IAEA-TECDOC-1718

Characterization of Swelling Clays as Components of the Engineered Barrier System for Geological Repositories

Results of an IAEA Coordinated Research Project 2002–2007

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CHARACTERIZATION OF SWELLING CLAYS AS COMPONENTS OF THE ENGINEERED BARRIER SYSTEM FOR GEOLOGICAL REPOSITORIES

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FOREWORD

At the request of the Member States, the IAEA coordinates research into subjects of common interest in the context of the peaceful application of nuclear technology. The coordinated research projects (CRPs) are intended to promote knowledge and technology transfer between Member States and are largely focused on subjects of prime interest to the international nuclear community.

This report presents the results of a CRP carried out between 2002 and 2007 on the subject of swelling clays proposed for use as a component in the engineered barrier system (EBS) of the multibarrier concept for disposal of radioactive waste.

In 2002, under the auspices of the IAEA, a number of Member States came together to form a Network of Centres of Excellence on Training in and Demonstration of Waste Disposal Technologies in Underground Research Facilities (URF Network). This network identified the general subject of the application of high swelling clays to seal repositories for radioactive waste, with specific emphasis on the isolation of high level radioactive waste from the biosphere, as being suitable for a CRP.

Existing concepts for geological repositories for high level radioactive waste and spent nuclear fuel require the use of EBSs to ensure effective isolation of the radioactive waste. There are two major materials proposed for use in the EBS, swelling clay based materials and cementitious/concrete materials. These materials will be placed between the perimeter of the excavation and the waste container to fill the existing gap and ensure isolation of the waste within the canister (also referred to as a container in some EBS concepts) by supporting safety through retardation and confinement. Cementitious materials are industrially manufactured to consistent standards and are readily available in most locations and therefore their evaluation is of less value to Member States than that of swelling clays. There exists a considerable range of programme development regarding qualification of swelling clay materials. Some Member States with advanced repository concepts have identified potentially suitable swelling clay materials for use in these underground facilities. Given the quantities of clay material needed for a repository application, the identification of potentially suitable materials located within or near to Member States is of considerable interest, particularly for those with more limited resources to apply to their repository programmes.

This report provides:

— An overview of the repository concepts of the Member States involved in the CRP;

— A general description of key material properties and a means of assessing those properties of relevance to screening potentially suitable swelling clays for repository use;

— A summary of the properties measured for the candidate materials selected by each of the participating Member States and a preliminary assessment of their potential suitability for use as a component of the EBS of a deep geological repository.

Overall, it is concluded that the major technical goal to assist in the identification of suitable clay deposits in Member States has been achieved. Moreover, by participating in the CRP the Member States appear to have advanced their programmes in a number of other ways. Specifically, by their involvement, the participants have joined and enlarged the international group of experts able to assist in the safe disposal of high level radioactive waste.

This publication was developed with the assistance of Member State representatives to this CRP through several RCMs, written contributions by these individuals and regular communications between the participants. The IAEA wishes to express its appreciation to all those who were involved in the preparation of the report and its revision. Special mention is due to D. Dixon (Canada) and S. Hossain (Germany) for finalizing the publication. The IAEA officers responsible for this publication were M. Gray (also CRP project officer), B. Neerdael and P. Degnan of the Division of Nuclear Fuel Cycle and Waste Technology.

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CONTENTS

EXECUTIVE SUMMARY

An uneven level of technical experience related to the sealing of underground repositories for radioactive waste exists between several Member States (MSs). Some programmes are well-advanced with regards to repository development for deep geological disposal of High Level Radioactive Waste while many other MSs are just beginning to consider this option for safely isolating used or reprocessed reactor fuel.

An important component to the development of a repository that will meet the safety and licensing requirements for a DGR in many geologic media is the development of an engineered barrier system (EBS) that through its design and the properties of its components will contribute to keeping the facility isolated from the biosphere. A commonly identified component of the EBS is swelling clay, used to ensure that any openings generated by repository excavation do not become preferential pathways for contaminant migration.

MS with relatively advanced DGR concepts or programmes have spent approximately 30 years characterising candidate swelling clay materials for repository application and then undertaking full-scale tests to evaluate EBS performance under simulated repository conditions. This has required development of special underground research laboratories (URLs) and substantial expense in development of personnel and technologies for testing of the EBS. MS with limited quantities of HLW are unlikely to be able to afford to develop such facilities and many are still in the earliest stages of identifying domestic sources of swelling clays for repository use.

In order to facilitate and accelerate the process of swelling clay identification, characterisation and evaluation for potential suitability for DGR applications, the IAEA established a CRP that brought together specialists from MS with advanced EBS development programmes with representatives from MS with less advanced programmes. The participants in this CRP have worked through the initial stages of swelling clay identification, characterisation and qualification as it relates to use in an EBS. This was accomplished via a co-operative process of materials testing and information exchange, whereby the participating MS utilized their technical expertise to complete a preliminary assessment of previously un- or incompletely-characterised candidate clays being considered for EBS use. The CRP resulted in identification of several potential deposits of swelling clay that have the potential for use as components of the EBS. Some of these materials are potentially suitable for use in an SKB-type highly compacted bentonite buffer while others may be more suitable for use in tunnel and shaft backfilling applications.

1. INTRODUCTION

1.1. Background

The Network of Centres of Excellence on Training in and Demonstration of Waste Disposal Technologies in Underground Research Facilities (URF Network) identified the general subject of the application of highly swelling clays to seal repositories for radioactive waste with specific emphasis on the isolation of high level radioactive waste from the biosphere as suitable for an IAEA Coordinated Research Programme (CRP). Several Member States with advanced repository concepts have proposed that high level waste should be disposed in specially built repositories excavated in deep geological strata. This approach to disposal had become the accepted and internationally preferred method. Swelling clays are proposed for use in closing and sealing the openings made for the operation of the facility. Swelling capacity combined with low intrinsic hydraulic conductivity allows for efficient and long term sealing of the openings. Moreover, the fine grained, highly surface active character of the clays provides them with an ability to limit the rate of radionuclide contaminant transport through them as a result of their physical-chemical properties and processes associated with them.

Proposals for participation in the CRP were received from twenty-eight Member States. Considering the funding limitations, the technical quality of the proposals and administrative factors, eventually, proposals from the following MSs were accepted for IAEA support: Canada, China, Czech Republic, India, Japan, Russian Federation, Republic of Korea, Sweden, South Africa and Ukraine.

The participating MSs can be classified in accordance with the degree of development of their national radioactive waste management programmes. Canada, Japan, Republic of Korea and Sweden each have advanced repository programmes and offered leadership to the group. The repository programmes of China, Czech Republic, India, Russian Federation, South Africa and Ukraine, although less advanced regarding repository and sealing materials selection, are moving forward with their plans for repository design. These Member States provided expertise for inclusion in the CRP and, it was judged, could benefit significantly from association with the project.

It was considered that the work would lead to a verified approach for materials screening and selection for repository construction that could be applied not just by the MSs forming the CRP but also by all Member States that intend to develop repositories that will include use of swelling clays as part of their engineered barrier system.

1.2. Project objectives, scope and structure of report

Although the general subject of the CRP (i.e. Swelling Clays) was pre-defined at the time of establishing the project, it was necessary to accommodate the wide range of status regarding repository design and the variety of expertise represented by the individuals who formed the research team. Based on this it was necessary to develop specific technical objectives for the work that could be achieved within the time available for this project. After a period of discussion and evaluation of the priorities of the various MSs, the research team decided that the objectives of the CRP would be as follows:

- To transfer knowledge regarding the repository concepts and approaches being considered by the participating Member States through preparation of an overview of the repository concepts considered by each Member State involved in the CRP;

- To undertake cooperative laboratory work and clay properties collection that would allow for a general description of key material properties and means of assessing these properties that are of relevance to screening potentially suitable swelling clays for repository use;
- To develop a summary of the materials properties measured for the candidate materials selected by each of the participating MSs and provide a preliminary assessment of their potential suitability for use as a component of the engineered barrier system of a deep geological repository.

Based on the above listed objectives and general scope agreed to by the research team Member States would best benefit if the reporting of this work was structured as follows:

- The repository concepts being developed in each of the participating MSs have been reviewed and summarized. Important developments being made in MSs that were not part of the CRP were briefly examined for relevance. Common aspects of the national programmes, with particular emphasis on the use of swelling clay in engineered barriers are identified. This review also includes a brief summary of the technological approaches being considered for installing waste packages and sealing materials in a repository for high level nuclear waste (see chapter 2).
- A common set of definitions related to swelling clays has been developed as they relate to the specific application of radioactive waste disposal. Relatively simple methods to identify and characterize the swelling clays were developed as they apply to the specific task of screening clays that have potential for use in repository sealing. (see chapter 3)
- The generic evolution of a repository was defined and the properties of the clays relating to determining the engineering performance of candidate swelling clays and their long term behaviour when used in repositories as seals examined. These aspects of behaviour apply, in general terms, to the initiation of a process of evaluation and qualification of swelling clay materials for repository use and subsequently safety assessments of repositories (see chapter 4).
- The screening techniques developed within the project have been tested by application to materials identified through searches for suitable materials within China, Czech Republic, India, Republic of Korea, Russian Federation, South Africa, and Ukraine. A preliminary assessment of the characteristics of the candidate materials provided by the CRP participants was developed. This assessment is based on comparison of candidate clays to well characterized swelling clays proposed for use by MSs with more advanced repository programmes (see chapter 5).
- A brief comparison of the characteristics of each of the candidate clays relative to the MX-80 reference material has been provided (see chapter 6).

2. REPOSITORY DESIGN CONCEPTS, NEEDS AND PRACTICALITIES

2.1. Management and repository concept descriptions

Each of the national programmes represented in this report is examining potential approaches to nuclear waste isolation in geological media. Each has its own unique set of materials to be emplaced, system requirements, constraints and solutions that must be considered when undertaking a safety assessment and subsequently, requesting a licence for construction and operation of a nuclear waste repository. Common to all these programmes is the need to find a means of safely isolating the waste product(s) in various geological media for long enough that the material(s) no longer pose a threat to the biosphere. In order to accomplish this, most nuclear waste isolation concepts include the use of both a deep geological environment and engineered barrier systems (EBSs) intended to keep these nuclear materials isolated in the repository. Common to all of the concepts examined in this section is the use of swelling clay as a component in one or more of the engineered barriers proposed for use in sealing a repository.

Several approaches are being considered for the management of nuclear fuel waste - used fuel (UF) or reprocessing waste - by various national programmes. This section briefly outlines the direction that each of the participating organizations is taking in managing these radioactive materials. These programmes and their status are briefly described below in alphabetical order by national name. Additional information on the repository concepts of several IAEA Member States can be found in a recent IAEA document [1].

2.1.1. Canada

Canada has been using nuclear technology to generate electrical power for more than 40 years and so has been accumulating used nuclear fuel for this entire period. The current practice in Canada is to store the used nuclear fuel in water-filled pools and then after approximately ten years, move the bundles into steel lined concrete dry storage containers at the various nuclear reactor sites. This approach has proven safe but requires continuous control (physical security, monitoring and storage system maintenance) and has not dealt with ultimate disposition. In an effort to develop a means of dealing with the long term management of UF the concept of emplacing it and other high level waste (HLW) in a deep geological repository (DGR) within the stable plutonic rock of the Canadian Shield was examined [2, 3].

The generic Canadian DGR concept consists of a series of multiple engineered barriers that include the waste form (i.e. currently used CANDU® reactor fuel bundles), a corrosion resistant container (many other programmes refer to this as a canister and for the purposes of consistency, the term canister will be used in this document), and a repository sealing system that includes use of an engineered barrier system (EBS) that contains swelling clay [2]. At present Canada is not considering reprocessing the used nuclear fuel but disposing of the used fuel bundles directly. The DGR repository concept in crystalline rock involves installing the durable canisters at a depth of between 500 to 1000 m in the stable plutonic rock of the Canadian Shield and then sealing the repository. Sedimentary rock formations may also be a suitable DGR host medium [4] and is currently being studied in other countries (e.g. Belgium, France, Japan, Switzerland) with the Canadian programme recently becoming active in examining this option [3]. An illustration of the current generic DGR conceptual design options under consideration in Canada is shown in Fig. 2.1.

The canister emplacement geometries shown in Fig. 2.1 all utilize similarly dimensioned canisters but reflect some of the options available for their installation. These geometries represent variations of the Swedish in-floor (KBS-3V) and horizontal borehole (KBS-3H) emplacement concepts (upper and lower sketches), as well as an in-room option being considered. Each of these options has operational advantages and disadvantages associated with it and final selection will come once safety assessment and licensing is completed and a geological medium is selected. Clay based sealing materials would be used to surround the used fuel canisters and would completely fill all remaining voids in each emplacement room to ensure low permeability and that suitable chemical and biological conditions are maintained within the repository. Each of these sealing and backfilling materials must be compatible with the other system components (canister, rock, concrete). Beyond the regions shown in Fig. 2.1 there are additional materials required to seal the access shafts and isolate hydraulically active regions from the repository. Swelling clay materials are expected to play an important role in achieving this goal, thereby isolating the repository from the surrounding geosphere and biosphere.

Ultimately, after all the emplacement rooms are filled and sealed but before all access shafts and tunnels are sealed, a period of monitoring would occur to confirm that the facility is performing as expected. During this monitoring period, the repository and its surroundings would be experiencing heat and water movement. After a sufficient period of post-operational monitoring to confirm that the system is evolving satisfactorily, the remaining excavations would be backfilled and sealed such that long term safety of the facility would not depend on institutional control. This generic repository concept also includes extended monitoring of the repository and the potential to retrieve the used fuel canisters, if required for any reason prior to final facility closure.

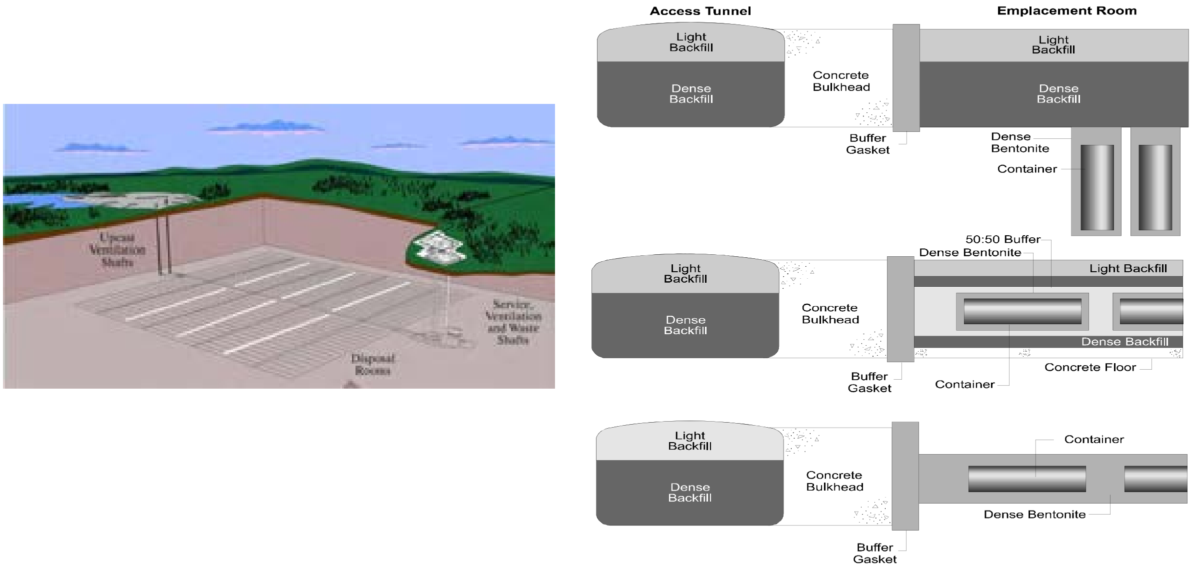


FIG. 2.1. Canadian repository and emplacement concepts [2-4].

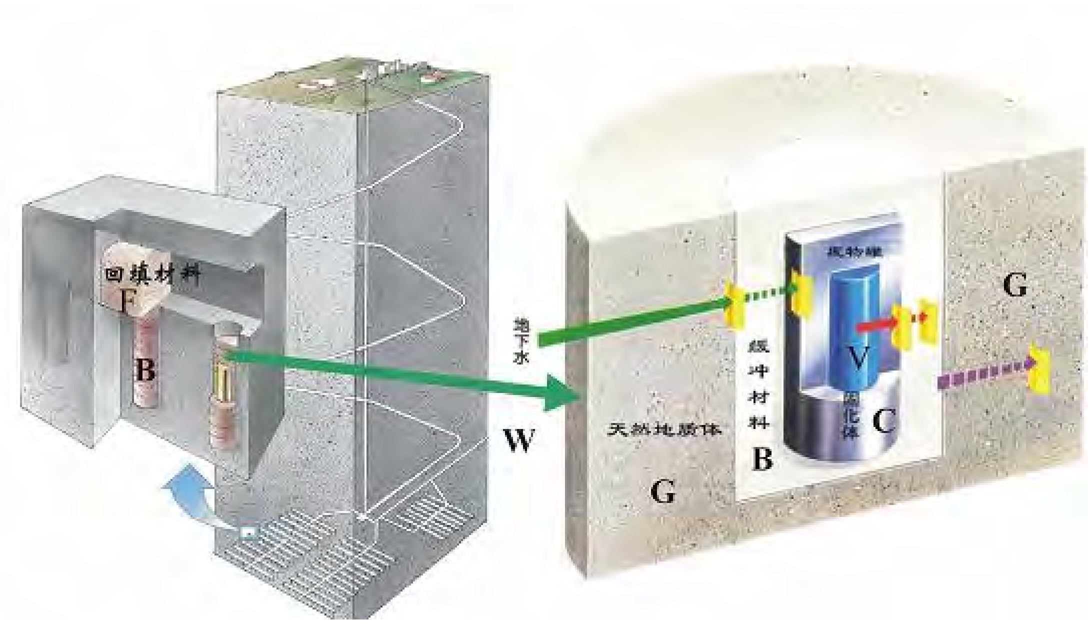
2.1.2. China

China is in the process of rapidly expanding its nuclear power generation capacity. In association with such expansion will be the generation of considerable quantities of radioactive waste material including used reactor fuel. At present it is estimated that the 1000 tons of UF will be generated by 2010, increasing to 2000 tons by 2015. Beyond 2020, about 1000 tons of UF will be produced each year. With such a rapid development of the nuclear industry, the development of final safe disposal of high level radioactive waste is an important aspect in sustainable development of the nuclear industry in China.

In China, according to the national high level radioactive waste management policy, the UF in China will be reprocessed first, followed by vitrification and final geological disposal. Similar to the conceptual approach to HLW waste disposal in many countries, the repository concept currently adopted by China is a shaft-tunnel system, located in saturated zone in granite. The generic repository concept anticipated to be adopted by China is similar to that of many other nations in that it involves installation of canisters in boreholes drilled in the floor of tunnels located deep underground (Generic KBS-3V geometry) as shown schematically in Fig. 2.2 [5].

The approach of selecting a site that could become the repository, based on common siting of an underground research laboratory (URL) and the repository, has been adopted in China. The result is a 3-step strategy for repository development:

(1) Site selection and site characterization both for the URL and the potential high level waste (HLW) repository;
(2) Construction of a URL at the site selected as a potential repository;
(3) Construction of final repository if appropriate URL results are achieved.



B: Buffer; C: Canister; G: Geosphere; F: Tunnel backfill; V: Vitrified HLW;
W: direction of groundwater movement shown by green arrows

FIG. 2.2. HLW disposal concept considered in China [5].

A long term plan has been developed for HLW disposal in China and is based on the expected growth in the need for disposal of reprocessing wastes generated. At present it is expected that by 2050 a repository site will have been identified and constructed in China [6]. The current target schedule for this repository siting and construction process is:

- 2006–2020: sitting and site evaluation, site confirmation, complete site characterization and feasibility study of URL, conceptual design of repository;
- 2021–2040: construct and test of URL, in situ test and demonstration on disposal in URL, detailed design and construction of repository;
- 2041–2050: complete construction of repository.

The repository is to be sealed through use of an engineered barrier system (EBS), including a solid waste form, an overpack and fill materials containing swelling clay (buffer and backfill) placed between the waste package and the surrounding granite. China has many large bentonite deposits within its borders but these materials have typically not been exploited commercially and so their properties and composition are currently not well enough defined to allow for the definition of a reference material. A screening process using the approach developed in this CRP is now being applied in an effort to identify potentially suitable domestic materials for repository utilization.

2.1.3. Czech Republic

A concept for radioactive waste and UF management in Czech Republic has been approved by the Czech government on 15 May 2002 [7, 8]. The Czech concept for radioactive waste disposal includes all levels of radioactive materials and follows two basic principles:

- Firstly, that radioactive waste and used nuclear fuel management in Czech Republic is and will continue to be provided by licensed private entities (collection, processing, storage and transportation) under the direction of the State-owned organization the Radioactive Waste Repository Authority (RAWRA). Ultimately, through disposal of existing radioactive waste, future reprocessing or disposal of UF RAWRA is supposed to be able to provide extended services to generators, if required.
- Secondly, that Low level radioactive waste (LLW) produced in Czech Republic will be safely disposed of at existing near surface repositories, whose safe operation will be assessed and optimized on a continual basis.

One method being considered for the disposal of all of these waste forms involves their placement in a deep geological repository. Before such a repository is put into operation, waste materials will be stored at the waste producer’s site. The technical aspects associated with the disposal of radioactive waste and the preparation of a deep geological disposal site will utilize the outcomes of foreign and domestic research and technical development programmes. The main objective of deep geological disposal of high level waste or used nuclear fuel is to provide for the permanent isolation of such materials from the environment without the intention of later removal. The principle of the deep geological repository is based on passive safety (i.e. no further human supervision). The repository system consists of a number of barriers, i.e. a combination of engineered (EBS) and natural (geological) barriers shown conceptually in Fig. 2.3. In addition, options for UF reprocessing and the employment of new techniques that will result in lower volumes or reduced toxicity of UF will be pursued but ultimately some volume of waste material will need to be disposed of.

The Czech repository concept includes the use of clay based materials as part of the EBS and so identification and qualification of domestic sources of potentially suitable swelling clay materials is of interest. Sources of domestic bentonite have been identified and a process of characterization and evaluation to determine their potential suitability for repository application has been initiated.

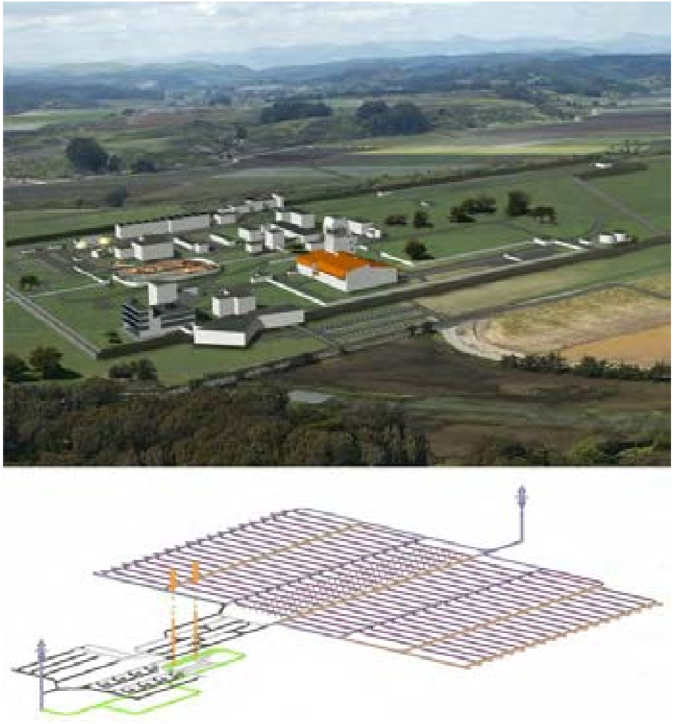


FIG. 2.3. Concept for deep geological disposal in Czech Republic [9].

