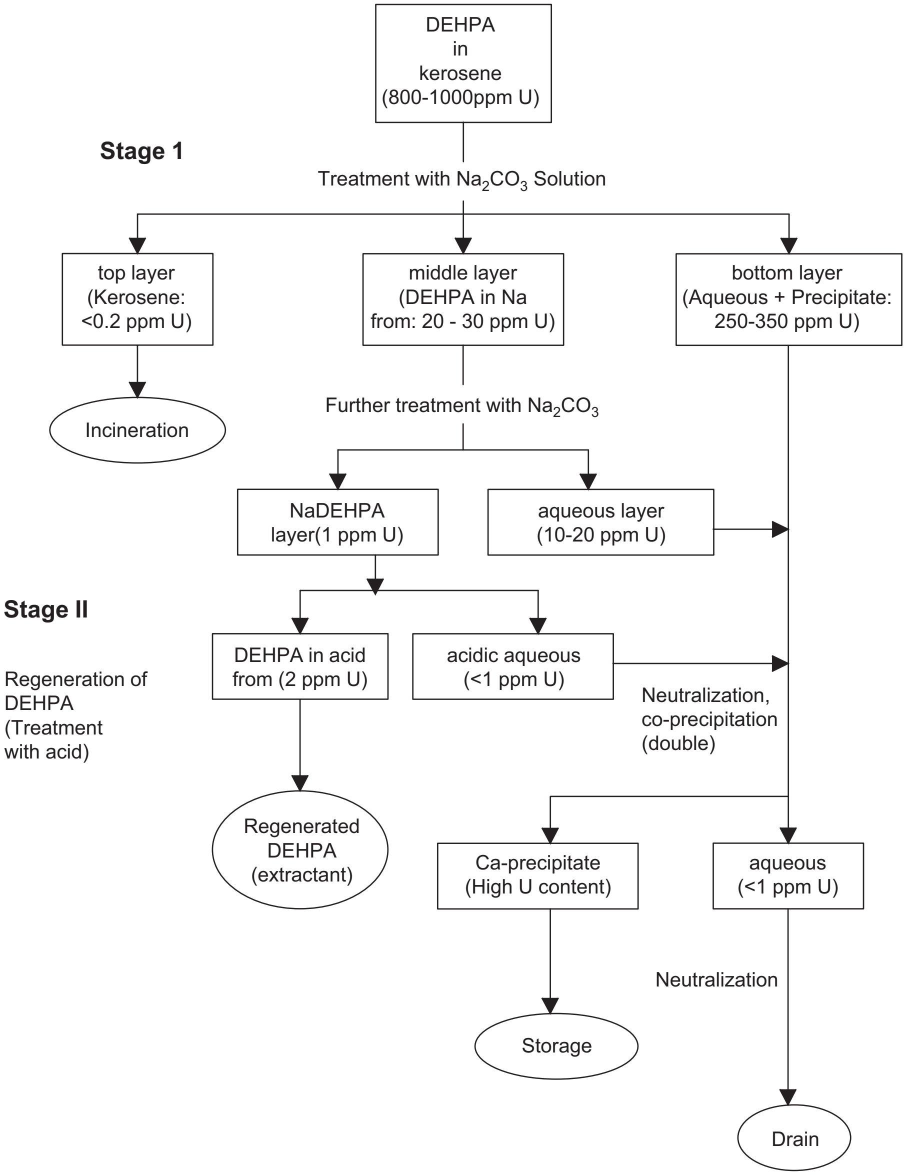

FIG. 43. Flow chart for experimental pre-treatment of DEHPA waste [315].

Appendix IV

NIOBIUM PROCESSING IN BRAZIL

IV.1. INTRODUCTION

The process of niobium isolation appears to be complicated. Niobium minerals usually contain both niobium and tantalum. Since they are so similar chemically, it is difficult to separate them. Niobium can be extracted from the ores by first fusing the ore with alkali, and then extracting the resultant mixture into hydrofluoric acid, HF. Current methodology involves the separation of tantalum from these acid solutions using a liquid–liquid extraction technique. In this process, tantalum salts are extracted into the ketone MIBK (methyl isobutyl ketone, 4-methyl-pentan-2-one). The niobium remains in the HF solution. Acidification of the HF solution followed by further extraction in MIBK gives an organic solution containing niobium. After conversion to the oxide, metallic niobium can be made by reduction with sodium or carbon. Electrolysis of molten fluorides is also used [316].

The two industries that are reported here are located in Brazil. One in the state of Minas Gerais, located in the southeast region of the country – which will be treated as Niobium Industry I — and the other located in the west central region — Niobium Industry II.