The project for the building and operation of a DGR within Czech Republic will be carried out stepwise, beginning with consideration of the potential for construction of new nuclear energy sources. Based on this review the need for construction of repositories for UF and HLW as well as a location for the disposal of radioactive materials other than HLW will be assessed. The timeframe for a deep geological repository in Czech Republic is based on three stages:

- Firstly, up to 2015 will be used to select two potential site for DGR and approve one in 2025 after successful completion of the safety assessment and licensing processes;
- The period between approximately 2050 and 2065 will be for the construction of surface and underground facilities;
- The final phase, beginning in about 2065 will be the start of repository operations.

2.1.4. India

The environment and achieving radiation protection goals have been among the major concerns in the planning of India’s nuclear energy programme. In the Indian nuclear fuel cycle, right from inception, a closed loop option has been adopted where UF is reprocessed to recover plutonium and unused uranium for reuse as reactor fuel.

A three-step strategy for management of HLW has been adopted in India and is shown in Fig. 2.4 [10]. This strategy involves:

- Immobilization of waste oxides in stable and inert solid matrices;
- Interim storage and surveillance of the conditioned waste under continuous cooling;
- Disposal in deep geological formations.

The DGR concept adopted by India envisages emplacement of vitrified wastes at depths of about 500 to 600 m in appropriate host rocks, e.g. granite, granite gneisses, charnockite, basalt or other suitable geological media. The DGR as well as near surface disposal facilities (NSDF) concept include multi-barrier systems consisting of waste form, canisters, and swelling clay based sealing materials. Figure 2.4 also shows the generic emplacement concept being considered by India. The swelling clay-based materials are expected to play a very important role in retarding the migration of radionuclides by delaying the contact with ground water and through various radionuclide sorption processes. The swelling characteristics of buffer and backfill material constructed using swelling clay as a component are also expected to enable sealing of block fractures, joints and other pathways that may be present in the engineered barrier system. In addition, due to its low permeability swelling clays may reduce the rate of oxygen supply to the canister surface, thereby reducing the rate of corrosion [11].

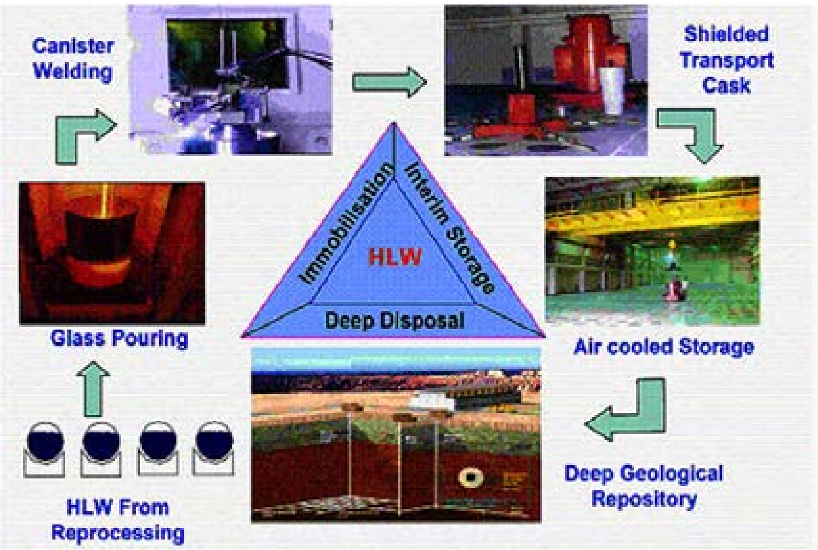


FIG. 2.4. The three stages of waste management that include the in-floor borehole emplacement concept being considered for use by India [10].

With the importance of the swelling clay component in the sealing system, the characterization and demonstration of efficacy of swelling clays in repository condition forms an integral part of studies ensuring the safe disposal of HLW. Associated with this demonstration process is a need to be able to effectively screen and qualify potential sources of swelling clay for their potential for use in a repository.

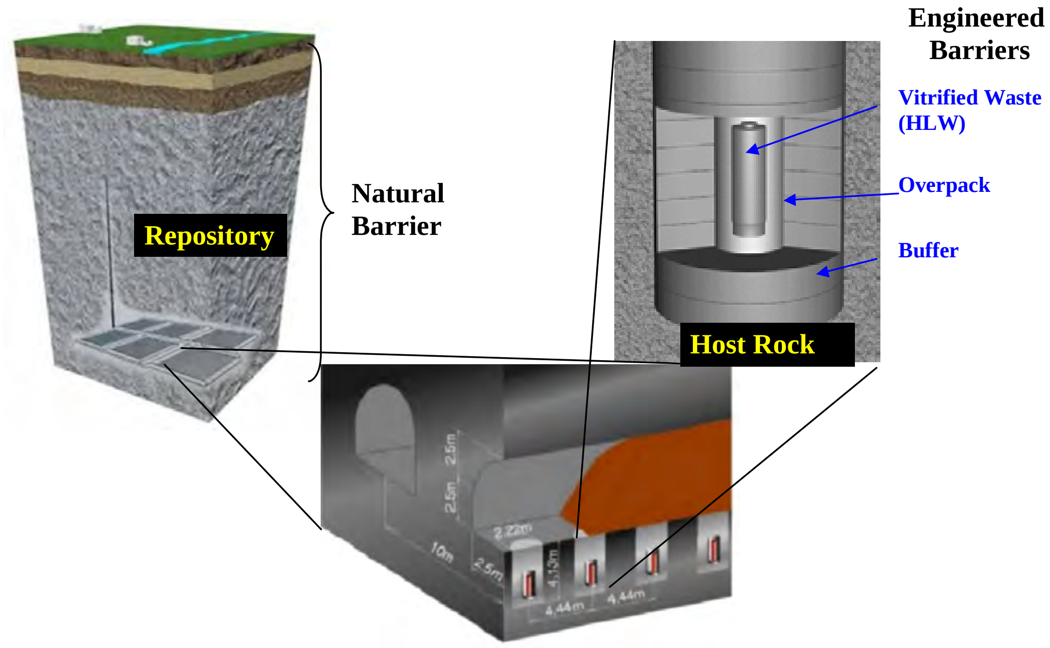
2.1.5. Japan

To ensure efficient use of resources, Japan has adopted a policy of fuel reprocessing. Once reprocessed, the resulting HLW has to be disposed of safely and reliably. After an appropriate period of cooling, the concept for HLW disposal in Japan calls for the disposal of these canisters in a stable geological formation at a depth of more than 300 m as described in JNC's H12 report [12].

The basic concept of Japanese HLW disposal presented in the H12 report is to construct a multi barrier system in a stable geological environment. The EBS consists of vitrified waste, a steel overpack that contains the vitrified waste and an engineered sealing system that includes swelling clay based sealing materials. These sealing materials are to have a very low permeability so that mass transport is diffusion controlled rather than advective. The repository will be located below the water table and will evolve towards a reducing groundwater environment. Under reducing conditions, the corrosion rate of the overpack should be slow, preventing the vitrified waste from coming into contact with water during the early stage when the radioactivity level of the vitrified waste is high.

The Japanese multi-barrier system is illustrated in Fig. 2.5. There is as yet no selected geological medium in Japan and both crystalline (granitic) and sedimentary rock types are under active consideration. At present there are Underground Research Laboratories being constructed in both granitic and sedimentary media to allow for direct evaluation of both the geological systems for suitability as a host forrmation. Neither of these sites is currently being considered as a potential repository site, and both are being used for generic research purposes only. When a potential disposal site is selected in the future, an optimized design for the engineered barrier system will be determined in the light of safety and economic considerations, taking into account the geological conditions specific to the site.

It is assumed that the steel overpack will eventually fail and groundwater will come in contact with the vitrified waste. At this stage, the release rate of radionuclides is expected to be extremely slow since it is limited by slow glass matrix dissolution. In addition, reducing conditions limit the solubility of many radionuclides. The swelling clay material also has a filter function, preventing the migration of colloids containing radioactive materials. In the rock formation surrounding the EBS, the transport of radionuclides is further limited by slow groundwater flow and interaction with the rock.



Disposal tunnels (vertical emplacement option)

FIG. 2.5. Generic disposal concept considered by Japan [12, 14].

Sodium (Na) type smectite is being considered as a candidate swelling clay material for use in Japan. In the H3 report of JNC [13], an evaluation of material properties and a performance assessment were carried out using 100 wt% clay compacted to a dry density of 1800 kg/m^3. In the subsequent H12 report of JNC, a mixture of bentonite and silica sand was considered as an alternative composition for the sealing materials. In these studies a bentonite produced and sold commercially in Japan (as Kunigel-V1) has been selected as their reference swelling clay component. This material has been extensively characterized over a 25-year period and is felt to be potentially suitable for repository application, hence its classification as a reference material. Work has continued to improve the characterization and better qualify this material as a suitable for use as the swelling clay component in JNC's EBS concept.

2.1.6. Republic of Korea

Republic of Korea has been operating nuclear power plants since 1978, with 16 currently in operation and has plans to build additional plants (5th lonterm electric power development plan, [15]). The resulting increase in the amount of UF is becoming an urgent national issue and so the development of a workable waste management concept and construction of a HLW repository has become a priority in Republic of Korea.

In 1997 the Republic of Korea Atomic Energy Research Institute (KAERI) launched a three-step R&D programme to develop a reference repository system for HLW by 2006. The first step of this programme set out a disposal concept utilizing generic information and data. In the second step, it developed a preliminary reference repository system focusing on the near field components. The last step of the programme is now devoted to the refinement of the preliminary reference repository system using more reliable information and data.

The concept for disposal of HLW in Republic of Korea is based upon a multi-barrier system composed of engineered barriers and the surrounding plutonic rock [16]. A repository is proposed for construction in bedrock of several hundred metres below the ground surface. The EBS consists of HLW disposal canisters, and barriers constructed using swelling clays in a configuration similar to that considered by many other countries (e.g. in-floor borehole geometry). In this configuration the HLW are encapsulated in disposal canisters, which are deposited into boreholes on the floor of the emplacement rooms. The gap between the canister and the wall of a borehole is then filled with a swelling clay based buffer material and the inside space of the emplacement rooms with a clay based backfill material.

The Korean Reference HLW disposal System (KRS) involves construction of disposal panels, access tunnel, and shafts within a suitable geological host medium. The various reactor wastes will be emplaced separately because of differences in the heat generation characteristics of the waste forms. Based on 40 m emplacement tunnel spacing, each Pressurized Water Reactor (PWR) UF disposal panel consists of 42 emplacement tunnels, and the Canada Deuterium-Uranium (CANDU) UF disposal panel contains 38 emplacement tunnels. The length of each emplacement tunnel is 250 m. The spacing of the boreholes will be such that neither the maximum canister surface temperature nor the maximum buffer temperature of 100°C are exceeded.

Figure 2.6 shows a schematic picture of the EBS of the KRS, which consists of disposal canister encapsulating the UF, buffer, and backfill. The dimensions of emplacement tunnel and borehole shown in this figure were based on preliminary design calculations [17].

Over a twenty-year period extensive characterization and assessment of Korean bentonite materials for suitability as buffer/backfill materials has occurred and at present they appear to be an appropriate material for use. Inclusion of bentonite from Republic of Korea in this screening study provides an opportunity to compare its behaviour and properties to other materials being considered by other international programmes.

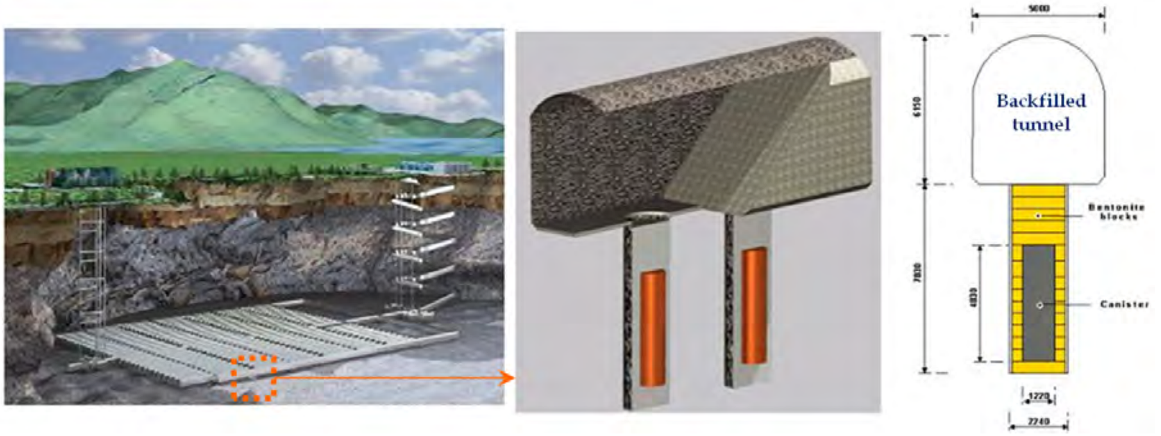


FIG. 2.6. The engineered barrier system of Republic of Korea's repository concept [16].

2.1.7. Russian Federation

The Russian State Corporation for Atomic Energy "ROSATOM" is responsible for the development of national concept(s) for HLW disposal. At present time, only three main regions have been included in the search of prospective geological sites. The location of each of these sites is shown in Fig. 2.7 and includes the Krasnoyarsk (radiochemical plant MCC), PA Mayak (radiochemical plant) and Novaya Zemlya (former nuclear test site) regions.

Historically, most of the work on locating an appropriate geological site for a DGR has been focussed in the Krasnoyarsk region near MCC. A team of scientific experts has identified Nizhnekanskiy granitoid massif in this region as the most promising site to build a repository of HLW [18]. This massif is one of the largest granitoid massifs of Middle Siberia; its outcrop area is more than 1500 km^2. It consists of granitoids of several different time phases, among which the most widespread types are biotite granites and granodiorites.

The Federal special purpose programme of Russian Federation "Management of radioactive wastes and used nuclear materials, their utilization and disposal for the years 1996–2005" (No. 1030) also included into the list of priority work a search on the Nizhnekanskiy of a site suitable for disposal of the MCC's solidified HLW. As a result of the site screening at scales of 1:100 000 to 1:25 000 in the area occupying the northern part of the Nizhnekanskiy massif and its Archean-Proterozoic structures 13 potential sites were identified, 8 of which were placed within the limits of the previously allocated promising areas. Two sites named "Itatskiy" and "Kamennyi" (about 7 km^2 each), located about 25 km from the MCC were determined as the most promising for further detailed evaluations.

FIG. 2.7. Simplified map of Russian Federation with possible locations of future HLW repositories.

While the detailed design of a HLW repository has not yet been completed and the form(s) that the immobilized HLW will be in is as yet undecided, it is anticipated that a repository will require the use of engineered barriers that will include the use of swelling clay materials. While such clay deposits are known to exist within the borders of Russian Federation, substantial commercial exploitation and development of these deposits has not yet occurred. The development of screening tools and methods of characterizing potentially suitable swelling clay materials for future use is therefore of considerable interest to Russian Federation.

ROSATOM has recently adopted the approach of developing “regional repositories” for the immobilization of medium and low level wastes. Such repositories would be built in the most developed industrial regions, in particular, near nuclear power stations or radiochemical plants. The Government of St. Petersburg is considering the creation of a local geological repository for Sosnoviy Bor Nuclear Power Plant (Leningrad region) in a formation of “blue” clays. Bentonites from the recently discovered Pikalevskoe deposit (Leningrad region) are of interest as they might provide a domestic and nearby source of swelling clay for use as a component of engineered barrier system in this repository. Established bentonite resources of the Pikalevskoye deposit are about 4 million tons but this deposit is expected to contain about 300 million tons of smectite-rich material. This Pikalevskoe deposit was therefore selected for inclusion in the screening studies described later in this document.

2.1.8. South Africa

South Africa is currently in the process of developing a national policy and strategy for radioactive waste management. A draft policy was formulated in 2003 and is currently under review. According to this draft policy, the South African Government shall establish a National Executive Committee on Radioactive Waste Management, which will oversee the implementation of this policy and strategy. The draft National policy recognizes that deep geological disposal is currently the most internationally acceptable solution for UF or HLW, apart from other management options, and as such will require very careful consideration [19].

In a formal site selection programme for geological disposal it is the view of the South African Nuclear Energy Corporation (Necsa) that a transparent, countrywide study would be required. Such a study would ideally identify a number of potential sites from which one would be selected for detailed site confirmation. Vaalputs National Radioactive Waste Disposal Facility in the Northern Cape Province is currently licensed [20] for the disposal of L/ILW and would likely be considered as one of these candidate sites because of its very favourable characteristics.

A site selection programme to locate a suitable site for the storage/disposal of radioactive waste in South Africa was originally initiated in 1978 and the selection criteria of Corner and Scott [21] led to the choice of Vaalputs in 1986 as a site for L/ILW disposal but with the recognition that it also had the potential for high level radioactive waste (HLW) disposal. This site is shown in the aerial photograph provided as Fig. 2.8 and was found to contain a volume of megacrystic granite gneiss with excellent geotechnical qualities at 200 m through 1000 m depth. This granite gneiss, dated at about 1060 Ma, has virtually no weathered zones or faults and very few joints and could potentially be a suitable geological medium for a repository. The lack of a formal national policy for radioactive waste management led to cessation of further deep geological site evaluation investigations at Vaalputs at the end of 1996.

There are a number of sources of radioactive waste materials in South Africa. The main generators of UF and other long lived waste destined for a potential HLW geological disposal facility in South Africa are the South African Nuclear Energy Corporation (Necsa) with its Safari-1 reactor for research and isotopes production and the Koeberg nuclear power station (Eskom). Beyond the conventionally derived, reactor generated waste materials, mining companies as well as the Pebble Bed Modular Reactor programme (PBMR) are also expected to utilize a future geological repository for the disposal of their long lived used fuel. Additionally considerable volumes of other long lived bulk waste and an unknown quantity of industrial and medical sources could potentially also be earmarked for geological disposal.

FIG. 2.8. Aerial photograph of the Vaalputs low to intermediate level radioactive waste disposal site.
(Photo shows the building hosting the waste receiving facility at the end of the main road to the south, and two operational trenches and associated activities to the north.)

Recently, feasibility studies were initiated in South Africa that began to review options for site selection for the deep geological disposal of UF, the encapsulation of such fuel, repository design and engineered barriers [22]. A part of this study involves the identification and characterization of available bentonite deposits in South Africa for potential use as an engineered barrier in the proposed HLW repository.

2.1.9. Sweden

Sweden has formally adopted the concept of deep geological disposal in crystalline rock as its reference method for managing its used nuclear fuel. This concept is generically known as the KBS-3V method, shown in Fig. 2.9. In the KBS-3 concept the reactor fuel is first cooled until its heat generation has reduced to a level that will allow for its installation within the repository without causing unacceptably high temperatures to develop. The fuel is installed in a canister consisting of a cast-iron insert to hold the fuel and provide mechanical strength and a copper shell that provides corrosion protection.

In the KBS-3 concept presented in report SR-97 [23, 24] and in later interim safety assessment reports [25], the approximately 1.05 m diameter by 4.835 m long copper-shelled canister would be installed in a borehole drilled into the floor of an access tunnel located approximately 500 m below the surface in a granitic rock mass. These approximately 24.6 tonne canisters would be surrounded by highly compacted bentonite clay (HCB), separating the canisters from the adjacent rock (Fig. 2.9). Once the rooms containing these boreholes were filled the rooms, tunnels, ramps, shafts and other excavations would be filled with low permeability backfill materials that also contain a bentonite clay component. Plugs and seals would then be installed in locations where they are necessary to provide support to backfilling materials or to prevent advective water movement within the repository. Variations of the emplacement geometry, mainly as a horizontal emplacement borehole (KBS-3H), similar to that shown in Fig. 2.9, extending outwards from the tunnel walls [26, 27], have also been examined as part of options and optimization exercises but no final decision has been made on the specific emplacement geometry to be used in a repository.

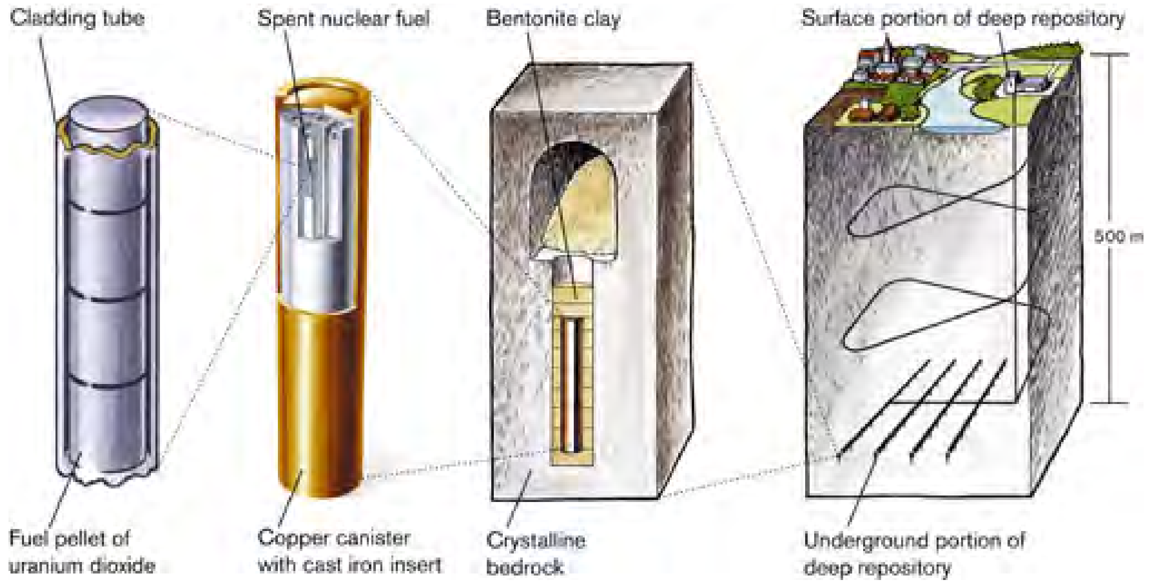


FIG. 2.9. The generic KBS-3 concept for used fuel disposal [25].

Sweden has no economically viable swelling clay deposits within its borders and so has selected a commercially available sodium bentonite produced in USA as its reference clay for concept development purposes. Over the course of nearly 30 years this material has been extensively characterized and tested both in the laboratory and in mock-ups constructed at one of the two underground research laboratories in granitic rock that have been operated during this time period. Using the extensive knowledge base developed for Wyoming bentonite (MX-80) it is expected that alternative sources of supply will be evaluated and qualified for potential use once a repository is licensed and operations are scheduled.

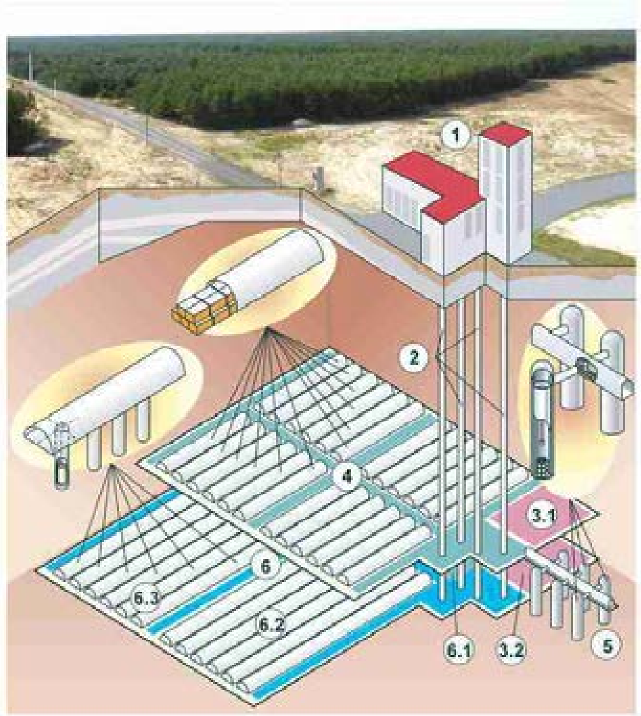
The first underground laboratory to examine HLW management issues associated with sealing was the international Stripa Project from about 1980 to 1990. The abandoned Stripa iron mine in Sweden was used to examine the hydrological and mechanical performance of the rock, and, in particular, on the function of engineered barriers, i.e. buffers, backfill, plugs and grouts. The successor facility to Stripa in Sweden is Äspö. This facility is being used to develop geological knowledge and test sealing strategies that would be useful in an actual repository site. The Äspö facility has all of the basic features anticipated to be necessary when an actual repository is constructed. Work is continuing at the Äspö site towards refining the materials performance specifications and emplacement technologies for use in repository sealing through the conduct of a number of large-scale sealing demonstrations and long term monitoring exercises. With the success of its concept development process, Sweden has moved into the siting and licensing phases of its nuclear fuel disposal programme. As of 2005, there were two regions receiving attention as potential repository sites, located in the municipalities of Osthammar and Oskarshamn. In early 2009 an announcement was made that the candidate site selected for detailed evaluation, and if found to be suitable through a safety assessment and licensing process, will be the location of the Swedish HLW repository is the Osthammar site near the Forsmark nuclear power station.

2.1.10. Ukraine

The general concept for radioactive waste isolation in geological formations in Ukraine is based on the experience of countries having more developed waste management programmes, the IAEA basic principles, and technical criteria adapted to geological, socio-economic and ecological conditions in Ukraine. Based on these considerations a law has been passed in Ukraine “On Radioactive Waste Management”. It provides for the ultimate disposal of HLW and ILW in deep geological formations. According to Article 17 of this law, HLW (including waste originating from processing of used fuel from Ukrainian NPPs at foreign enterprises and then returned to the Ukraine) is subject to long term storage and/or disposal in deep geological formations. A decision on management of used fuel after its interim storage will be made after approval of the used fuel management strategy. Used fuel that is not subject to processing shall be stored, after appropriate cooling, in special used fuel storage facilities equipped with multi-barrier confinement and protection system and with equipment for retrieval of the fuel from such a storage facility [28].

The deep geological repository concept for radioactive waste disposal being evaluated or adopted internationally and the potential for this concept to be implemented successfully is reflected in the laws passed by Ukraine that address this issue. The concept of geological disposal in Ukraine is therefore similar to that considered in other countries, being based on a system of multiple barriers consisting of the geological environment (geosphere) and the EBS. The conceptual layout of a DGR in Ukraine is presented in Fig. 2.10. The clay barriers component of the EBS is expected to provide a mechanically stable environment for the canisters by absorbing stresses and deformations, sealing gaps in the adjacent rock and retarding groundwater inflow to the canister surface. The clay is also expected to slow the migration of radionuclides released once canister failure and dissolution of the used fuel has occurred [29].

A process of regional investigations involving evaluation of the territory of Ukraine and the selection of geological regions and formations potentially suitable for radioactive waste isolation has been completed. Two zones within the geological region known as the Ukrainian Shield have been identified as being potentially favourable sites for a repository for radioactive waste disposal. These are the Korosten Pluton and a group of Proterozoic gabbro-anorthosite structures in the Middle Dnieper area [30].



1: Repository’s surface facilities;

2: Pit-shafts (hoist) and ventilating shaft;

3: Underground research laboratory;

4: Long lived waste from PVLROs/PZROs of the Exclusion Zone and Object Shelter;

5: Section for deposition of the long-lived non-heat-generating radioactive waste;

6: Section for deposition of HLW;

6.1: Service/decontamination equipment.

6.2: Area for temporary storage of SF;

6.3: Vitrified waste and SF RBMK

FIG. 2.10. Generic concept for a deep geological repository in Ukraine [29].

As the deep geological concept being considered by Ukraine includes the use to swelling clay as part of the EBS, a need has developed to identify potential domestic sources of material for use in the repository. A number of deposits of swelling clay are known to exist within Ukraine. However they are as yet incompletely characterized and need to be screened and evaluated in order to determine their potential suitability as components of the EBS. One of these materials has been selected for use in the screening studies described later in this document.

2.1.11. Summary of approaches to HLW management

The HLW management concepts and the status of the programmes in the 10 nations initially involved in this IAEA sponsored Coordinated Research Project (CRP) on swelling clays (Canada, China, Czech Republic, India, Japan, Republic of Korea, Russian Federation, South Africa, Sweden and Ukraine) have been reviewed. Each of the national programmes represented in this project has unique features and is at different stages of programme development, ranging from initial concept evaluation and development through to detail siting. Despite these differences, each of these programmes is considering the use of swelling (bentonite) clay materials as part of the EBS intended to achieve long term isolation of the radioactive materials placed in the repository. As the repository concepts considered by the participants in this CRP will require considerable volumes of swelling clay, it is important that the key features required of swelling clay be identified and means of screening potential sources of bentonite clay be established. A number of potential swelling clay materials are identified and their properties evaluated in this report.

Beyond the programmes outlined in this report are other national programmes that have or are considering use of swelling clays as part of the EBS. As these programmes were not represented in this CRP they are not discussed in detail but some of the more advanced concepts/programmes are as follows:

- **Finland**: Selection of a candidate site has been made (Olkiluoto) and site evaluation is currently ongoing. The concept adopted by Finland is much like that of Sweden and is outlined in the Posiva Environmental Impact Assessment Report [31].
- **Switzerland:** Concept assessment and evaluation is ongoing with active underground research laboratories in both granitic rock and sedimentary rock.
- **Belgium:** An active programme evaluating the use of a sedimentary geological formation is ongoing at the Mol Underground Research Laboratory.
- **France**: Evaluation of both crystalline rock (granite) and sedimentary geological environments are ongoing. An active programme associated with concept development and EBS evaluation is underway in France. An underground research facility is under construction in sedimentary rock at the Bure site in France. Work on crystalline rock concept development is being done at URLs located in Sweden, Switzerland and Canada,

2.2. Preparation of smectite rich materials for use in sealing

The generic concepts for high level nuclear waste disposal described in Section 2.1 all include the use of a buffer (defined as a dense, clay based material used immediately adjacent to the canisters) and a Backfill (generally a clay and/or clay-aggregate mixtures used to fill the other portions of the repository excavations (e.g. rooms, tunnels, shafts)) as part of the EBS.

As high density is generally considered to be a positive feature, in both buffer and backfill materials there is a need to develop practical and reliable means of compacting these materials. In most cases the buffer component is expected to be pre-compacted and then installed around the canister. Backfill materials may be pre-compacted or else compacted in place, depending on a variety of factors (e.g. density requirements, emplacement geometry, geological conditions).

2.2.1. Buffer

Considerable work has been done to develop and assess means of generating high-density smectite-rich materials (either clay-only or sand-clay mixtures) for use as buffer. These pre-compacted masses are usually prepared by pouring clay powder, granulates or clay-aggregate materials at predetermined water content into a rigid-walled form and compacting it uniaxially or isostatically under high pressure (typically 50 to 150 MPa), to form highly compacted blocks. Where clay-only materials are used to produce the buffer, the dry density achieved is typically in the order of 1800 to 2000 kg/m^3. Sand-clay mixtures will have densities that are dependent on the ratio of clay and sand as well as the compacting load applied.

Figure 2.11 shows a full-sized buffer block prepared by uniaxial compaction of commercial bentonite powder (prepared using Wyoming bentonite (MX-80) powder under 100 MPa uniaxial pressure). This block is being placed into deposition holes according to the concept shown on the right side of this figure [32]. Smaller blocks composed of clay only or sand-clay mixtures have also been used in large-scale “mock-up” experiments in Sweden, Japan, the Czech Republic, Canada and Belgium.

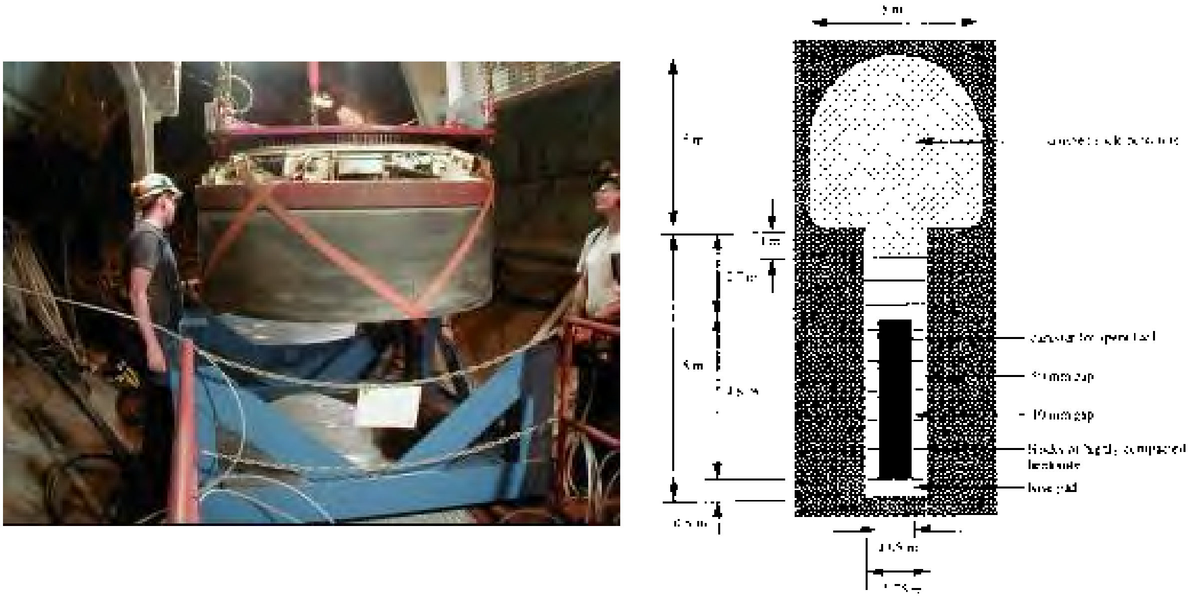


FIG. 2.11. Large pre-compacted clay block [32].

2.2.2. Backfill

Backfills are often designed using natural clays with lower smectite content than buffers or mixtures of smectite-rich clay with non-swelling and/or non-clay ballast for use as fillings of drifts, tunnels, and shafts where it is not necessary to reach the same very low conductivity and high expandability required of buffer. Backfill can be placed in a loose form and then compacted directly into place, or can be produced as compacted blocks that can be placed in stacks with a minimum of space between them. These block materials can be produced in much the same manner as buffer and so are not be treated separately in this document.

2.3. Technologies proposed for use in buffer and backfill placement

All of the programmes described in Section 2.1 use sealing materials in the excavation volumes not occupied by the waste packages. The installation of these materials is not necessarily a straight forward process. The technologies required for placing swelling clay materials will depend on the emplacement geometry selected, the emplacement concept selected and the nature of the waste packages themselves. Common to all generic emplacement concepts outlined in Section 2.1 is the need to emplace canisters holding highly radioactive materials. The radiological field present on the surface of the canisters to be emplaced will be determined by the type of radioactive materials in the canisters (UF, reprocessing wastes, etc.), the canister itself (e.g. metal used, thickness of walls, details of internal components), and the time between removal from the reactor (or reprocessing) and actual emplacement. As a result of these uncertainties in canister conditions it will be necessary to develop technologies for their emplacement that are unique to each national situation.

Despite the limitations imposed by the nature of the waste canisters and waste forms considered by each national programme, there are certain aspects of emplacement of the sealing materials that are common to most generic concepts. In each of the concepts outlined above there is a clay based buffer component of the EBS that is immediately adjacent to the waste canister. Its composition may vary depending on the details of the EBS approach selected but it is typically a dense, high bentonite content material that has been pre-manufactured (it is often referred to as buffer). This pre-manufactured buffer material may be installed in several manners, including:

- Emplacement of full-sized segments in the boreholes prior to canister installation (Fig. 2.11) [25];
- Emplacement in tunnels prior to in-room canister emplacement (Fig. 2.12); or
- Installation as part of a pre-assembled canister-clay “super container (SC)” as proposed for the KBS-3 and 3H concepts (Figs 2.13 and 2.14).

The buffer may under some circumstances also be compacted directly into the borehole depending on the composition chosen and the properties required of the buffer [34]. Compaction directly into a borehole may be done by leaving a centrally located volume for canister installation or may involve coring out of the material occupying the volume needed for canister installation [2, 12].

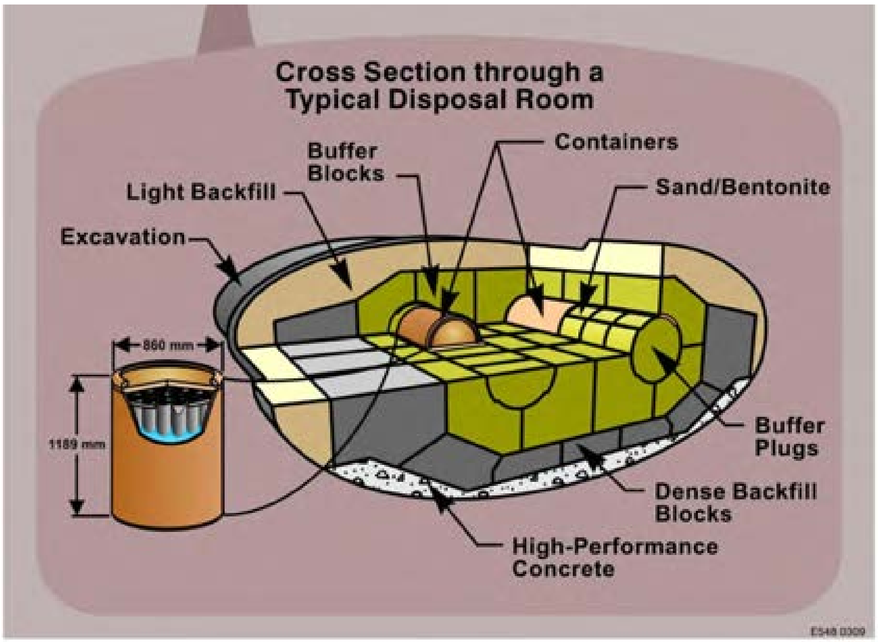


FIG. 2.12. Example of use of pre-compacted clay based materials [33].

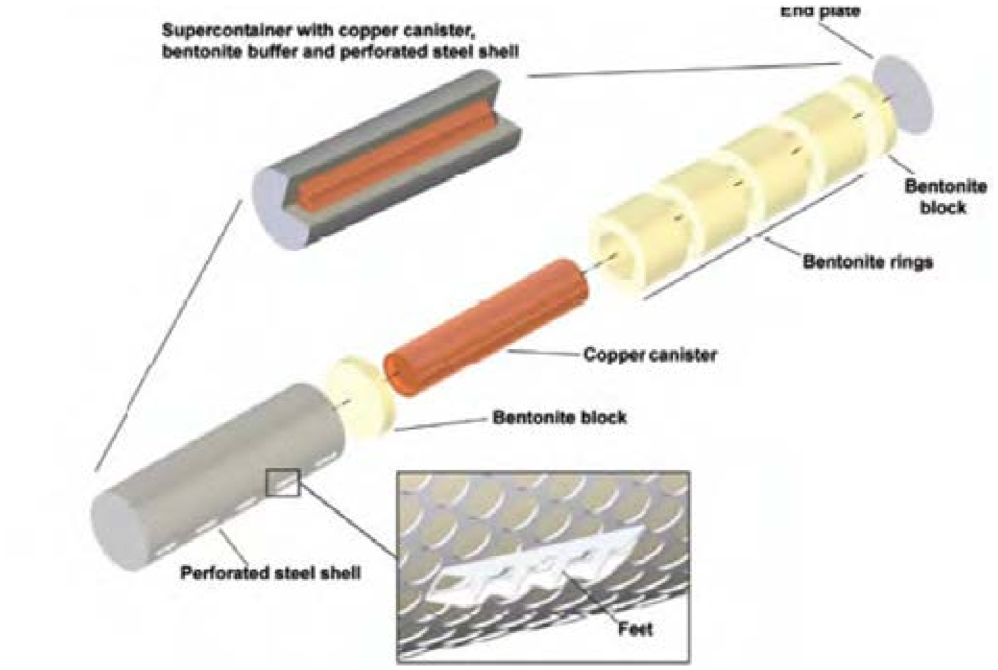


FIG. 2.13. KBS-3H super-container concept [27].

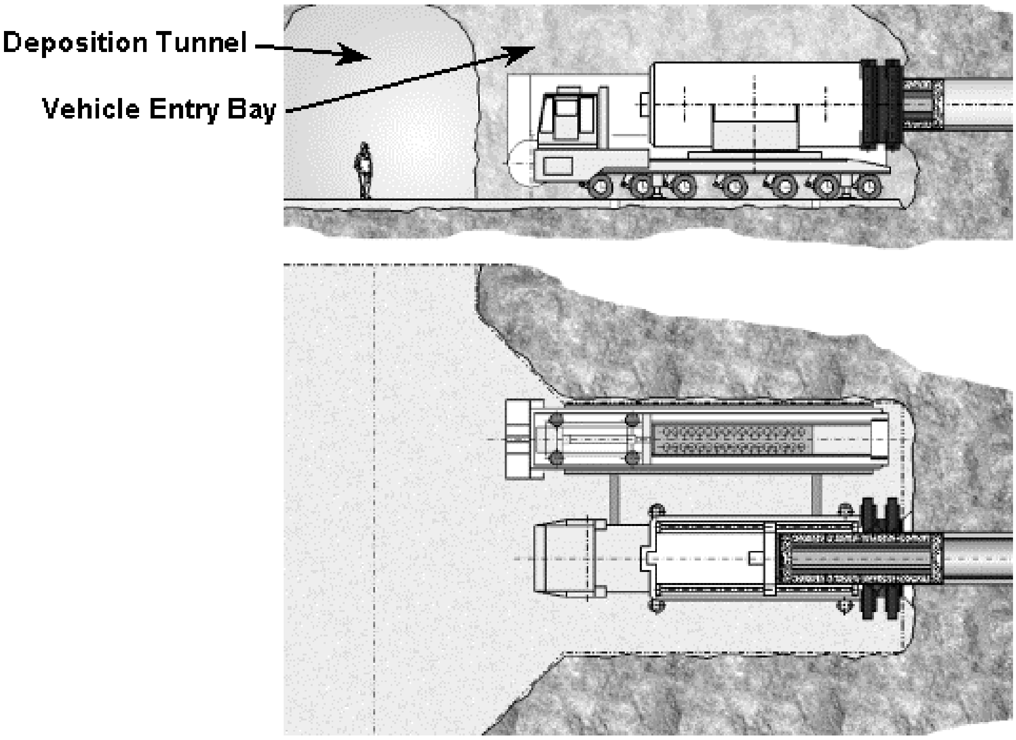


FIG. 2.14. Horizontal installation of a used fuel super container [26].

As described in Sections 2.1 and 2.2, beyond the buffer there are other clay based components of the EBS. These may include densely compacted materials with high bentonite content and then further components of backfill as is the case for the concept outlined in Ref. [35] or may include densely compacted backfill materials only. Concepts of how such materials might be installed have been presented by a number of researchers and designers [33, 36–38]. Examples of how installation of backfill might occur are provided in Figs 2.15 to 2.18. Beyond the conventional emplacement concepts and geometries an alternative emplacement concept that avoids the use of much of the handling equipment proposed for the in-room geometry has been outlined by work done in Switzerland [39, 40]. This concept, presented in Fig. 2.18 involves the placement of a large volume of highly compacted bentonite pellets (backfill) to fill the remaining tunnel volume and Fig. 2.19 shows a trial installation.

Installation of backfill-type components has been undertaken in several underground laboratories as part of technology development and demonstration activities. At the Canadian URL both pneumatic and dynamic emplacement technologies have been attempted as part of various EBS studies [42–44]. At the Äspö facility SKB has also examined the installation of backfill materials in full-scale tunnels using dynamic in situ compaction as part of their tunnel plugging experiments [44, 45]. Each of these technological approaches show promise for repository application but have also identified limitations to their use in terms of the achievable installed density, the geometry in which it can be effectively installed and the installation rate.

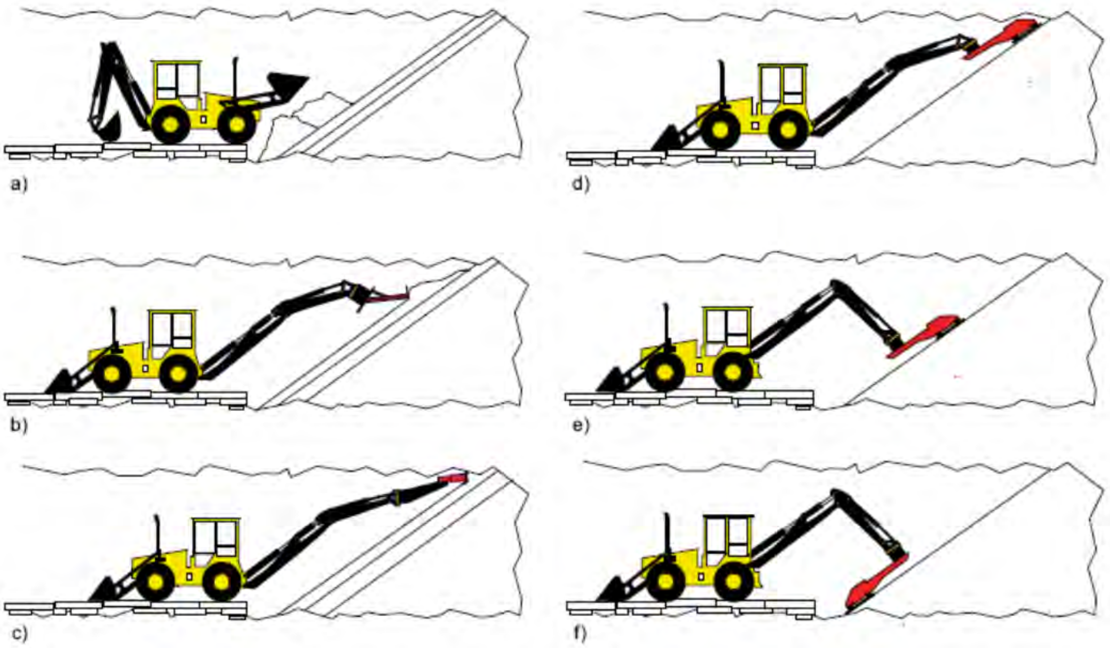


FIG. 2.15. Process for installing tunnel backfill using in situ compaction [36].