IV.2. NIOBIUM INDUSTRY I

Magmatic and pneumatolitic processes have shaped the appearance of most of the radioelements in the earth's crust. In the magmas and their hydrothermal derivatives, uranium and thorium may appear together in pyrochlore minerals, $(Ca,Na)_2(Nb,Ta)_2O_6F$. In Niobium Industry I the element occurs as barium pyrochlore which comes to be essentially an oxyhydroxide of niobium, barium, titanium, rare earths from the cerium group and thorium. The industry mines the ore to produce Fe–Nb concentrate, niobium oxide and metallic niobium.

The industry has six main units: mining, concentration, calcination and acid leaching, production of Fe–Nb league, production of niobium oxide and metallic niobium. The installation has two settling ponds where the residues from the physical and chemical processing are disposed of, and a slag deposit. None of the settling ponds release liquid effluents into the environment.

Mining of the ore is carried out in an open cast pit and the waste rock is deposited in waste rock piles.

The operational process is depicted in Fig. 44. It includes the steps of ore mining, grinding/crushing, magnetic separation, de-sliming, floatation, calcinations, leaching, drying and aluminothermic reduction. After floatation the content of pyrochlore in the concentrate is about 96%. Table IL depicts the radioactivity concentrations in the residues generated in each step of the operational process. Table L shows the radioactivity fluxes generated in each step of the operational process.

Subsequently, the pyrochlore concentrate is transferred to the calcinations unit and leached with 5% HCl in order to remove impurities such as phosphorus, sulphur and lead. In this step the solubilization of barium is observed. Afterwards, barium is precipitated by means of adding sulphuric acid to the process. The precipitated material is then disposed of in a settling pond.

The concentrate from the leaching process is sent to the metallurgical unit for aluminothermic reduction. The recovery of niobium in this step is about 95% efficient. The slags are removed from the unit by trucks and deposited in a provisional pond.

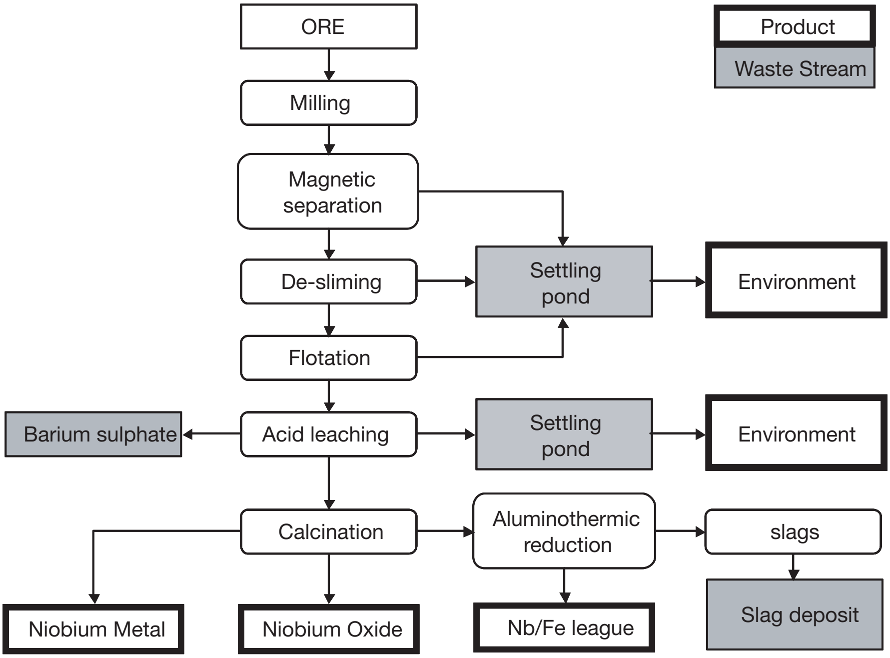

FIG. 44. Operational flow chart of Niobium Industry I.