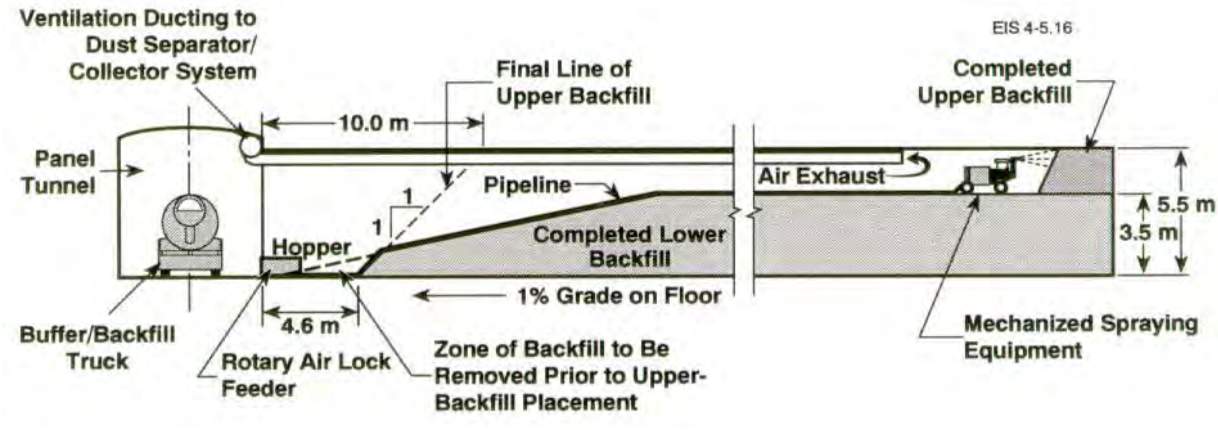


FIG. 2.16. Light (upper) backfill placement [37].

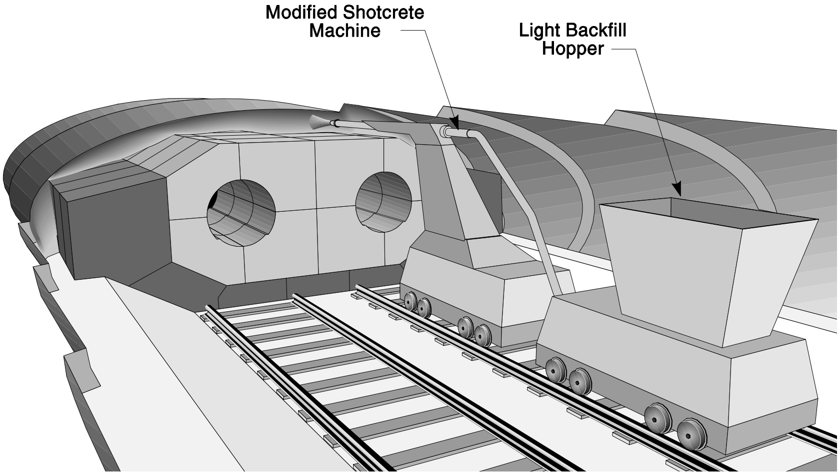
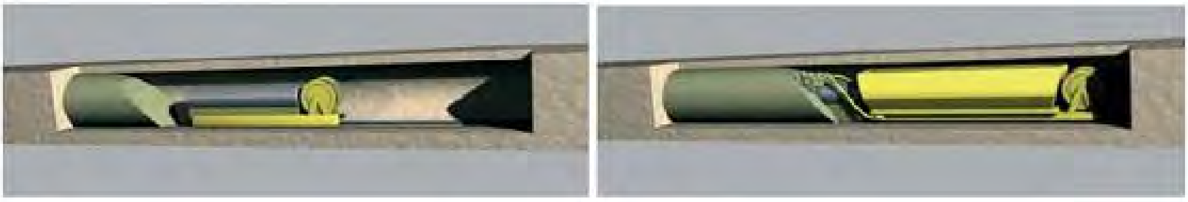


FIG. 2.17. Concept for light (upper) backfill placement (in-room emplacement method) [37].



(a) Emplacement of canister and base blocks (b) Installation of bentonite pellets

FIG. 2.18. NAGRA in-room emplacement concept: (filling most of tunnel volume with bentonite pellets) [39].

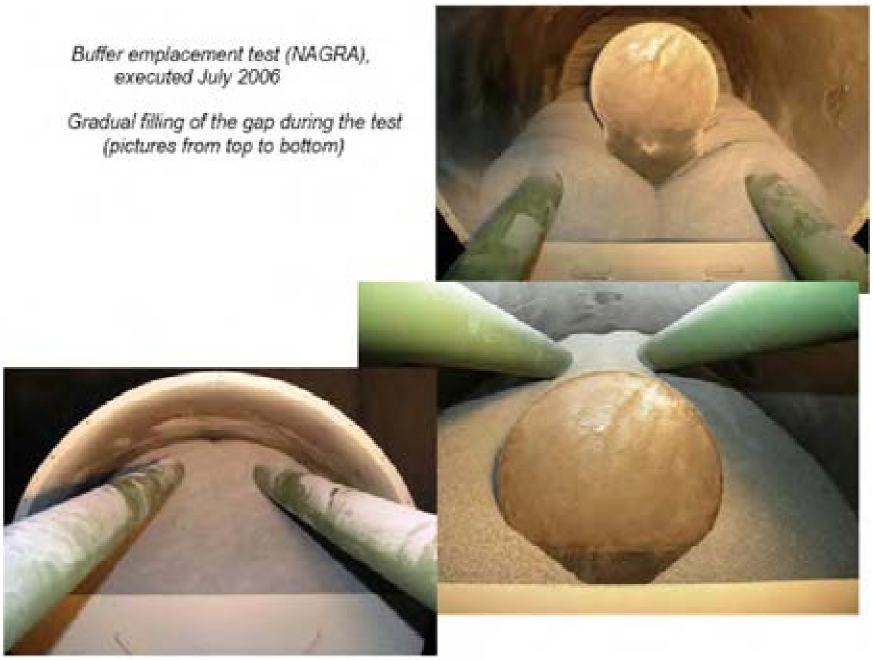


FIG. 2.19. EB experiment examining the NAGRA in-room concept [41].

2.4. Technological issues associated with the installation of swelling clays

There are a number of technological issues related to the installation and use of swelling clays in a repository. Section 2.2 provided descriptions of some of the emplacement geometries and technologies being considered for use in installing swelling clays in a repository. They are largely based on conceptual designs with only limited demonstration of their viability in actual repository conditions having been done to date.

Assuming that the clay based materials selected for use as buffer and backfills are adequate for the purpose of engineered barriers in a repository environment, these swelling clay materials need to be both practical to place or manufacture and durable enough to survive transportation to their emplacement location.

The production of full-scale pre-compacted masses of buffer, their storage, handling and durability in a repository environment are not trivial issues. These very dense and large segments of buffer may tend to become brittle or subject to damage induced by transportation following their construction. The generation of such large segments of highly compacted clay also requires use of very high pressure compression equipment, the operation and maintenance of which may also be a technological challenge. Work on this issue has been conducted in Sweden [46, 47] and in Japan [48].

As is the case with the buffer, the manufacturing of dense backfill materials proposed for placement, as large pre-compacted masses also requires considerable technological development. The manufacture and perhaps more importantly, their transportation to and within a repository, together with their accurate placement requires a robust handling technology. This is a topic of ongoing study by SKB and Posiva as part of their repository design processes [49].

Beyond the technological issues and challenges associated with the use of clay based sealing materials that are being addressed by the various national programmes described in this document, there are also a range of other issues that are undergoing active international investigation. Amongst them are those associated with the identification of the long term viability of using swelling clays as part of an EBS and what environmental aspects could potentially affect their long term performance. A number of these topics are discussed in subsequent sections of this document. In order to develop a fuller understanding of the likely long term stability and performance of the swelling clay materials proposed for use in a geological repository, the properties of these materials have been undergoing extensive evaluation by a number of laboratories and in a range of underground research laboratories. Section 3 outlines some of the key materials properties that are looked for in a candidate buffer material and how certain environmental and other factors may influence the effectiveness of these materials in isolating radioactive materials.

3. SWELLING CLAYS: THEIR DEFINITION AND PROPERTIES KEY TO SCREENING

As described and discussed in Section 2 of this document, swelling clays are being considered as a primary component of several engineered barriers for use in isolating HLW in a geological repository. This chapter provides a guideline for use in determining the potential suitability of swelling clays from previously uncharacterized deposits as candidates for use as a component in the engineered barrier system of a repository. Such a general guideline for material characterization will allow preliminary screening of potential candidate clays and although detailed assessment will ultimately be required, a long and expensive material qualification process may be avoided.

3.1. Clays

3.1.1. Definition of clay minerals

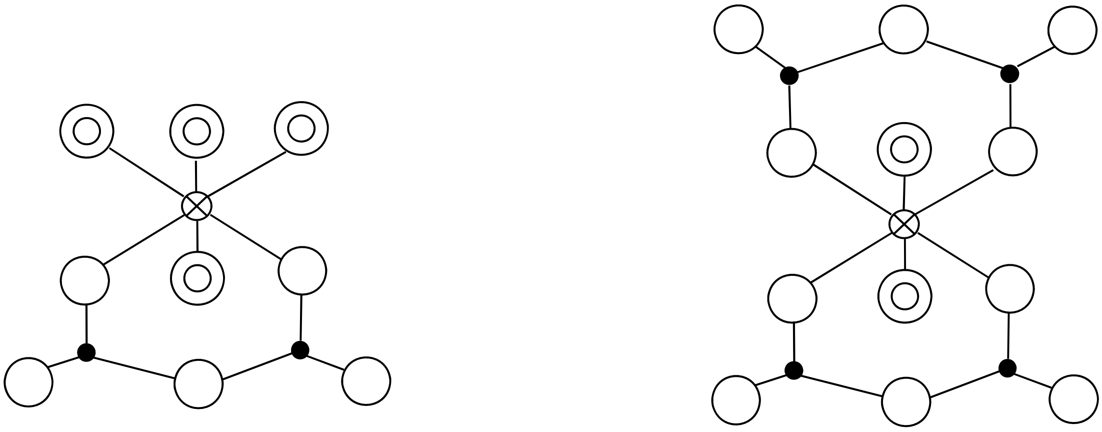
Clay is a term that is commonly used to describe either the size of the individual particles present in a deposit (clay size fraction) or else specific minerals (clay minerals). Because clay minerals tend to be very small in size (<0.002 mm in dimension) particles that fall into this size range are defined as clay-sized but are not necessarily clay minerals. Understanding of this distinction is important in the assessing of clay materials because two samples of clay having identical particle size can have very different behaviours based on the clay minerals present.

Clay minerals are formed in a variety of environments and take on an almost infinite variety of mineral structures as a result of natural variations in the availability of their constituent atoms. Clay minerals do however break down into a somewhat more limited number of mineral families that share similar microstructures. Depending on the structure developed in the course of mineral formation, different clay minerals will exhibit very different physical, chemical and mechanical behaviours. For example in the presence of free water montmorillonite clay minerals will swell to many times their initial volume as a result of taking water into the layers between the individual unit layers of clay, while kaolins will remain essentially unchanged. As a result of these substantial differences in the behaviour of the various clay mineral types determining the types and proportions of clay minerals in a particular clay deposit is one of the first and most important of the steps necessary in screening a source material.

Swelling Clays are a widely distributed type of geological material usually containing more than 50% of mineral particles less than 2 microns in size. Their main mineral components are dispersed layer silicates such as illite, kaolinite, montmorillonite and other clay minerals. The well known and unique properties observed in clays such as plasticity, swelling, cation exchange and high absorption capacity are caused by dispersed layer silicates. Fine grained particles of quartz, mica, carbonates, iron oxides, sulphides and organic material also occur in clays. The unique crystal structures of clay minerals are discussed in detail in Ref. [50] and so are not dealt with in detail in this document.

Most clay minerals are characterized by their platey-layered structure, which are similar to those observed in mica [32, 50, 51]. These minerals are composed of well-defined parallel layers of atoms containing silicon, aluminium, oxygen and hydroxyl groups with or without magnesium, iron and other minor elements. Within the layers the atoms are arranged in parallel sheets of a highly symmetrical, generally hexagonal, manner. The atoms are firmly bonded together to form relatively simple and stable structures. The forces holding the layers together are generally weaker and it is the manner in which the layers are arranged and bonded that leads to a large variety of crystal structures.

In the same manner as the various layers, the atoms within each of the layers are arranged in closely coordinated groups. Tetrahedral groups of oxygen atoms surrounding silicon, aluminium are linked together in sheets of hexagonal symmetry and likewise octahedra of oxygen and hydroxyl group surrounding aluminium, magnesium, and iron. The dimensions of these planar networks are very similar and consequently composite tetrahedral-octahedral-layers are readily formed. These layers form units, may be two- or three-layered (Fig. 3.1) [50].



⊕ : Hydroxyl; ●: Aluminium; ○ : Silicon; ◎ : Oxygen

(a) Two-layer unit cell (b) Three-layer unit cell

FIG. 3.1. Diagrammatic representation of two types of clay unit cell.

Two-layered sheets are formed by tetrahedral and octahedral layers in pairs. The part of free electrical charge associated with the tetrahedral layer may be neutralized by hydrogen with substitution of oxygen in the hydroxyl group. Three-layered sheets composing an individual layer of the mineral are formed by two tetrahedral with one octahedral layer located between them. These layers are bonded through the common oxygen ions.

Ions forming the octahedral layer may occupy a varying number of sites depending on ion charge: all sites in the cases of magnesium and iron (2+) or only two thirds of the ions sites for aluminium or iron (3+). Mineral groups with these different octahedral layer occupations are called tri-octahedral and di-octahedral based on the particular case.

The structural layers may or may not be electrically neutral, depending on isomorphous replacements of atoms within the layer (or the absence of isomorphism). The substitution of aluminium for silicon in the tetrahedral sheet as well as the substitution of magnesium, iron and aluminium for each other in the octahedral sheet is very important to distinguish main mineral groups of layer silicates (clay minerals, micas, etc.). For example, replacement of silicon by aluminium in muscovite produces an electrically negative layer that is counterbalanced by positive potassium ions that occupy positions between layers and thus forming stronger bounds:

$$Si^{4+} \leftrightarrow Al^{3+} + K^{+}$$

One group of minerals, kaolin, is composed of neutral or largely neutral layers. Their structural formula is: $Al_2Si_2O_5(OH)_4$. Isomorphous replacement and the presence of external cations needed to give an electrically neutral structure tends to very limited. As a result this mineral tends to be very stable, durable and relatively non-reactive.

The montmorillonite (smectite) group of minerals has extensive isomorphous replacement in both the tetrahedral and the octahedral layers, and the external ions required for electrical neutrality play an important role. Integrated chemical formula can be presented as: $(Na,Ca,Mg,H_2O)(Al,Fe,Mg)_2(Si,Al)_4O_{10}(OH)_2$. Owing to the weak binding forces between the tri-octahedral unit layers of this type of mineral, water and other molecules readily penetrate, giving rise to characteristic swelling phenomena. Cations, such as H^+, Na^+, Mg^{2+}, Ca^{2+}, etc. lie between the silicate layers. They are readily exchangeable and form Na-montmorillonite, Ca-montmorillonite, etc., which are identified based on their dominant exchangeable cation.

All varieties of clay and mica minerals are generated due to type of batch (two- or three-layered), type of cation (two- or three ion charge – therefore the type of cation sites occupation: all sites, three octahedral or two third sites, dioctahedral) and at last the type of present inter-batch layer (the lack of it, alkali ion, water molecules or brucite layer). Mixed-layered silicates can be formed by sequence of structure layers of most of above mentioned main groups. Another constitutive feature of layer silicates which leads to the large number of varieties occurring in nature is the possibility of rotating and shifting of structural layers and units, generating polytypes. Some of polytypes can sufficiently differ in morphology of crystallites forming the clay rock itself that different physical and mechanical properties may exist.

It should be pointed out that interchanging of layers occurred along one structural axis: c and is evidently fixed in interlayer distance. The value of this distance is becoming the diagnostic parameter for all the groups of layer silicates. This diagnostic value and main groups of layer silicates may be characterized by the diagrammatic representation in Fig. 3.2.

The layer silicates the most frequently occurred in clays are: kaolinite, montmorillonite, hydromicas (illite) and mixed layered silicates (illite-montmorillonite, illite-smectite, etc.). Their position in layer silicates classification is listed in Table 3.1 and is marked by bolded text.

Each mineral group has its diagnostic value of interlayer spacing – d (001), Å and this is a key feature for use in assessing the composition of natural clays.

3.1.2. Methods of clay mineral identification

At present, means of identifying the various basic clay mineral groups are quite well defined. Beyond basic identification, further identification of structural details of mineral species is useful in characterizing a source material. Therefore, methods of identification typically incorporate two stages: firstly, mineral group and then mineral variety (chemical composition and structural features) identification.

The treatment of the sample under investigations includes fine clay fraction separation to eliminate carbonates, quartz. The main methods of analyses are: X ray powder diffraction XRD, scanning-electron and transmission-electron microscopy (SEM and TEM) and thermal analysis (DTGS). Structural specification of swelling layer silicates can be revealed by saturation of the sample with organic molecules (glycerol, etc.) as well as by thermal dehydration.

Kaolinite group – chemical formula: $Al_2Si_2O_5(OH)_4$

Polytypes: kaolinite, dickite, nacrite, halloysite (incorporate water molecules in interlayer spaces). The reference x ray diffraction pattern for kaolinite is presented in ASTM PDF card #14-164[1] and can be accessed from that source. The diagnostic values for the first, and strongest diffraction angles d (001) and (002) are: 7.17 Å and 3.58 Å respectively.

Montmorillonite (smectite) group – chemical formula: $M_{0.33}Al_2(Si, Al)_4O_{10}(OH)_2 \cdot n\ H_2O$

The Montmorillonite group includes a number of mineralogically distinct sub-types which have slightly different chemical composition and structure but all of which are able to expand considerably in the presence of free water. It should be noted that the term bentonite is often used interchangeably with montmorillonite or smectite. While also a mineral subspecies of the montmorillonite-group, the name bentonite is more commonly used to identify commercial products that consist of crushed, montmorillonite-rich shales that are mined from a variety of sources around the world.

The main mineral varieties are:

– Montmorillonite:	$(M_{0.33} \cdot nH_2O)(Al_{1.67}Mg_{0.33})\ Si_4O_{10}(OH)_2$
– Beidellite:	$(M_{0.33} \cdot nH_2O)Al_2(Si_{3.67}, Al_{0.33})\ O_{10}(OH)_2$
– Nontronite:	$(M_{0.33} \cdot nH_2O)Fe_2^{3+}(Si_{3.67}, Al_{0.33})\ O_{10}(OH)_2$

"M" is a mono-valent cation or molecule: Li^+, Na^+, K^+, NH_4^+ or in some cases: $[Al(OH)_2]^+$ or organic molecules RNH_3^+. Also, the exchangeable ions can be substituted for 0.5 divalent cations: Ca^{2+}, Mg^{2+} etc. Mineral varieties often show low crystallinity due to superposition of sheets in completely random fashion, giving rise to "turbo-stratic" structure. The diffraction patterns generated by these materials reveal a number of broad peaks. The type of isomorphous cations and quantity of interlayer water molecules are connected with the interlayer distance, thus giving diagnostic values for the first diffraction angle d (001) in a range of 12 to 15 Å. Diffraction patterns of Montmorillonite are presented in ASTM PDF card #29-1498 and #13-135.

The characteristic swelling behaviour of Montmorillonite is widely used for its identification. In order to specifically identify the mineral type organic molecules of known dimension are used to saturate the interlayers (e.g. glycerin or ethylene glycol) and the resultant changes in the x ray diffraction angles are measured. The diagnostic values for d (001) are for: air-dried mineral 12-15 Å, saturated mineral 17.6 Å, then dried at 600ºC for two hours to eliminate all the inter-layer water will result in a mineral having a d (001) of 9.6-10 Å.

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[1] Here and in other parts of this section references have been made to host institute PDF card # for quality assurance purposes.

TABLE 3.1. CLASSIFICATION OF LAYER SILICATES

Batch type	Interlayer spacing occupation	Octahedral layer occupation			
		Di-octahedral	d (001), Å	Tri-octahedral	d (001), Å
Two-layer (1:1)	-	Kaolinite group $Al_2Si_2O_5(OH)_4$	7	Serpentine group $Mg_3Si_2O_5(OH)_4$	7
	Water molecules	Halloysite group $Al_2Si_2O_5(OH)_4$	10		
Three-layer (2:1)	Water molecules, M (Na, Ca, Mg)	Montmorillonite (smectite) group $M_{0.33}Al_2(Si, Al)_4O_{10}(OH)_2 \cdot nH_2O$	10 or higher	Saponite group $M_{0.33} Mg_3Si_4O_{10}(OH)_2 \cdot nH_2O$	10 or higher
	Water molecules, M (Na, Ca, Mg)			$M_{0.33-0.5} Mg_3(Si, Al)_4O_{10}(OH)_2 \cdot 4\text{-}6\ H_2O$	
	K, Na	Muscovite group $Kal_2(AlSi_3)O_{10}(OH)_2$	10	Flogopite group $KMg_3(AlSi_3)O_{10}(OH)_2$	10
	Water molecules, M (Na, K)	Al-Hydromicas group Illite $K(Al, Fe, Mg) (Si, Al)_4O_{10}(OH)_2$ Mg-Fe-Hydromicas group Glauconite $K(Fe, Al, Mg) (Si, Al)_4 O_{10}(OH)_2$	10	Mg-Fe-Hydromicas group Hydroflogopite $KMg_3(AlSi_3)O_{10}(OH)_2$	10
				Chlorite group	14
Mixed layer alumo-silicates	Same types of batches	Illite-Montmorillonite		Chlorite –Vermiculite-Montmorillonite	
	Different types of batches	Kaolinite -Montmorillonite			

Illite group – chemical formula: $K(Al, Fe, Mg)(Si, Al)_4O_{10}(OH)_2$

This variety of hydrous mica incorporates small quantity of water molecules in comparison with its primary mineral equivalent muscovite, and its diffraction pattern is almost the same as of muscovite.

The diagnostic diffraction pattern of illite is presented in ASTM PDF card #43-685 and its first and most readily identifiable d (001) value is at 10 Å.

Glauconite group – chemical formula: $K(Fe, Al, Mg)(Si, Al)_4O_{10}(OH)_2$

This hydrous mica is an iron-containing variety of illite. The reference diffraction pattern of Glauconite is presented in ASTM PDF card #9-439. The key diagnostic value d (001) for this mineral is found at 10 Å.

Mixed layer alumino-silicates

Two general types are defined among mixed layer alumino-silicates. The first and most widely found type of mixed-layer mineral is formed by exchanging of interlayer cations and water in the sequence of alumino-silicate layers. Minerals of muscovite, illite and montmorillonite groups have the same layer composition (Fig. 3.2) and differ in their interlayer filling. One variety of mixed layer silicate formed by combinations of the unit layers of these minerals is called illite-montmorillonite. Another possible combination of this type of mixed layer system is the presence of chlorite, vermiculite and montmorillonite unit layers. The second major type of mixed layer system involves exchanging of various alumino-silicate layers within the mineral. This type of exchange involves two-component mixed-layered mineral, two-layer and three-layer thick sections are exchanged in the structure of kaolinite-montmorillonite.

Identification of the specific type of mixed-layered silicates present requires saturation of the clay sample with organic molecules in order to determine their “real” interlayer distance(s). The x ray diffraction data for various layer superpositions are usually calculated according to the theoretical structure. Data available in PDF files from ASTM should be used as a guide in the identification process of mixed layer systems (e.g. diffraction pattern of illite-montmorillonite is located in ASTM PDF card #35-652 and chlorite-vermiculite-montmorillonite in ASTM PDF card #39-381). The diffraction pattern of kaolinite-montmorillonite is presented in ASTM PDF card #29-1490.

Two examples of typical untreated powder XRD analysis of a natural clay sample (two from the western region of India and one from Saskatchewan Canada) are shown in Fig. 3.3. These traces illustrate both the variety of minerals that can be present in a natural material as well as the general similarity in the composition of these two materials. This figure also shows that both of these materials have high content of montmorillonite that would initially qualify it as passing the initial screening for swelling clays. Further testing would however be necessary to establish the chemical, physical and mechanical suitability for use as a candidate material for use in a repository application.

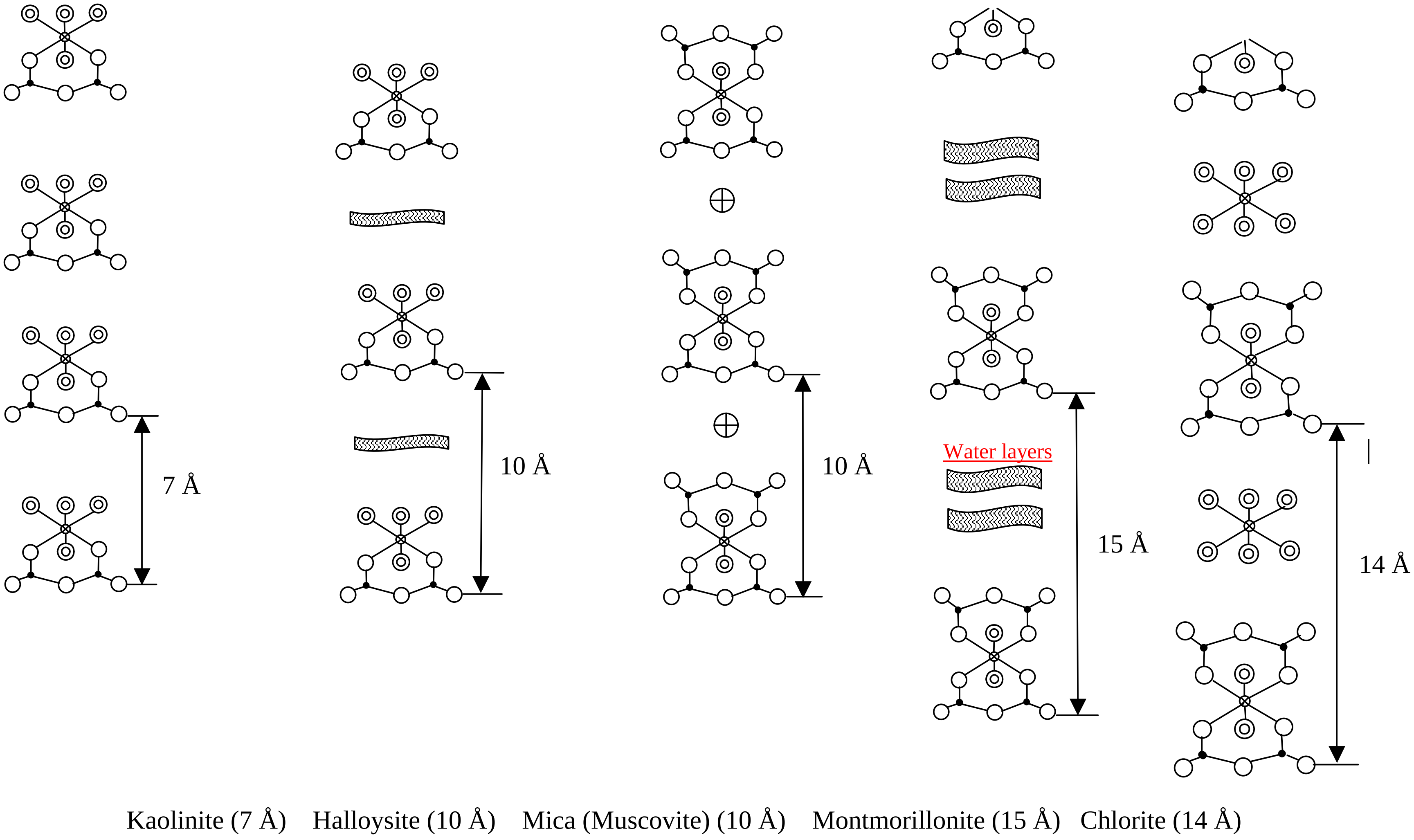

FIG. 3.2. Diagrammatic representation of the succession of layers in a number of layer lattices silicates. (The approximate basal spacings are given in Å (10^{-10} m) [50]).

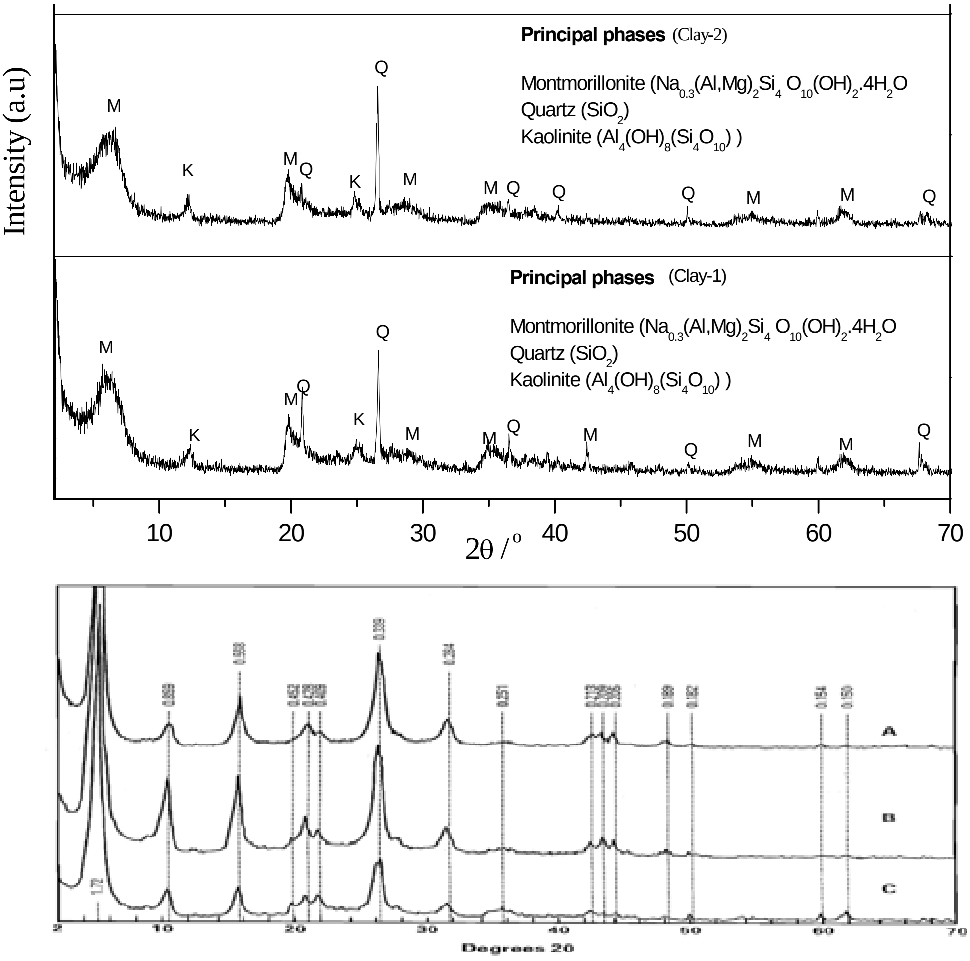

FIG. 3.3. XRD patterns of three swelling clays. (Upper traces: Clays 1 and 2 from western region of India; lower trace is for three samples of Saskatchewan bentonite (all from the same deposit). Identification of reflections of Montmorillonite (M) – PDF #29-1498; Quartz (Q) – PDF # 88-2302; Kaolinite (K) – PDF #78-2110) are provided.)

3.2. Preliminary screening

3.2.1. Guideline for screening

The process of screening potential materials for use in a repository is not one that will typically pre-exist in any nation that has not been involved in this activity previously. Most of the component tests of the screening process will be known to various specialized organizations in most countries but may not be recognized by the organizations responsible for dealing with waste management issues. Examples of the technical and scientific specialties that must be brought to bear on the issue include: civil and geotechnical engineering, geology, geochemistry, mineralogy and chemistry. Using information compiled from a variety of sources for well-characterized reference materials (such as Bentonite MX-80 and Saskatchewan bentonite [52-54]), the main characteristics that must be present in raw material to be considered for the potential material are broken down into two categories. The first category are those characteristics that can be used as screening criterion and the second are properties that must be determined based on more complex materials testing.

Screening Criterion:

- The presence of a considerable portion of swelling material (preferably a Na-montmorillonite dominated material);
- High cation exchange capacity;
- High swelling capacity (volumetric and swelling pressure);
- Low permeability when compacted to density to be used in repository;
- Preferably having accessory minerals that are poor in potassium and iron.

Detailed Material Assessment:

- Suitable thermal conductivity;
- Potential to adsorb radionuclides;
- Ability to retard canister corrosion;
- Reasonable stability under the chemical and thermal conditions expected.

To be economically viable, the clay deposit and the raw material from it can be judged according to the following criteria:

- The exploitable amount of material from a particular clay deposit should be at least twice the required mass for the buffer and backfill required for the waste repository;
- The mineral composition of a potential clay source should not vary by more than 10%.

3.2.2. Generic screening tests

Once a deposit of clay (or clay shale) has been identified as containing swelling clay of potentially suitable character, the following combination of screening tests should be performed:

- Mineralogical composition:
    - Clay contents by x ray diffraction;
    - Clay content by DTGS technique.
- Basic physical characteristics;
- Swelling properties;
- Coefficient of permeability.

The above listed screening tests are the minimum currently perceived as being necessary to identify potentially appropriate materials and could be expanded. It is understood that the above listed tests do not qualify a material for use in a repository, but rather they help to separate clearly unsuitable materials from those that have the potential for use. More detailed characterization of materials that pass the initial screening process will be necessary before they can be considered as suitable.

3.2.2.1. General mineralogical and chemical composition

Swelling materials (clays) are found in many places around the world. The dominant mineral components are dispersed layer silicates, such as montmorillonite, illite, kaolinite, and other minerals such as fine-grained quartz, mica, carbonates, iron oxides, sulphides, and organic materials. Their structure was described in detail in Section 3.1 and since each mineral group has its diagnostic value of interlayer spacing it is relatively straight-forward to determine what minerals are present using X ray diffraction technology.

It is also possible to determine the mineralogical composition of a sample through chemical analysis of the major and trace elements present in the solid phase of the raw material. The experimental procedure described in the Febex project provides the method commonly used to do this analysis [55]. The major cations present in the pore fluid or in solution outside of the minerals themselves can be determined by a test called Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES). Sodium and potassium contents can also be determined by Flame Atomic Emission Spectrometry (AAS-Flame) and trace elements with ICP-MS. The presence of anions can be determined with the aid of ion chromatography. Afterwards, the total alkalinity of the samples was determined by potentiometric titration. The detailed chemical analyses of the materials examined in this CRP are presented in Section 5.4.

Other techniques such a thermal gravimetry (TG) and differential thermal analysis (DTA), as described in Ref. [56], can also be used for element and mineralogical determinations (Fig. 3.4). These tests evaluate the temperature at which water is lost from a specimen as well as the mass that is lost, thereby providing an indication of the amount of strongly-held water present (normally associated with swelling clays). Slight differences in the temperature at which water loss occurs also provide information on the presence of non-swelling or mixed layer minerals.

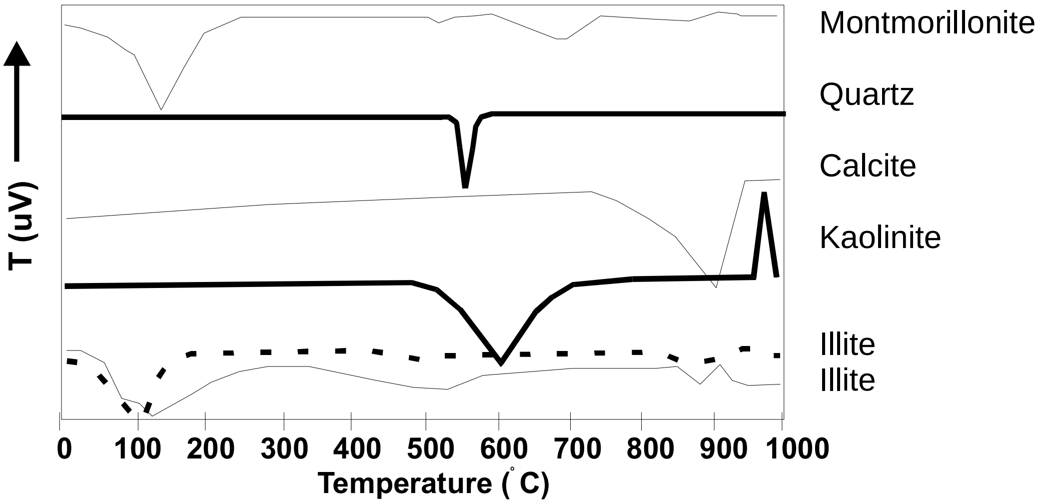

FIG. 3.4. DTGS analyses of different minerals.

Another characteristic of swelling clay is the high interlayer cation population in the clay structure that results in a high cation exchange capacity (CEC). The high number of cation exchange sites is the result of the very high surface area of the clay (up to 800 m^2/g of clay) and the high negative charge on these surfaces. Tests to determine not only the total number (cation exchangeable capacity (CEC)) but also the type of cations (exchangeable cations (EC)) associated with the clay surfaces provide valuable information on the swelling clay content of an uncharacterized material. The presence of weakly sorbed cations on the clay surfaces may allow for the exchange of these non-radioactive cations with positively charged radioactive elements or compounds, resulting in retardation of their movement through the barrier. Given the relative simplicity of EC and CEC analyses, they allow rapid screening of potential materials (a low CEC indicates a low smectite content or a low-reactivity clay, both of which are undesirable). This tool for assessment is used in Section 5 where various previously uncharacterized clay materials supplied by participants in this CRP are evaluated.

The nature of the interlayer cations affects not only the ion exchange properties of clay but also has a very substantial influence on the swelling capacity, hydraulic behaviour as well as rheological behaviour as will be discussed later in this document.

3.2.2.2. Basic physical characterization

In association with the basic mineralogical tests described above that can be run to determine the clay minerals present in an uncharacterized sample of “clay” there are three basic geotechnical tests that provide fundamental information on the nature of soil materials. These tests are known as the Atterberg (consistency) limits (liquid limit, plasticity limit, plasticity index)), the particle size distribution and specific density.

(a) Atterber (consistency) limits

The Atterberg (consistency) limits of a soil are a very simple and quick means of assessing the cohesive behaviour of a soil (hence an indication of the clay content and type). There are three components to these limits, the liquid, plastic and the difference between them (plasticity index). This test is widely used in agriculture and geotechnical engineering and the method of carrying out this test is standardized and is included in the standard engineering materials testing specifications of many nations (e.g. CSN 72 1014 (Czech Republic); ASTM 423-66 (USA)).

Liquid limit - w_L

The behaviour of cohesive soil depends on its water content. As the gravimetric water content increases, the rheological behaviour of the soil changes until it is on the verge of becoming a viscous fluid. The water content at which the soil shows some shear strength is considered to be the boundary between liquid and plastic consistency. It is called the liquid limit w_L. Depending on the liquid limit, soils can be specified as soils showing the following plasticity:

- L - low w_L below 35%;
- I - medium w_L = 35-50%;
- H - high w_L = 50-70%;
- V - very high w_L = 70-90%;
- E - extremely high w_L exceeding 90%.

Plasticity limit - w_p

After significant reduction of its gravimetric water content, the soil obtains characteristics of a brittle material, and when exposed to strain it fails. This state is called stiff consistency. The range in gravimetric water content between the uppermost plastic behaviour and stiff consistency is called the plasticity limit. The means of doing plasticity limit measurements are also standardized in the engineering (soil) materials testing specifications of many nations (e.g. CSN 72 1014 (Czech Republic); ASTM 423-66 (USA)).

Plasticity index - I_p

Plasticity is expressed by means of the plasticity index, which is the range of gravimetric water content over which soils show plastic behaviour. The plasticity index is expressed as the difference between the liquid and plasticity limit

$$I_P = w_L - w_P$$

where w_L is water content at liquid limit and w_p water content at plasticity limit.

The relation between the plasticity index and water content at the liquid limit for clay and other material (e.g. loam) can be expressed in a Casagrande plasticity diagram as indicated in Fig. 3.5. However materials dominated by minerals such as montmorillonite will have w_L and I_p values that are off this chart (beyond the upper right corner) and materials showing these values show good initial potential for suitability as candidate swelling clays.

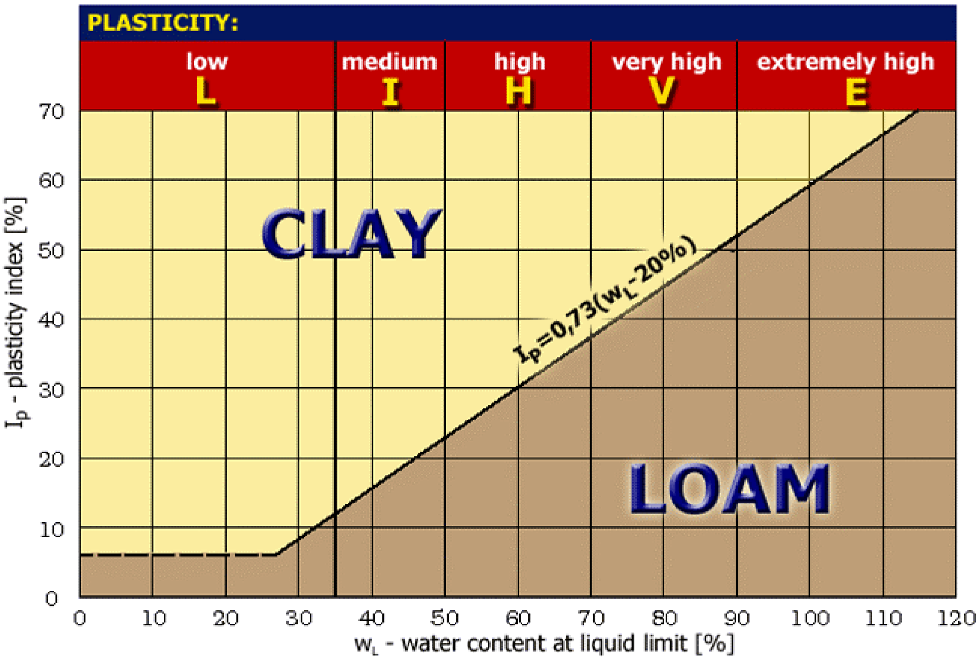

FIG. 3.5. Casagrande plasticity diagram [57].

(b) Particle size distribution (grain size analysis)

Grain size analysis is universally used in the engineering classification of soils and the information gained can be used to predict soil-water movement, although permeability tests are more generally used. The presence of very large fine contents is also a useful indicator that the sample may contain desirable clay minerals.

The particle size distribution (grain size analysis) pattern as illustrated in Fig. 3.6 can be determined in accordance with the CSN 72 1017/ASTM D422-63 soil characterization procedures and various other national and international standards.

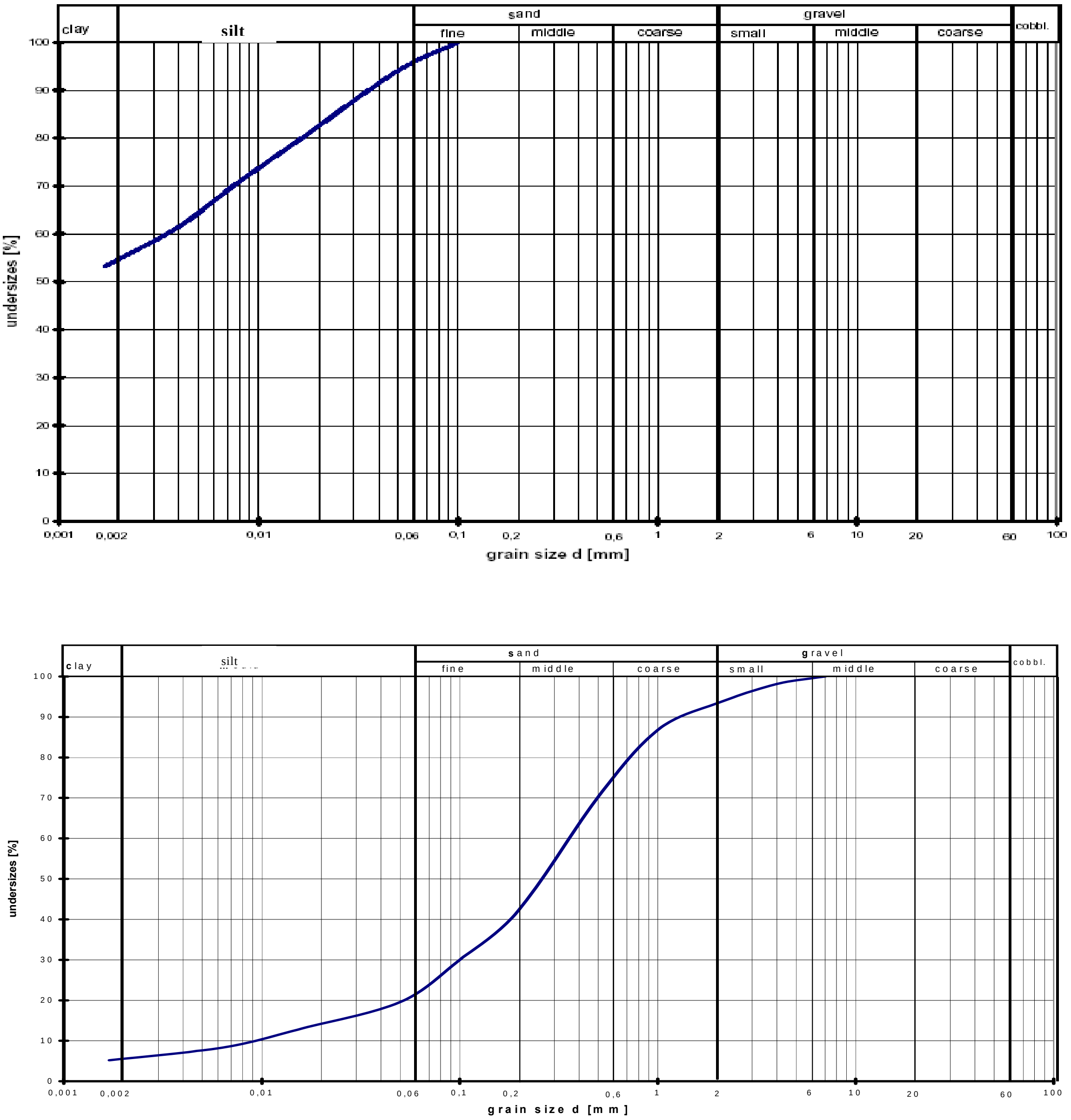

FIG. 3.6. Particle size distribution diagrams of different materials. (Upper plot is a smectite-rich clay and lower is a non-clay soil [57].)