TABLE IL. RADIOLOGICAL CHARACTERIZATION OF RESIDUES GENERATED IN THE OPERATIONAL PROCESSES OF NIOBIUM INDUSTRY I
(data from Ref. [317])

Operational step/residue	Activity concentration ($Bq \cdot kg^{-1}$)				
	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Ore	930	805	1 330	6 390	5 176
Magnetic separation	1 091	340	720	863	750
De-sliming	1 265	1 381	2 029	8 996	5 278
Floatation	770	990	2 440	3 100	1 440
Pyrochlore concentrate	14 620	1 370	710	83 000	8 900
Barium sulphate	42	26 250	7 400	43	197 300
Slags	23 138	3 290	2 400	117 600	20 200

The radiological impacts associated with this activity can be divided into occupational and environmental exposures (operational and post-operational scenarios). In the first case the main aspects are associated with external exposure to gamma radiation in the case of the slags, where exposure rates of about 4.0 mR/h have been recorded. Operational environmental impacts associated with discharges of liquid effluents into the environment can be considered negligible. However, the contamination of groundwater by ^{226}Ra and ^{228}Ra in the area of influence of the settling ponds containing barium sulphate precipitate is a relevant issue. Typical concentrations of about 5.0 Bq/L for each of the above mentioned radionuclides have been observed. In this case, the

TABLE L. RADIOACTIVITY FLUXES GENERATED IN THE OPERATIONAL PROCESSES OF NIOBIUM INDUSTRY I
(data from Ref. [317])

Operational step/residue	Radioctivity fluxes (10^{12} $Bq \cdot a^{-1}$)				
	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Ore	1.1	0.98	1.6	7.8	6.3
Magnetic separation	2.0	0.063	0.13	0.16	0.14
De-sliming	0.18	0.20	0.29	1.3	0.77
Floatation	0.58	0.75	1.8	2.3	1.1
Barium sulphate	0.00021	0.13	0.037	0.00022	0.99
Slags	0.58	0.083	0.061	3.0	0.51

adopted management strategy/mitigation used by the company is to pump the water from the aquifer and treat it before release into the environment. If it is assumed that an individual from the general public drinks groundwater at a rate of 2 L/d, the effective dose is estimated to be about 2.0 mSv/a.

In terms of future (potential) exposures, the main concern is the use of some of the operational process residues as building material in the foundation of houses. Effective doses of about 400 mSv/a are predicted for the slags and about 50 mSv/a in the case of the use of waste rock and the residues deposited in other settling ponds. The main contributor to the total dose would be external exposure to gamma radiation (especially ^{232}Th daughters), accounting for about 50% of the total dose in the case that slags are used for the above mentioned purposes. It is important to observe that at time T_0 the dose attributable to ^{232}Th daughters will be approximately zero and will increase up to 250 mSv/a after 50 years. The main issue here is that the exposures to be observed in the long term have to be considered carefully. Besides external exposure to gamma radiation, the exposure associated with the inhalation of radon will also be of concern, amounting to about 100 mSv/a, this value being constant along the simulated period of time, i.e. 1000 years.

The company concerned is taking care to bury the residues properly to avoid the general public having access to them in the future, as well as to avoid undue external exposures to gamma radiation in the case of the site being occupied. The main concern is finding a permanent solution to the avoidance of groundwater contamination by radium isotopes once the pumping and treatment strategy cannot be considered as a permanent solution. Remediation may include removal of the material and isolation of the floor of the settling pond with some sort of impermeable/semi-impermeable liner, or reduction of the infiltration of rainwater through the deposited residues. The niobium mineralization utilized by Niobium Industry II is the result of a residual enrichment, caused by the leaching of carbonatites, which have apatite and pyrochlore as main constituents. The niobium reserves are estimated as being of about 20 Mt with an ore grade above 1.3% in Nb_2O_5 or 40 Mt with the ore grade above 0.5%.

IV.3. NIOBIUM INDUSTRY II

Unlike Niobium Industry I, this industry produces Fe–Nb league mainly for export to be used in the production of speciality steels. All the residues generated in the process are disposed of in only one tailings dam. In the past, the slags used to be disposed of in blocks in a separated ore. The ore is mined in an opencast pit. Drainage water from the mine pits is directly discharged to the open environment. The waste rock is deposited in a waste rock dump.

The operational process is similar to that of Niobium Industry I and is shown in Fig. 45. As the niobium ore content is lower than that of Niobium Industry I, there is an additional step that includes the grinding of the slags to recover the niobium still present in this material. Table LI shows the activity concentrations of different radionuclides in the materials generated in the operational process and Table LII the radioactivity fluxes.