There are two components to the determination of particle size distribution in soil: screen analysis and densimetric (or settlement) test. In screen analysis the sample is washed through a series of standardised sieves to separate out grains bigger than 0.06 mm (sand to gravel) and the proportion captured in each of the sieves are weighed and used to plot a grain size distribution for the coarse component. If there is essentially no fines content then it is unlikely that the material being tested is suitable as a candidate swelling clay since the size of montmorillonite particles are typically smaller than 0.06 mm. Density based particle size analysis is carried out on those materials that pass through all of the sieves (grains smaller than 0.06 mm (silt)). This material is mixed with a predefined volume of water to produce a homogeneous solution that is placed into a settling tube and the rate at which it settles out is assessed based on Stokes' sedimentation law expressing the relation between the diameter of spherical grains and the velocity of their sedimentation in a liquid. For swelling soils, the densimetric method is not particularly suitable due to their plate rather than spherical shape and their tendency to remain in suspension almost permanently. Determination of the finest grain fraction must be done using a particle sizing technique that can deal with these fines (e.g. laser light technique that looks at reflection and transmission of light through fines in suspension).

Figure 3.6 provides an example of samples of materials initially described as “clay” but that have very different grain size distribution characteristics. These plots show one of the key reasons for distinct differences in the behaviour of these two “clay” materials. The upper plot shows a material with a high “clay-sized” content while the lower shows essentially no clay-sized component. It can therefore be assumed that the material represented in the lower plot is not going to contain a substantial swelling clay component. While useful in preliminary screening, particle size analysis does not provide any information on the mineralogy.

(c) Specific density

The Specific density of soil particles also provides some initial indications of what the composition of a candidate material may be. Results of tests that fall substantially above or below the commonly accepted range for swelling clay minerals are indicators of potentially undesirable minerals.

Specific density of soil is expressed by the ratio of the weight of individual solid particles of soil dried at a temperature of 105-110ºC and their volume:

$$\rho_s = \frac{m_d}{V_s}$$

where m_d is weight of solid particles of dry soil and V_s volume of solid particles.

However, for more accurate determinations the pycnometer (basic apparatus for determining specific density of soil) can be used. The results for specific density (based on the standard methods described in CSN 72 1011/ASTM D1556-64) of different raw materials including reference clays, are presented in Table V-V. The specific density can then be calculated from the following relation:

$$\rho_s = \frac{(m_2 - m_1)}{V \times \rho_k + m_2 - m_3} \times \rho_k$$

where V is pycnometer volume, m_1 weight of empty pycnometer, m_2 weight of pycnometer with a dry sample, m_3 weight of pycnometer with a sample and additional liquid.

3.2.2.3. Swelling capacity of clay minerals

The detailed mineralogical and surface exchange characteristics of swelling clays have been discussed earlier in Section 3.1 of this document and it is the ionic composition and mineralogical structure that will control the swelling of a clay soil. Since montmorillonite have the majority of the cations associated with them active in balancing the negative charge of the mineral surface the composition of the ions will also dictate the swelling behaviour of these materials. Unlike most other clay minerals, when exposed to water, dry montmorillonite absorbs water in the interlayer region and the repulsive forces present between adjacent sheets are able to push the sheets apart. The degree of expansion observed depends on the cations located in the interlayer region. If the cation is monovalent and strongly hydrated (as Na^+), the repulsion between the sheets is stronger and the expansion is larger than for less hydrated cations.

Depending on the expansion between two montmorillonite sheets, two types of swelling mechanisms may be distinguished; crystalline and osmotic swelling. Crystalline swelling is a result of the hydration of the surface. Osmotic swelling is the repulsive osmotic force due to the interaction of the layers, limited only to some extent by Van Der Waal’s forces. For example, if the interlayer cation is polyvalent, the strong electrostatic attraction between the cation and montmorillonite surface limits the hydration of the cation and the spacing between clay sheets does not normally exceed 10 Å. If small and monovalent cations are present then the clay can take up more water and the interlayer spacing may increase abruptly up to 30-40 Å, continuing to increase to several hundred ångströms with water content.

There are two basic swelling-related tests that should be done as part of an initial screening process. The first is a simple free-swell test and the second a swelling pressure test.

Simple free-swell test is intended to provide a rapid indication of the swelling properties of the clay constituents present in raw materials. The test essentially consists of a known volume of dry clay being allowed unlimited access to free water and observing the volume that the hydrated clay occupies. In the absence of a well-defined swelling clay component (e.g. non-smectite or mixed layer clay), the measured mass to hydrated volume ratio would be low. It should be noted that this measure is highly sensitive to the composition of the water and so care must be taken to ensure that all tests are done using the same fluid. In order to compare reference clays with possible candidate clays, “free swelling” was determined using the technique described in the Febex report [55] as well as by the Centre for Experimental Geotechnics (CEG) of the Czech Technical University in Prague (Czech Republic) and these results are presented in the analysis of the test materials in Section 5.

A swelling pressure test involves measurement of the force applied by the clay to its confinement as it takes on water and achieves saturation. Ideally this can be done using a rigid confining cell that allows movement of water into the specimen while measuring the force developed, although use of a constant confining load and, allowing the specimen to swell (or compress) to a volume equilibrium, produces comparable results. A wide variety of cell sizes have been used to measure this parameter and a summary of information available from the literature was undertaken in the year 2000 [54] and this has been updated to provide an even larger body of data for comparison and discussion (see Sections 4 and 5). At lower densities the development of swelling pressure is strongly affected by the pore fluid present and so knowledge of the groundwater conditions present at a repository site is important in determining what swelling pressure can be developed by a particular bentonite material. Tools to predict the swelling pressure developed by a candidate material using mineralogical and chemical information have also been developed [58].

4. PROPERTIES AND FACTORS IMPORTANT TO POST-SCREENING PROCESS, EVALUATION AND QUALIFICATION

4.1. Introduction

This chapter provides a brief review of the process of buffer saturation and evolution within a repository environment where elevated temperatures, varying groundwater chemistry and ion-exchange processes are active in determining system behaviour and how they will affect the development of the key behavioural properties listed above. The most commonly identified properties needed of clays to be used for embedding canisters with HLW are:

- To minimize groundwater flow in the region immediately surrounding the canisters;
- To minimize transport of ions including radionuclides in the clay surrounding the canisters;
- To provide possibilities for gas caused by corrosion and radiolysis to be released at acceptable pressures;
- To allow heat transport from the canisters to the surrounding rock without causing unacceptably high temperature of the canisters and the embedding clay;
- To eliminate growth and migration of microbes;
- To provide sufficient bearing capacity and support for the canisters;
- To retain their physical properties for a sufficiently long time.

Associated with each of the above listed key properties are dynamic processes that will determine if a candidate clay is potentially suitable for use as a buffer. Through identification of key parameters, measurement of basic material properties and developing an understanding of how they interact, a candidate clay can be screened for its potential compatibility for a particular repository environment.

Processes that are of particular importance include:

- Water redistribution during pre-saturation period;
- Hydrothermal redistribution of soluble compounds and potential cementation of clay particles;
- Equilibration of pore water cations with surrounding groundwater and effects on hydraulic and swelling behaviour;
- Role of buffer density on hydraulic and swelling behaviour of buffer;
- Nature of gas transport within and through buffer an potential effects on subsequent contaminant movement;
- Transfer of heat from canister surface to surrounding rock mass and effects of density, water content and mineral composition on this process;
- Migration of contaminants within the buffer and effects of mineralogy and density;
- Microbial activity and their role in corrosion of canister;
- Deformation behaviour and strength of buffer materials under canister or other applied loading;
- Mineral alteration to non-swelling minerals under repository environment.

Each of these items are briefly discussed in the remainder of this section and representative references are provided to direct the reader to more detailed discussion of each topic.

4.2. Evolution of the clay surrounding the canister

4.2.1. The early saturation stage

The following changes with respect to temperature (T), hydraulic (H), mechanical, i.e. stress/strain (M), chemical (C), biological (B) and radiological (R) conditions will occur in the system of highly compacted smectitic blocks, confined by the overlying backfill and the surrounding rock, as can be seen in Fig. 4.1:

- Thermally induced redistribution of the initial pore water implying desiccation in the hottest part of the clay and wetting of its colder part that contacts the rock;
- Uptake of water from the rock and contacting backfill;
- Expansion of the clay leading to an eventual tight contact with rock and overlying backfill;
- Chemical processes within the clay and at the contact of clay and canisters. The dominating processes are those associated with salt, primarily Ca, Cl and SO_4 will migrate with the water that migrates from the rock. Precipitation in the hot part of the buffer will produce sodium chloride and gypsum, contributing thereby to the corrosion of the canisters.

The processes described above are often coupled in a complex way and so will cause transient changes in the material properties.

Water saturation of the pores present in the clay may require a decade or more for a canister-embedding clay in richly water bearing rock, several decades in tight rock of this type, and hundreds to thousands of years in very low permeability or unsaturated (e.g. argillaceous) rock. Many potentially important physico/chemical processes may proceed during this long period leading up to saturation, and may lead to significant changes in material performance, especially in the early phase when the temperature gradient is high. The ability of the surrounding rock to supply the buffer with water is therefore a key function in the design and performance assessment of repository concepts.

The initial degree of water saturation of the clay (S_r), which is expressed as the ratio of the volume of pore water and the total pore volume, is much less than 1.0. At low S_r-values the hydration potential, which is the driving force for saturating the clay, is very high which means that suction by the clay is strong. As degree of saturation increases this suction decreases to very low levels. The rate of saturation is controlled by the hydraulic conductivity and density of the clay but not significantly by the water pressure in the surrounding materials.

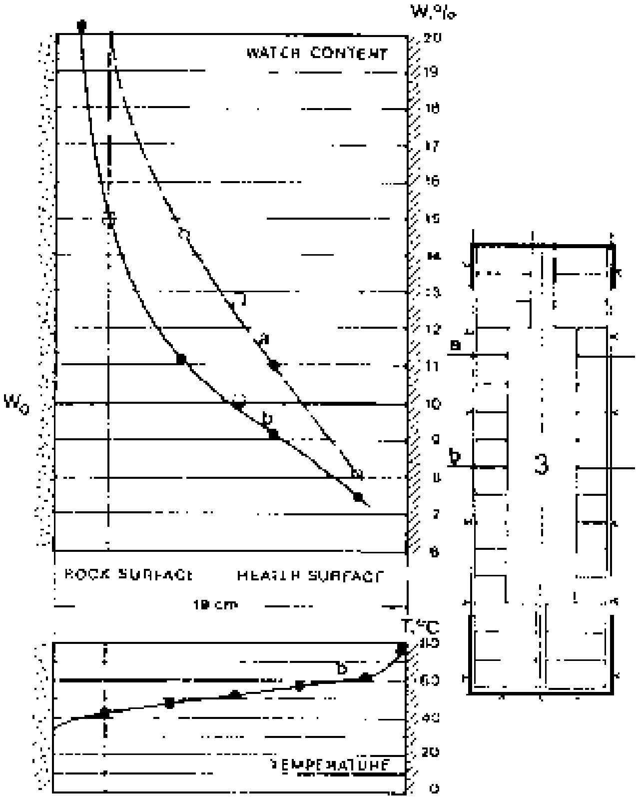

FIG. 4.1. Temperature-driven redistribution of water in very dense MX-80 clay surrounding a 600 W heater in the Stripa field experiment (Buffer Mass Test) after a few months [50].

4.2.2. The hydrothermal period

After complete water saturation the buffer enters a period of several hundred to thousand years in duration, under which hydrothermal (elevated temperature and full water saturation) conditions prevail. Under such conditions smectite degradation can occur and this process consists of dissolution and various alterations in the mineralogical composition of the buffer. In response to hydrothermal processes the smectite component of the clay can also undergo partial conversion to non-expanding minerals. In extreme cases this may result in the clay (or regions within it) becoming brittle due to cementation caused by precipitation of silica and iron released from dissolved minerals. Ideally, the amount of Fe- and K-bearing minerals associated with the buffer should be at minimum in order to limit these processes. Cementation of the otherwise expansive smectite particles can also result from chemical interaction between canisters and buffer, i.e. by formation of iron compounds. Some of these changes are slow and some may be reversible when the temperature gradient ultimately vanishes, thereby not threatening the overall long term performance of the buffer. Studies to quantify these processes are ongoing in a number of programmes examining the long term performance of smectites under repository conditions. Gamma radiation generated by the materials in the waste canisters is concluded to have only a very small impact on the physical and chemical states of smectite minerals. This has been documented by exposing clay samples (MX-80) to strong gamma radiation under hydrothermal conditions [50].

4.3. Physical properties

4.3.1. Hydraulic conductivity

A general picture of the importance of the mineralogical composition of clays with respect to the hydraulic conductivity is given by the diagrams in Fig. 4.2, which illustrates the dependence of the hydraulic conductivity (*K*) on density at saturation for differently composed clays. The upper curve in each diagram represents saline (ocean-type) pore water while the lower represents saturation and percolation with distilled water. The hydraulic conductivity of montmorillonite with a saturated density of 1600 kg/m^3 (dry density of 945 kg/m^3) is around 100 times lower than that of illite and as much as 100 000 times lower than that of kaolinite. For a saturated density of 2000 kg/m^3 (dry density of 1570 kg/m^3) the difference in hydraulic conductivity between these materials is smaller but still significant [32]. It should be noted that Fig. 4.2 represents very pure clay mineral materials. Most natural clays contain also non-clay minerals and are therefore more pervious than the materials presented in Fig. 4.2.

The risk of non-uniform distribution of the smectitic (e.g. montmorillonite) clay component in clays being considered as candidates for use as a buffer makes it necessary to require higher gravimetric clay contents than are theoretically needed to provide a material of acceptably low hydraulic conductivity. Thus, for practical purposes the minimum swelling content needed to guarantee uniform distribution and density of the clay component is about 10-15 %. The clay content is often taken as the content of smectite particles but this is not correct. Thus, using Wyoming MX-80 which like all other bentonite products is a mined natural material and therefore variable in nature material as clay component to preparing a mixture with 10 % clay means that the true weight percentage of smectite will only be about 7, since this bentonite has only about 70 % smectite content but can occasionally be less than that. The sensitivity of the quality of the swelling clay component becomes even greater in systems where the clay already has low smectite content (e.g. Friedland or Kunigel-V1) or is admixed with non-clay materials (e.g. quartz sand or graphite).

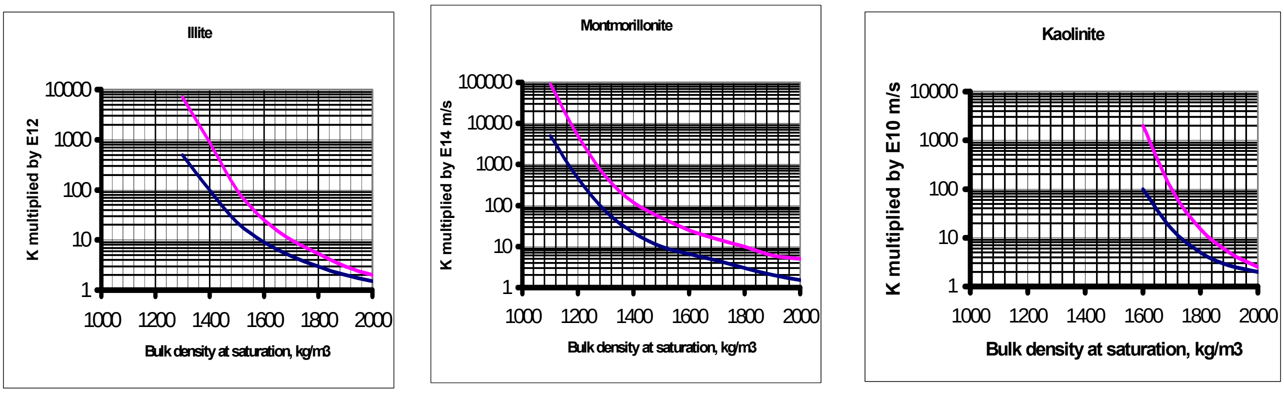

FIG. 4.2. Hydraulic conductivity (K) of pure clays as a function of density at saturation (the upper curves represent saline pore water; the lower ones represent distilled water).

In an effort to provide a means of comparing the hydraulic character of a variety of smectite-rich clays there has been considerable work done on the development of “normalizing” parameter(s). If it is possible to identify parameter(s) that can be applied to a variety of smectitic materials one will be able to estimate the hydraulic conductivity (*K*) of various material mixtures by determining only a few, key properties. Based on the already established hydraulic dominance of the smectite clay component of natural bentonites, work was originally done to normalize the hydraulic properties of a variety of smectite – sand materials and smectites of “similar” mineralogical and chemical character. The first stage was the use of a parameter defined as the effective clay dry density (ECDD), defined as the mass of “clay-sized” smectite-rich material divided by the volume occupied by the sum of the volumes of the clay-sized component and the voids. This parameter assumes that the non-smectite components including sand or other mineral additives are inert filler material that does not influence material behaviour. This approach was successful in generating a normalized hydraulic conductivity (K) – ECDD relationship for water-saturated “Wyoming-type” and Canadian bentonites, both of which have similar smectite contents and particle size distributions. With the generation of a relatively consistent relationship for these materials, others of substantially different smectite content were evaluated using this parameter. While the general relationship between ECDD and K was maintained these other lower smectite content materials generally exhibited values on the higher end of the range of hydraulic conductivity as can be seen in Fig. 4.3 (centre). Using the ECDD parameter it is possible to estimate the hydraulic conductivity of almost any smectitic or smectite-admixture knowing only the non-clay component content and the dry density of the compacted material.

As the ECDD normalizing parameter proved to be less than entirely satisfactory for lower smectite content clay materials (e.g. Kunigel-V1 bentonite of Japan, Friedland clay of Europe, Canadian glacial lake clay) further evaluation was undertaken. In an effort to broaden the applicability of the ECDD parameter, a revised parameter known as the effective montmorillonite dry density (EMDD) has been developed [52]. EMDD is defined as the mass of smectitic clay (typically montmorillonite) divided by the sum of the volumes of the smectite clay component and the voids. This means that the volume occupied by all non-smectite minerals or other components is subtracted from the unit volume before density is calculated. EMDD is

$$EMDD = \frac{f_m f_c \rho_d}{\left[1 - \frac{(1-f_c)\rho_d}{G_s \rho_w} - \frac{(1-f_m) f_c \rho_d}{G_n \rho_w}\right]}$$

where f_c is the clay fraction and f_m is the montmorillonite fraction in the clay and G_s and G_n are the specific gravities of the sand and the non-swelling clay.

This method of normalizing the density of the sealing materials has proven to be very effective, resulting in the generation of a generic EMDD–K relationship for smectite-dominated materials as is shown in Fig. 4.3, which indicates that the EMDD parameter can be used to estimate the hydraulic conductivity for a wide range of smectite-rich materials for which the swelling clay content has been determined. . The effects of not accurately knowing the swelling clay content can be seen in Fig. 4.3 and then again in Fig. 4.8, where Friedland clay was incorrectly attributed a swelling clay content of 45% rather than the 30% later identified as being more representative. The result of such compositional uncertainty is a plot that indicates that the clay has swelling pressure and hydraulic conductivity characteristics that cannot be normalized using the EMDD parameter. Although not shown in Fig. 4.3 and Fig. 4.8, the use of the more appropriate swelling clay content of 30% results in the Friedland clay plotting within the general range of the other materials examined. This also highlights the importance of quality control in production of swelling clay materials for repository use.

In Table 4.1, the dominating effect of pore fluid chemistry on hydraulic conductivity is shown. The type of smectite mineral is important in this context as illustrated by this table and Fig. 4.4 [54]. As shown in this figure it is necessary to have a good indication of the pore fluid chemistry as well as the exchangeable cation composition in order to obtain the greatest accuracy in using EMDD to estimate the K of smectite-dominated sealing materials. Provided that the type and content of smectite in the clay and the salinity are known it is possible to roughly estimate the hydraulic conductivity of the system.

TABLE 4.1. HYDRAULIC CONDUCTIVITY OF MX-80 AND FRIEDLAND CLAY PERCOLATED BY HIGH SALINITY FLUIDS [32]

	Density at saturation, kg/m^3				
		1600	1800	2000	2050
	Dry Density, kg/m^3				
		950	1260	1570	1650
Permeant % salt	Clay type	Hydraulic conductivity, m/s			
0.5 NaCl	MX-80	7E-12	2E-12	2E-13	6E-14
3.5 NaCl		-	E-11	E-12	E-13
0.5 $CaCl_2$		E-10	2E-12	2E-13	9E-14
3.5 $CaCl_2$		E-9	3E-12	3E-13	9E-14
2 NaCl	Friedland	2E-8	-	-	-
10 NaCl		5E-8	E-9	5E-11	2E-11
3.5 $CaCl_2$		E-9	6E-10	2E-11	-
10 $CaCl_2$		-	5E-9	3E-11	2E-11

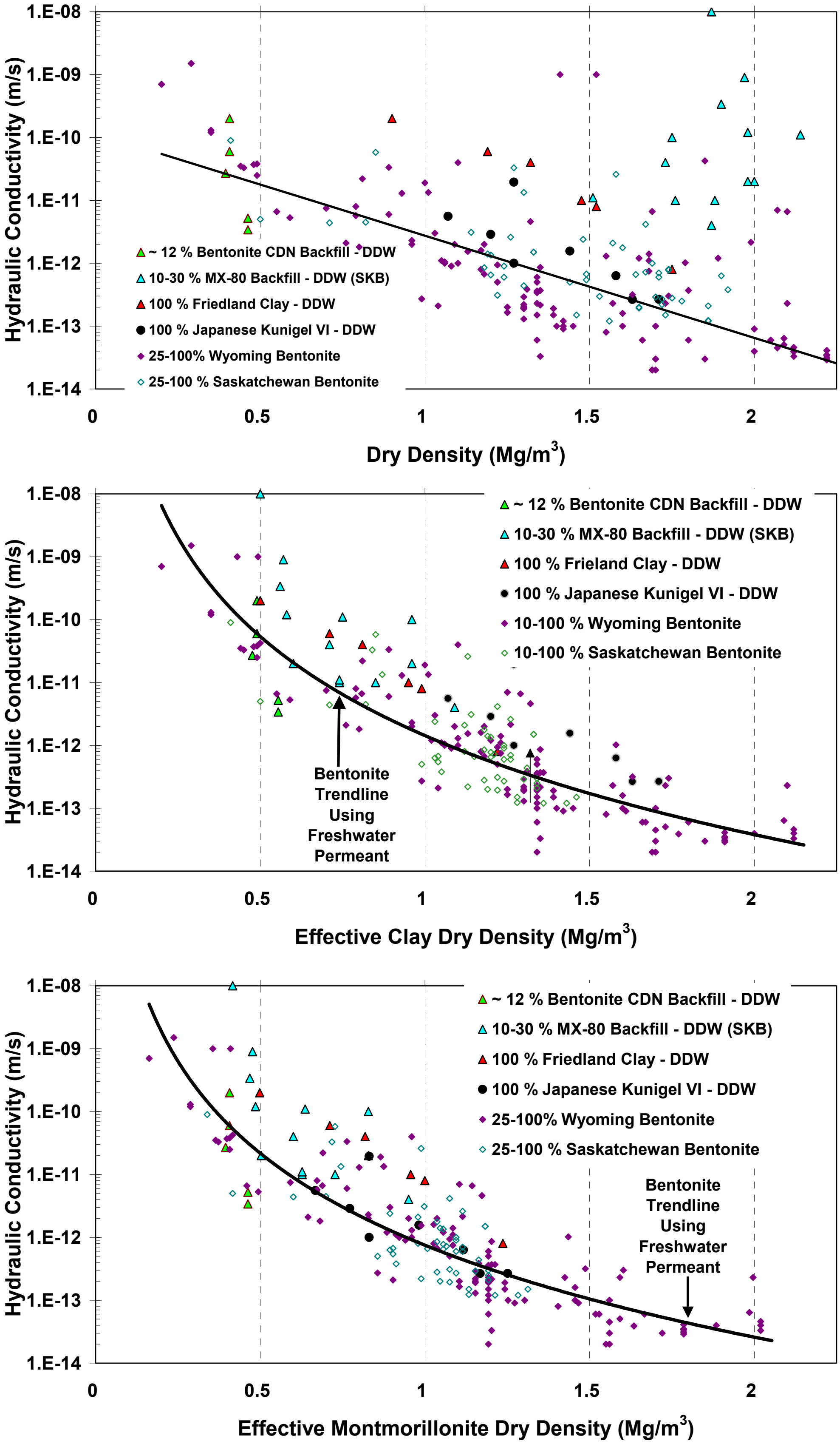

FIG. 4.3. Means of normalizing the density versus hydraulic conductivity for various swelling clays and clay-admixtures [52].

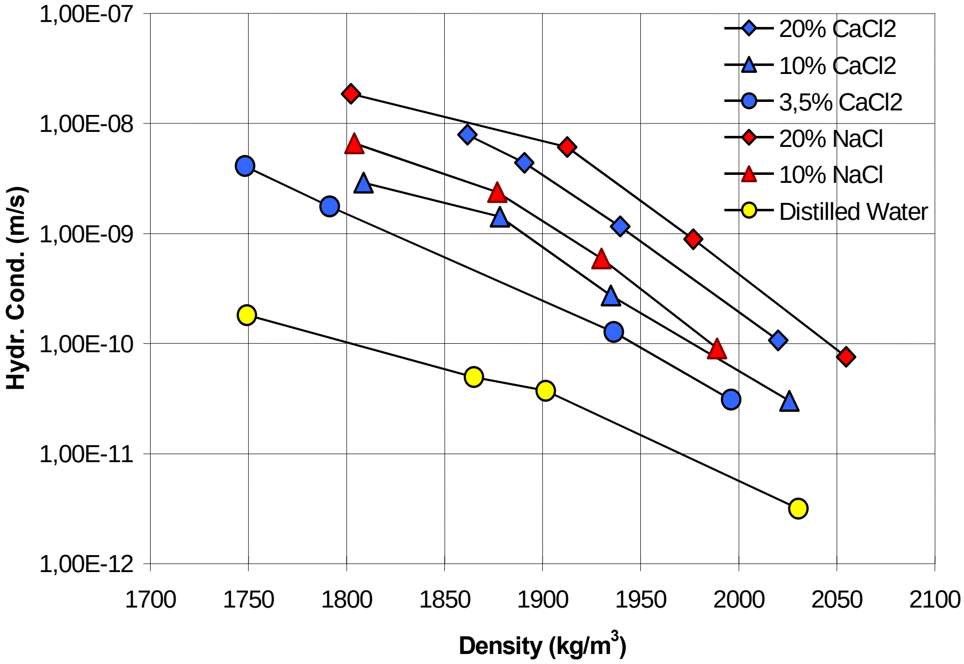

FIG. 4.4. Effect of pore fluid salinity on hydraulic conductivity of Friedland clay [32].

4.3.2. Gas conductivity

In practice one can assume that the gas conductivity is about a thousand times higher than that of water. This means that once gas has made its way through canister-embedding clay and further out through even more permeable geological units it will flow at a rate that is more dependent on the availability of pressurized gas than on the gas conductivity. The most important issue is therefore the gas pressure that leads to penetration of gas through the clay, i.e. the “critical gas pressure”. For temporarily stagnant gas in clay voids, the solubility of the gas in question — air, hydrogen or organic gases — is an important matter since the bubbles will shrink and the dissolved gas will diffuse out of the system.

According to current hypotheses the micro-structural heterogeneity has a decisive influence on the critical gas pressure: gas makes its way where it finds least resistance, which is along continuous channels of neighbouring larger voids where the capillary retention is at minimum or when the bond strength of adjacent particles is overcome [59]. Both views imply that the critical gas pressure is on the same order of magnitude as the prevailing effective pressure (“grain pressure”). Examples of gas pressure tests in 300 L permeameter are shown in Fig. 4.5. The initial drop of inflow rate observed was attributed to elastic strain of the permeameter and the clay after which steady flow took place. The increased inflow rate after a certain period of time was due to the successively increased pressure gradient present across the remaining specimen length when the gas front approached the discharge filter.

At low densities and gas pressures, gas entering the clay may cause consolidation and local densification, which may raise the critical pressure if the pressure steps are small or the gas pressure is left on for a long time. At high densities such impact is smaller but practical difficulties in determining the true critical pressure may arise since leakage along the confining cell and the clay may take place.

The nature of the host rock determines the possibility of gas accumulated in the buffer to dissipate. The excavation disturbed zone (EDZ) around holes and tunnels in crystalline rock in combination with the ubiquitary permeable fractures certify that gas is released at a pressure that only insignificantly exceeds the piezometric pressure. For argillaceous rock it can be different because of its tightness although an EDZ is also developed in that rock.

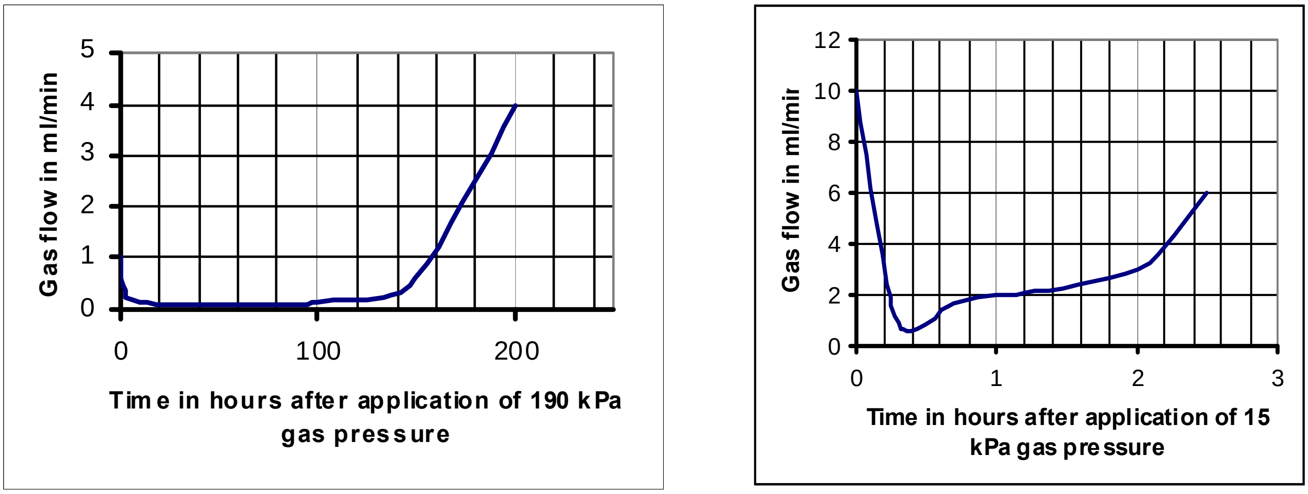

FIG. 4.5. Examples of gas penetration process at the critical pressure. (Left: Smectite-rich bentonite with 1680 kg/m^3 density at saturation with seawater (swelling pressure 200 kPa, K=4E-11 m/s); right: 10/90 mixture of Na-bentonite and graded ballast, density at saturation with seawater 2200 kg/m^3 (swelling pressure 50 kPa, K=2E-9 m/s) [32].)

4.3.3. Heat conductivity

Heat transport is a function of interacting parameters, mainly: compacted density, degree of water saturation, clay mineral composition and the type and quantity of non-clay minerals (especially coarser-grained crystalline particles such as quartz, graphite or crushed rock). The blending of smectite and non-clay components into a material for use in repository sealing applications will ultimately be a decision that is made based on the balancing of the needs for thermal conduction, swelling and hydraulic properties, and longer term processes active in a repository.

The heat associated with the radioactive decay in the waste should be effectively transferred to the surrounding rock since temperature rise to more than the boiling point can otherwise cause vaporization of the pore water and desiccation of the buffer and speed up degradation of the smectite minerals. Pore water, quartz and well-crystallized graphite are good heat carriers but for high densities water is a less effective heat conductor and addition of quartz and graphite particles can be made to raise the conductivity if this is required. Attempts to do so have been successful in large-scale experiments by AECL in Canada and RAWRA in the Czech Republic. The risk of non-homogeneous distribution of any admixtures must be considered, however.

Commercially available smectite-rich clays with a water content of 10–15%, which corresponds to 1 to 2 inter-lamellar hydrate layers, and compacted to a dry density of 1500 kg/m^3 have a heat conductivity of about 0.75 W/m·K. An increase in water saturation from 50 to 100% raises the heat conductivity by around 100% and further significant rise can be reached by adding a quartz component to the system. Fig. 4.6 shows the change in the thermal conduction characteristics of a bentonite-sand system at various degrees of water saturation and it also shows the influence of saturation in a bentonite-only system. The effective stress and temperature are known to have a certain, rather small effect on the heat conductivity [32]. A high effective stress gives good inter-particle contact, which is manifested by the higher heat conductivity of denser clays. This rise is counteracted by a reduction in water content. Increased temperature slightly improves the heat conductivity since heat transfer is aided by the presence of convection circulation.

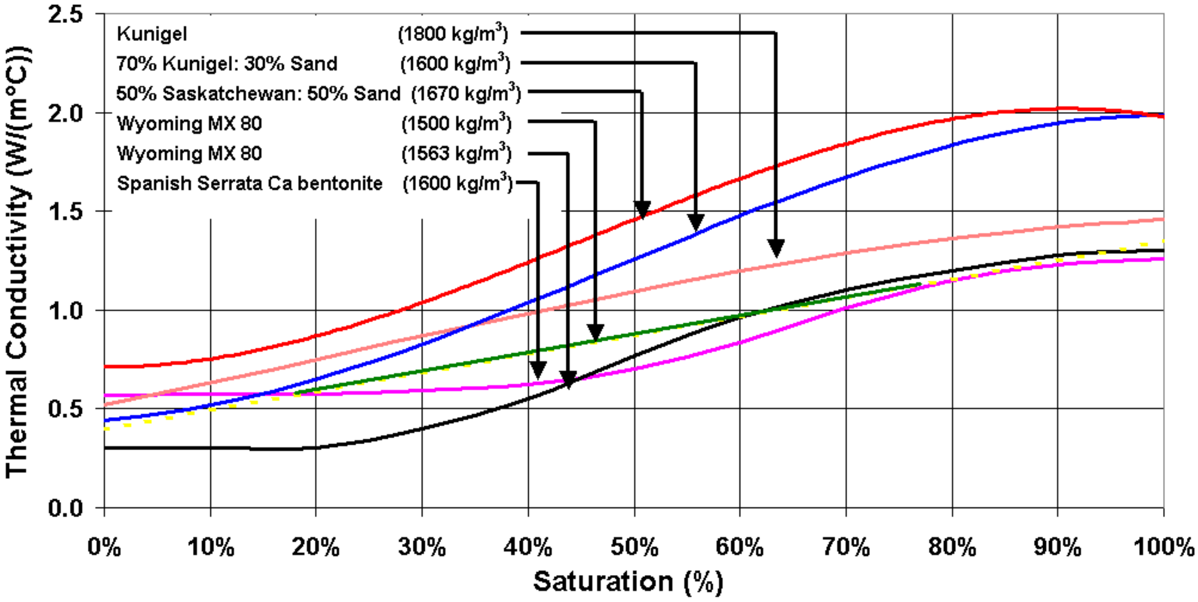

FIG. 4.6. Influence of degree of water saturation, dry density and aggregate component on thermal conductivity [37].

4.4. Ion migration and sorption

The transport rate of dissolved chemical material in clays depends on the diffusivity of ions and molecules under the influence of concentration gradients. Knowing these rates are important because they can determine the rates at which corrosion-inducing chemicals can reach, interact with and then move the corrosion products away from the canister or other system components (e.g. concrete). Additionally, the rate and manner of radionuclide movement will be strongly affected by the diffusion characteristics of the material(s) it must move through. Hence once a candidate swelling clay is identified by screening it will be necessary to determine its more complex behavioural characteristics.

The diffusion transport capacity is expressed by the “effective” diffusion coefficient, which refers to the actual “effective” porosity and which gives information on the ion transport on the micro-structural level in contrast to the “apparent” diffusion coefficient, which is derived directly from curve-fitting using recorded concentration profiles. The use of porosity-related diffusivities means that one can distinguish between transport of cations and anions, and between cation diffusion in continuous water-filled voids, along particle surfaces with electrical double-layers, and through the inter-lamellar space in smectites. The latter two mechanisms involve ion-exchange mechanisms for which a sorption parameter, K_d, is used [32].

In practice, the ion transport capacity can be predicted by applying Fick’s law and relevant values of the coefficient of the density-related “effective” diffusion, D_e. The density of the clay plays an important role except for monovalent cations as illustrated by Fig. 4.7. It demonstrates that D_e of the anion chlorine is reduced by two orders of magnitude if the dry density is increased from 500 to 1500 kg/m^3 (1200 to 1950 kg/m^3 at fluid saturation) for which D_e is about E-11 m^2/s. The curve for neutral molecules is approximately valid also for water.

The diffusive anion transport capacity is proportional to the ratio of the pore space of the voids between the stacks of smectite lamellae since they are excluded by the Donnan effect from the inter-lamellar space. With increasing density there is a strong reduction of the available space for migration and the diffusion coefficient of anions therefore drops significantly. Many cations move both by pore diffusion and surface diffusion including both the inter-lamellar space and the close vicinity of the stacks of lamellae where electrical double-layers with dominant cationic population exist. Hence, the retarding effect of raising the density on the diffusion capacity of cations is limited, especially for monovalent ions. Some typical effective diffusivity (D_e) data are presented in Table 4.2.

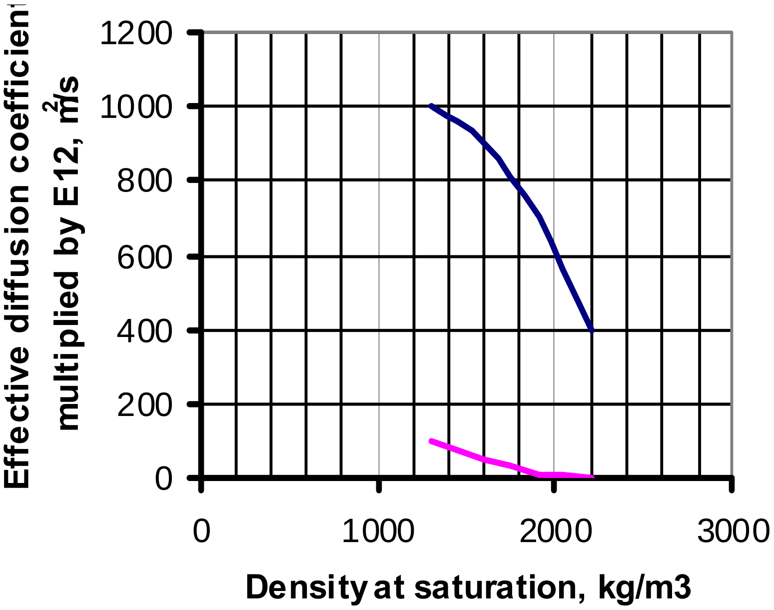

FIG. 4.7. Measured effective diffusivities for smectite clay. (Upper curve: monovalent cations; Lower curve: monovalent anions like chlorine [60]).

TABLE 4.2. EFFECTIVE DIFFUSION COEFFICIENT D_e FOR ELEMENTS MIGRATING IN MX-80 CLAY WITH A SATURATED DENSITY OF ABOUT 2000 kg/m^3 [50]

Species	D_e (m^2/s)
C-14	E-10
I-129	2E-12
Sr-90	2E-8
Cs-137	2E-9
Na-22	E-9
Pu-238	E-10
Am-243	E-10

4.5. Microbiology

Interaction of organic species with smectite clays in general has been an important issue for a long time especially with respect to the performance of microbes. In recent years the matter has become particularly important in conjunction with the design of repositories with radioactive waste. The requirement is that microbes must not penetrate the clay buffers since they can form organic colloids that can migrate to the biosphere bringing with them radionuclides. The problem has been studied from different perspectives, one of them being the micro-structural constitution and the possibility for microbes to survive and migrate that it can offer.

MX-80 clay with densities lower than 1200 kg/m^3 at water saturation has sufficiently large voids to host large amounts of bacteria and allow them to multiply and migrate by breaking gel bonds although fixation to smectite clay by hydrogen bonds puts limits to the mobility [32]. The frequency of relatively large voids in MX-80 buffer clay with a bulk density at water saturation of less than 1800 kg/m^3 may be sufficient to host bacteria. Certain bacteria like *desulfotomaculum nigrificans*, which play a role in the degradation of copper canisters, can survive in certain voids in the softest clay gels at this bulk density since they provide sufficient access to free water while enough nutrients for multiplication and spore production may not be available. At higher bulk densities bacteria are not able to move in the clay because of the limited space and the mechanical strength of the clay gel and they will likely ultimately lose their potential to produce spores.

4.6. Mechanics

4.6.1. Swelling capacity and swelling pressure

The expandability of smectitic clay is a critical property since it provides a self-sealing potential and makes sure that tight contact with the surrounding rock is attained. It is determined by use of a rigid oedometer equipped with a load gauge, determination of “free swell” offering a first indication of the expandability of candidate clays.

The major influencing factors on swelling capacity are the density, pore water salinity and type of smectite and adsorbed cation, and the temperature. The complete mechanism that results in swelling pressure is not fully understood. Most of the swelling pressure observed in a confined system results from the repulsive forces present in the hydrated system of cations in inter-lamellar positions and the osmotic pressure between basal surfaces of stacks of lamellae. Elastic energy stored in strained mineral crystals may add to these pressure components. Other environmental, structural and chemical factors will also play a role in what swelling pressure develops. The validity of the effective stress principle for smectites as well as for any other soil has been proven and it is assumed that the swelling pressure is equivalent to the effective compressive stress and the total pressure experienced by a surface adjacent to a swelling clay will be the sum of the swelling pressure and any pressure contribution from the pore water [32, 50].

Swelling pressure data for montmorillonite-rich clay (MX-80) and Friedland Clay saturated with low-electrolyte water are given in Table 4.3. It shows that montmorillonite-rich clay with its exchange capacity dominated by sodium has a swelling potential even at very low densities, while Friedland Clay has little to none.

TABLE 4.3. SWELLING PRESSURE (p_s) IN MPa OF A NUMBER OF TESTED BUFFER MATERIALS SATURATED WITH DISTILLED WATER [32]

Density at saturation, kg/m^3	1300	1500	1700	1800	1900	2000	2100
MX-80	0.06	0.2	0.4	0.8-0.9	1.4	4-5	10-12
Friedland	-	-	0.05	0.1	0.3	0.8-1	2-2.5

The “normalizing” parameter EMDD can be used for estimating the swelling pressure (p_s) of materials by determining only a few, key properties [52]. This method of normalizing the density of the sealing materials has proven to be extremely effective, resulting in the generation of a generic EMDD–p_s relationship for smectite-dominated materials as is shown in Fig. 4.8 where the various density parameters are plotted. It should be noted that the data shown in this figure was collected from as many international sources as possible and represent clay materials such as MX-80, Wyoming bentonite, Saskatchewan bentonite, Friedland clay, Kunigel-V1 and others. As noted in Section 4.3.1 the effect of not accurately knowing the swelling clay content of a candidate material can have a notable effect on the EMDD calculated for a material. This is illustrated in Fig. 4.3 where Friedland clay shows a consistently higher hydraulic conductivity than other bentonite materials examined and was caused by incorrect assumptions regarding its mineralogical composition (swelling clay content 30% rather than 45% initially assumed).

Like the hydraulic conductivity, the swelling pressure is a function of the density but also on the pore water chemistry as illustrated in Fig. 4.9. This figure clearly shows the effects of pore fluid chemistry and that positive swelling pressure is recorded even at very low densities, at which more smectite-rich clays show even higher pressures. Work done in Japan has also resulted in the development of a numerical method for prediction of swelling pressure development [58]. Lacking the means of physically testing to confirm swelling pressure behaviour in the initial screening process this numerical assessment approach can also be used.

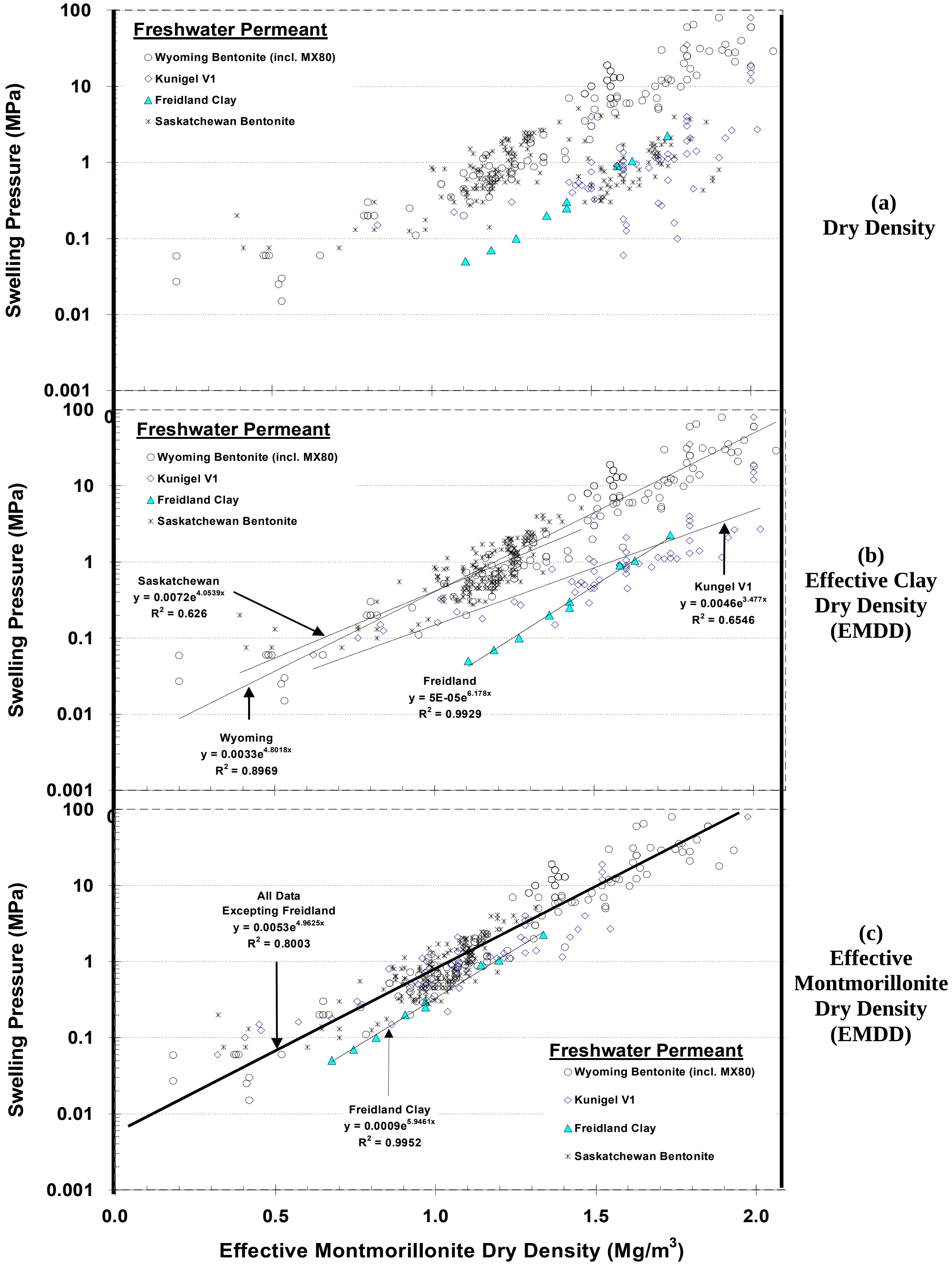

FIG. 4.8. Use of EMDD as a means to predict swelling pressure in smectite-rich clays [52].