Unlike in Niobium Industry I, the uranium content is higher than that of thorium. This finding has very important implications in terms of radiological impacts and also on the possible management of residues, as will be discussed later. It also illustrates that any projection to be made in terms of TE-NORM generation in the mining industry has to be done on a case by case basis even

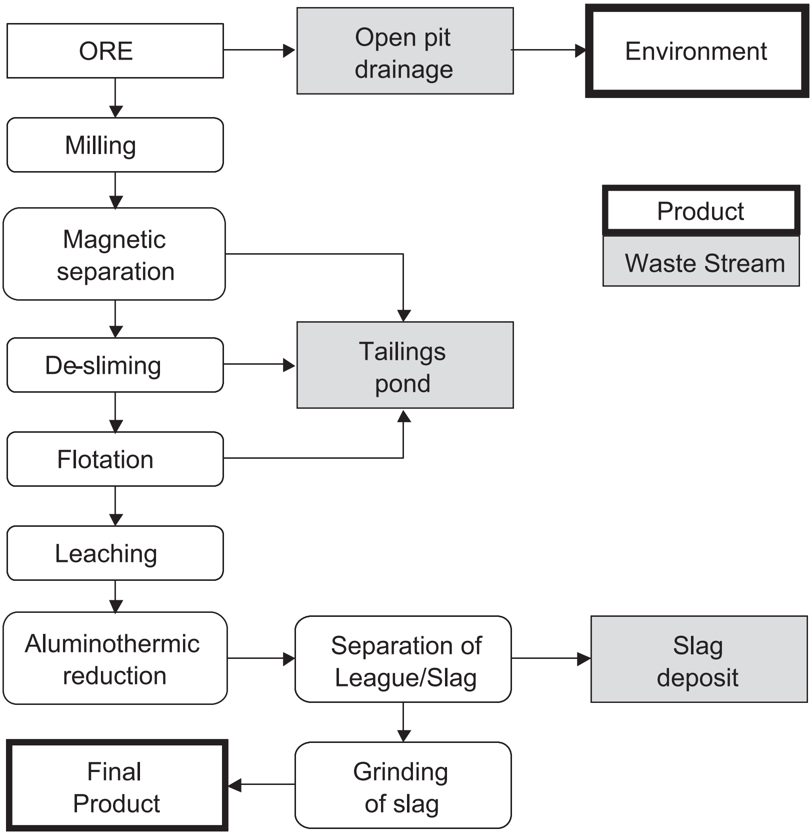

FIG. 45. Operational flow chart of Niobium Industry II.

TABLE LI. RADIOLOGICAL CHARACTERIZATION OF RESIDUES GENERATED IN THE OPERATIONAL PROCESSES IN NIOBIUM INDUSTRY II
(data from Ref. [317])

Operational step/residue	Activity concentration ($Bq \cdot kg^{-1}$)				
	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Ore	4 550	3 390	7 890	904	2 040
Magnetic separation	843	941	1 730	293	315
De-sliming	6 700	6 900	6 180	1 754	3 080
Floatation	4 985	3 400	7 530	1 040	1 840
Slags	34 819	5 160	435	16 700	6 450
Final product	64	41	—	23	258

if the metal to be extracted is the same and also if the ore mineral is of identical composition.

In a broad way it can be said that at least 60% of the total activity of each individual radionuclide leaves the operational process along with the residues generated in the floatation step. In the case of ^{238}U this figure reaches 70% of the total. This finding has a very relevant bearing on the economic recovery of uranium.

On the basis of the same assumptions as were considered in the case of the use of the process residues for landfill and the subsequent construction of houses on such sites, the predicted doses in this case will show quite different

TABLE LII. RADIOACTIVITY FLUXES GENERATED IN THE OPERATIONAL PROCESSES OF NIOBIUM INDUSTRY II
(data from Ref. [317])

Operational step/residue	Radioctivity fluxes (10^{12} Bq/a)				
	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Ore	2.5	1.9	4.3	0.49	1.1
Magnetic separation	0.092	0.10	0.19	0.032	0.034
De-sliming	0.45	0.46	0.42	0.12	0.21
Floatation	1.8	1.2	2.7	0.38	0.66
Slags	2.4	0.036	0.0030	0.12	0.44
Final product	0.00025	0.00016	—	0.000088	0.0015

patterns. Initially they will be lower than those of the Niobium Industry I study case — of about 100 mSv/a. However, after 100 years the dose related to the use of the slags begins to increase, reaching a maximum of about 800 mSv/a after 5000 years, while the doses associated with the tailings and waste rock materials remain constant for 1000 years, beginning to decrease after that.

In the case of slags, the external gamma irradiation represents 34% of the total dose while exposure to radon is responsible for approximately 66% of the total dose in time T_0. As time passes the contribution of radon (inhalation) increases due to the ^{230}Th decay.

Liquid effluent release at the present time does not constitute a major concern if only river water consumption by cattle is considered. In this situation the estimated doses would be negligible (below the exemption levels of 10 mSv/a as stipulated in Ref. [282]).

However, if other exposure pathways are introduced in the assessment (such as the use of river water for irrigation) the estimated doses can reach values up to 0.9 mSv/a.

Consideration of the exposure of the workforce [318] showed that niobium concentrations in faeces and urine samples from workers were 42.4 ± 39.6 and 1.34 ± 0.45 mg/d, respectively. The average niobium concentrations in faeces and urine samples from the control group were 3.4 ± 3.2 and 0.60 ± 0.45 mg/d, respectively. The amounts of niobium excreted by the workers were significantly different from the control group. The niobium concentration in faeces samples from workers suggests that the workers inhale coarse particles and ingest mineral dust particles, probably as they touch their mouths with dirty hands. The niobium concentrations in urine samples indicate that there is systemic incorporation of the element.

Thorium was present at all stages of the process, with average concentrations of less than 0.5 mg/m^3 and with a mass medium aerodynamic diameter from 0.8 to 14 mm. All workers presented measurable thorium concentrations in their faeces. However, the results did not correlate with the personal air sample data. The authors concluded that the thorium excreted in faeces is not a consequence of inhalation exposures during work. On the other hand, the amounts of uranium excreted in the faeces of workers were significantly higher than the average concentrations excreted by the members of the control group. Workers involved in mining operations presented the highest uranium concentrations in faeces, 110 mBq/g_{ash}, the lowest concentration values being reported for the personnel involved in ore crushing operations, about 20 mBq/g_{ash}.

IV.4. SUMMARY

The niobium industries cited in this report did not show any relevant environmental radiological impact due to the release of effluents into the environment. The main concern relates to the potential use of some of the process residues as landfill for house construction, a use for which very high effective doses may be observed. The contamination of groundwater, as was reported for one of the industries, is also of concern, when people have direct access (ingestion) to this water. Exposures (external gamma irradiation) of the workforce to radiation were relevant in the case of workers manipulating the slags of the metallurgical process. In terms of inhalation and ingestion of radionuclides by the workforce, the doses estimated did not seem to be of major concern.

Adequate management of residues so as to avoid undue exposure (use for building purposes) is envisaged in future scenarios.

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GLOSSARY

Please note that some of the definitions given below are broader than those given in the current IAEA Waste Safety and Waste Management Glossaries in order to accommodate the specific subject matter considered in this report.

amang. Malay term for tin ore processing tailings.

barren. Said of rock devoid of ore.

by-product. Any product from an (industrial) process that is not the intended primary product, but inevitable given the process implemented.

comminution. The mechanical process of rock breakage, excluding blasting, as ore is mined and prepared for mineral processing.

gangue. Those minerals in an ore body that are not the ore itself.

industrial process. This term is used very broadly in the present report to denote any human activity involving the application of technology, for example the mining, processing and drinking water treatment industries.

LCA. Life cycle analysis, a systematic set of procedures for compiling and examining the inputs and outputs of materials and energy, and the associated environmental impacts directly attributable to the functioning of a product or service system throughout its life cycle.

MFA. Material flow accounting or analysis, a method whereby the streams of, for example, materials, chemical elements and energy are assessed and possibly balanced. It is centred on the material/chemical compound, rather than on the product or service like LCA. MFA covers approaches such as substance flow analysis (SFA), product flow accounting, material balancing and overall material flow accounts.

milling (of ore). Understood here to include all the processes necessary to remove the metal sought from the ore.

NOR. Naturally occurring radionuclide.

NORM. Naturally occurring radioactive material.

overburden. Any rock or soil above the vein, seam or stratum of material of interest that has to be (partially) removed to provide access.

product. The intended output from an (industrial) process.

pregnant. In solution mining and in milling, the solution containing the dissolved metals, etc., of interest.

protore. Rock containing subeconomic material from which economic mineral deposits may form by geological concentration processes such as supergene enrichment. A protore may become profitable with technological advance or change in market value.

REE. Rare earth elements (or lanthanides), comprising 16 chemical elements, including those with atomic numbers 57 (lanthanum) through 71 (lutetium), as well as yttrium (atomic number 39).

SFA. Substance flow analysis. Similar to MFA.

tailings. Residues of a milling process that usually involves grinding, sieving, flotation and/or other physical or chemical treatment and is intended to remove as much as is feasible of the target mineral or chemical compound. Tailings consist of gangue and may include process water, process chemicals and portions of unremoved minerals and chemical compounds.

TE-NORM. Technologically enhanced naturally occurring radioactive materials.

UNEP. United Nations Environment Programme.

waste. Any material that is:

(a) The unwanted/substandard output from an (industrial) process that cannot be utilized under given circumstances (technological/economic perspective);
(b) Declared as unwanted and/or unusable (regulatory perspective).

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