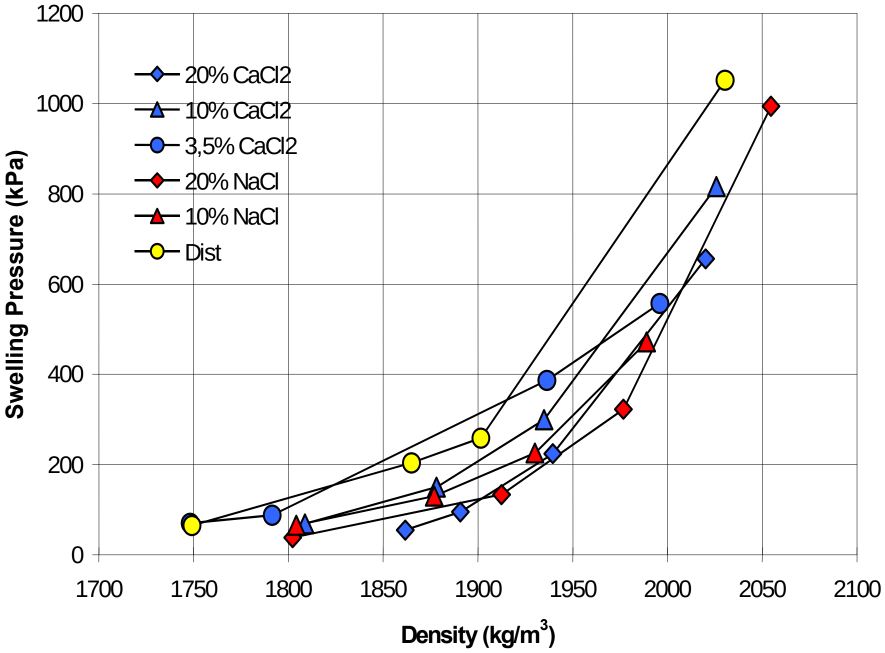

FIG. 4.9. *Swelling pressure tests on fluid-saturated samples of Friedland clay [32].*

4.6.2. Compressive and tensile strength

In the repository environment, there is a need for the sealing materials to be able to be placed into position without being mechanically compromised, to be able to support whatever mechanical load they are exposed to without physically failing, and then a need to remain stable over the longer term such that system performance is never compromised by their behaviour.

The basic measures of the ability of the sealing materials to achieve these goals are their compressive strength and their tensile strength. Both of these parameters are controlled by a combination of micro-structurally-determined characteristics, predominantly density and suction. The swelling clays proposed for use in a repository must be able to be placed at densities capable of supporting whatever mechanical loads are required (e.g. canister) and so an important post-screening characterization step will be to assess the fundamental stress-strain behaviour of the proposed materials. In principle, a high smectite content improves the mechanical strength.

4.6.3. Ductility

The way in which clays in general and smectites in particular can undergo large strain without losing coherence are particularly important in determining their suitability for use as a sealing material. The manner in which they strain and the results of this on the performance of sealing materials are factors that require careful consideration and assessment when doing post-screening evaluation of candidate smectite clays. The basic test known as consistency (or Atterberg) limits provides a screening tool that is used to assess the swelling clay content and begin the process of assessing ductility and mechanical performance. More complex physical characterization tests are required to develop a fuller understanding of the deformation characteristics of clay-based materials and complex numerical assessment tools are being developed in an attempt to add a longer-term predictive ability to materials behaviour evaluation.

4.6.4. Rheological properties

Determination of rheological properties is not a standardized procedure and is often made by different techniques and instruments in different laboratories. In principle, a high smectite content enhances the typical geotechnical properties of high-plastic clays and hence the importance of time-controlled processes like consolidation and creep. For predicting the performance in situ a number of hydro-mechanical (H/M), thermal/hydraulic/mechanical (THM), and a limited number of THM/chemical (THMC) codes and models have been and continue to be developed [32].

Compression and expansion

Compression and expansion of smectite-rich clays are largely reversible since there is, in principle, a unique relationship between density and confining effective pressure. Some hysteresis is involved but almost the same swelling pressure results from compressing soft smectite clay to a certain density and when denser clay of this type is allowed to expand to the same density. This is not the case for non-expandable clays, like illite and chlorite, because the microstructure undergoes largely permanent changes by compression.

Elasticity, plasticity, viscosity, creep

Buffers and backfills are ductile and typically behave as elasto/visco/plastic media. They have no true pre-consolidation pressure like natural sediments but are characterized by a certain relationship between density and swelling pressure, which is interpreted as effective pressure. Loading of buffer clay by a canister will cause consolidation, defined as time-dependent compression in conjunction with expulsion of pore water, the rate being controlled by the pressure increase, the hydraulic conductivity, and the shear resistance.

4.7. Longevity of smectites

The sealing materials in a repository are required to function for very long times following canister installation. As a result it is necessary to establish that the materials being considered for use are stable over the time required for effective waste isolation. The primary mechanism that has been identified as potentially compromising the long term durability of the sealing materials is hydrothermal alteration. Hydrothermal conditions can ultimately cause conversion of smectite minerals to illite and chlorite. The best example of this process at work is the well-documented successive alteration of deeply located sediments in deep-sea sediments of pre-Quaternary age. In these materials mineralogical alteration is associated with changes in the void size distribution and release of silica and aluminium in the presence of potassium ions [61]. The silica easily migrates and precipitates in regions where the temperature is lower, while the aluminium is more or less retained where it was released. These changes are expected also in canister-embedding clay in HLW repositories.

4.7.1. *Illitization*

Various investigations have shown that conversion of smectites yields non-expandable clay minerals, primarily as illite (hydrous mica) and chlorite. The general reaction is assumed to be:

$$\text{Smectite} + K^{+} + Al^{3+} \longrightarrow \text{Illite} + Si^{4+}$$

This type of reaction is believed to be valid for the entire smectite family but the activation energy for conversion and dissolution is different and for example in the beidellite-subtype potassium uptake may directly lead to collapse of the stacks of lamellae to yield illitic minerals.

While montmorillonite is the most common smectite species in nature, and has the best gelation properties, extensive experience in deep drilling shows that saponite is not affected by calcium and salinity as much as montmorillonite, and is considered to have a higher chemical stability at high temperature. This point to a higher activation energy for conversion of saponite to illite in comparison to a montmorillonite to illite conversion.

Numerous examples of illitization provided by the nature indicate that Pytte's theory for smectite-to-illite conversion can be used for estimating the risk of such changes [50, 32]. It is a thermodynamically based theory according to which the rate equation is:

$$-dS/dt = [Ae^{-U/RT(t)}]\,[(K^+/Na^+)^m S^n]$$

where: S = Mole fraction of smectite in *I/S* assemblages, U=Activation energy, R = Universal gas constant, T = Absolute temperature, t = Time and m, n = constant coefficients.

Figure 4.10 shows the rate of illitization for an activation energy of 24 kcal/mole to yield conversion from smectite. The model implies that heating to 50°C causes only insignificant loss of smectite in 10^6 years, while about 100% of the original smectite turns into illite in this period of time at 100°C. A typical temperature history for UF is: (a) 100-150°C in the first hundred years, (b) followed by a 500 year period with an average temperature of 50-100°C, and (c) less than 50°C in the subsequent 1000 years, leading to conversion of 15% of the initial smectite content to illite in 1500 years [50, 32].

4.7.2. *Precipitation of cementing compounds*

Conversion of the smectite minerals to non-expanding ones, primarily illite, is associated with release and migration of silica according to the equation provided above and precipitation of silica, where lower temperature prevails, yielding cementation [50, 32]. This process may be the most important degradation process in buffer clays but under the repository thermal and chemical conditions defined for an SKB-type repository it is not deemed to be so prevalent as to threaten the isolation function of smectite-rich canister-embedding clay [62].

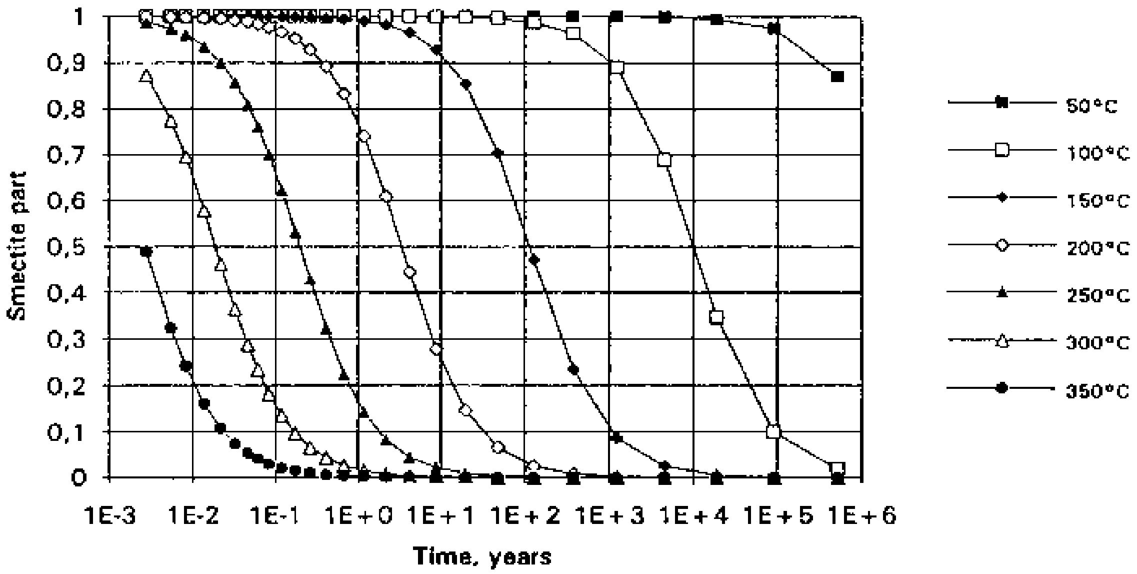

FIG. 4.10. Diagram showing predicted conversion of smectite to illite for the activation energy 24 kcal/mole [50, 32].

Experiments with Fe-rich smectite clays with Fe located in octahedral positions in the crystal lattice and containing Fe hydroxide compounds indicate that Fe can be released from heated clay and transported to become precipitated at cooling, causing cementation and drop in ductility of the clay [32]. As a result, the potential for reactions between clay and contacting iron and steel canisters that yield both cation exchange to Fe in the clay and precipitation of iron compounds at least early after disposal of the canisters when oxygen can still be available in the system cannot be discounted [50]. If either of these processes associated with Fe is active then there is a potential that the swelling clays might lose or see substantial reduction in their ability to expand to fill any voids or to close any cracks that might be generated in the buffer material during evolution of the system. Candidate clays will therefore need to be carefully assessed for Fe content and the nature of this iron determined.

5. RESULTS OF TEST CASES TO EVALUATE CLAY MATERIALS AS POTENTIAL SEALING SYSTEM COMPONENTS

5.1. Bentonites examined in this study and some basic materials properties

An important aspect of this CRP is the comparison of relatively unknown clay materials from member states, to more fully characterized clay deposits already identified as being potentially suitable as buffer material. This comparison provides a basis for assessing the likely suitability of these lesser known materials as candidate components in a repository sealing system. A short description follows of the candidate materials selected by the CRP members for test case studies.

5.1.1. China

Based on the literature investigation [63], swelling clays, such as bentonite, were identified in 23 provinces or autonomous regions in China. Up to now, 84 bentonite deposits have been discovered with various scales. Among them, there are 12 large scale bentonite deposits each with reasonably assured reserves of more than 50 million tonnes, 26 middle scale deposits, each with reasonably assured reserves of 5 million tonnes to 50 million tonnes, and 45 small scale deposits, each with reasonably assured reserves of less than 5 million tonnes.

Four selection criteria have been identified for use in locating a potentially suitable sodium bentonite deposit for use as the candidate supply of buffer/backfilling materials for China’s high level radioactive waste repository and these are as follows:

(1) Deposit size: The selected candidate bentonite is a very large-scale deposit (reasonably assured reserves greater than 50 million tons) and should be large enough in order to meet the demand for the installation of HLW repository;

(2) Quality: The selected bentonite should have the following properties:

- Low permeability;
- High swelling capacity;
- Suitable thermal conductivity,
- Reasonable stability (chemical and mineralogical);
- Na-bentonite with high content of montmorillonite and low contents of detrimental accessory compounds (chemical and mineralogical); and
- High cation exchange capacity.

(3) Location: Transportation of bentonite from the selected deposit to the candidate HLW repository site should be convenient and the cost should be as low as possible.

(4) Economic limitations and Environment Impact: The selected bentonite deposit should satisfy the mineability and economic feasibility conditions that include ease of open-pit mining and have minimal impact on the environment.

Based on these selection criteria, five large-scale bentonite deposits have been located and are shown in Fig. 5.1. Following systematic comparison and evaluation of these potentially suitable deposits, *Gaomiaozi* bentonite deposit, Xinghe County, Inner Mongolia Autonomous Region (Location 2 in Fig. 5.1), has been selected as the preferred candidate to provide swelling clays for HLW repository in China.

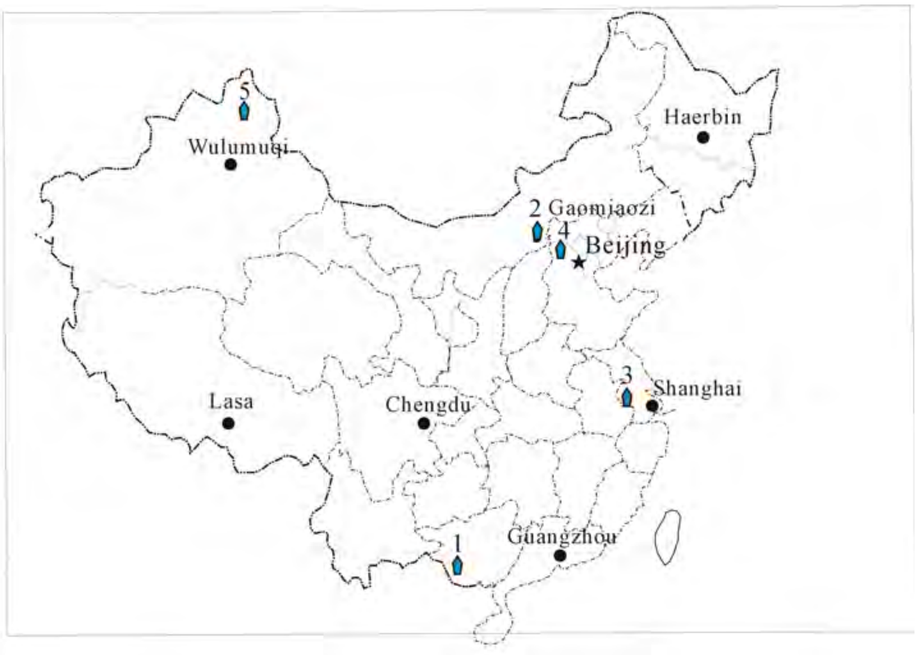

FIG. 5.1. Location of Major Bentonite Deposits in China (1. Guangxi Ningming deposit in Guangxi Autonomous Region; 2. Gaomiaozi deposit in Inner Mongolia Autonomous Region; 3. Jurong deposit in Jiangsu Province; 4. Xuanhua deposit in Hebei Province; 5. Wulanlinge deposit in Xinjiang Autonomous Region).

The Gaomiaozi deposit is the third largest bentonite deposit in China. The total reasonably assured reserves of the bentonite in Gaomiaozi deposit is about 160 million tons with about 120 million tons of Na-bentonite identified through detailed explorations. In the Gaomiaozi deposit, strata are dominated by upper Jurassic volcanic sequences unconformably overlapping Archean meta-sediments. Upper Jurassic rhyolite and tuffs are the main host rocks of the bentonite ore bodies. Five interbedded bentonite layers have been identified in volcanic sequences. There are four ore types of bentonite including: brecciaed bentonite; clayey bentonite; sandy bentonite and sandy bentonite with pebbles. Ore minerals are dominated by montmorillonite, with lesser amounts of illite as the main accessory clay mineral. While the gangue minerals are mainly composed of feldspar, quartz, calcite and zeolites. The deposit is located in an arid and remote area.

The geo-tectonics and hydrogeology conditions are relatively simple, which can satisfy the open-pit mining and have less impact to the environment. This material has therefore been selected for inclusion in this coordinated research project (CRP) in order to evaluate its properties relative to other materials considered for use in other national programmes.

5.1.2. Czech Republic

The Czech Republic has within its borders large bentonite deposits, the locations of which are shown in Fig. 5.2. The bentonites found within the Czech Republic are similar to each other and are mostly calcium-magnesium types. Na-bentonites are not found in the Czech Republic, and therefore, domestic bentonites must be enriched with sodium in order to activate them and thereby improve swelling, hydraulic and other properties. The Czech Republic extracts around 70 000 tonnes of bentonite per year, which amounts approximately to 0.75% of the world bentonite production [64]. Among bentonite producers in the Czech Republic, there are Keramost, a.s. (the biggest Czech producer of bentonites), Calofrig Borovany, a.s., Sedlecký kaolin a.s., and others.

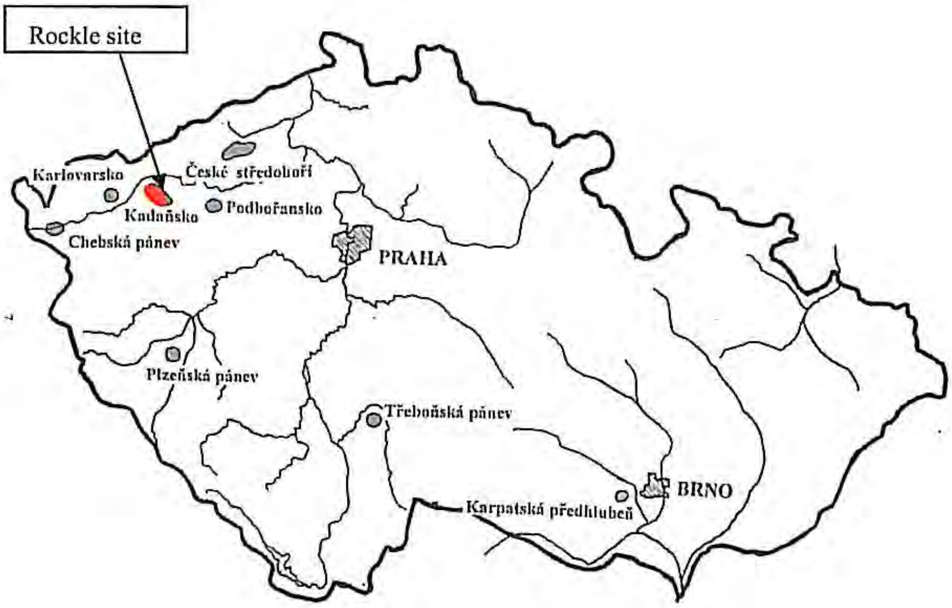

FIG. 5.2. Locations of bentonite deposits in Czech Republic.

Bentonites are found predominantly in the North Bohemian volcanic area (Fig. 5.2) – in České Středohoří and in Doupovské mountains area (Kadaň, Podbořany, Karlovy Vary) but also in the areas of Tertiary Basins (Cheb, Plzeň and Třeboň basins).

Following some basic mineralogical, physical, chemical characteristics screening, resource estimation and material evaluation, the bentonite found in the *Rokle* locality (RMN) near Kadarisko was chosen as a candidate for use in an underground repository in the Czech Republic. This commercially available material is a product that has not had its properties altered by chemical treatment and even without chemical activation, the bentonite from Rokle was classified as clay with extremely high plasticity (based on ČSN 73 1001 classification system).

5.1.3. India

India has substantial reserves of swelling clays located in various parts of the country. A smectite-rich deposit containing about 20 million tonnes has been identified in the western regions of the country. Other potentially suitable deposits of swelling clays are located in different regions of the country at Gujarat, Rajasthan, Bihar and Chennai (as shown in circled regions of Fig. 5.3).

FIG. 5.3. Location of bentonite deposits in India.

Naturally available swelling clay minerals like vermiculite, zeolites and montmorillonite have been used in India to augment the soil characteristics of the various waste disposal sites in near surface disposal facilities co-located with power/research reactors in various parts of the country for disposal of low and intermediate level solid waste.

Unlike most of the world’s bentonite deposits, clays from western region are the result of in situ alteration of older volcanic rock. As part of a preliminary screening process before identification of the most likely sources of materials, 26 swelling clay samples were recovered from different mineral deposit. These naturally available minerals have been analysed for CEC, physico-chemical characterization and sorption behaviour with respect to various radionuclides. Swelling material from the western area of India (possibly *Gujarat*) was forward as candidate material for evaluation in this CRP.

5.1.4. Republic of Korea

The general geology of the area in which the bentonite occurs in the Republic of Korea is shown in Fig. 5.4. The area contains Cretaceous igneous and sedimentary rocks and Tertiary volcanic and sedimentary rocks, in which the Tertiary rocks overlie those of the Cretaceous. The bentonite, as shown in the figure, is widely distributed in the Janggi Series and Yeonil Series. The Janggi Series, which is exposed in Janggi, Gampo, and Haseo areas, contains volcanic rock and tuff, whereas the Yeonil Series in Pohang area does not contain any tuffaceous rocks. Most of the sediments of the Janggi Series are in shallow fresh water, but those of Yeonil Series are deposited in a near-shore marine environment. The bentonite beds vary in thickness from a fraction of a centimetre to more than 20 metres. Some thick bentonite beds are composed of several layers and appear to represent the alteration products of a series of ash falls rather than single one. The reserves of the bentonite are estimated to be approximately 5.8 million tonnes [65].

The present design concept [66] of a HLW repository in the Republic of Korea includes use of domestic bentonite as both buffer surrounding disposal canisters and separating them from the host rock, and backfill for tunnels and access shafts. The performance criteria of buffer material, in this connection, were defined [67] and then the domestic bentonite deposits were investigated and screened to select a potential bentonite deposit that satisfied the performance criteria and provided sufficiently the required quantities of bentonite for a HLW repository. Jinmyeong mine, a bentonite deposit in Gampo area, Janggi Series was selected as a potential bentonite deposit for a HLW repository. The bentonite is referred hereafter to as *Gampo* bentonite.

Gampo bentonite is not a sodium-dominated material, its predominant exchangeable cation being calcium. This Gampo bentonite is a very well characterized material as the Republic of Korea has been evaluating this material for many years as part of its nuclear waste management programme.

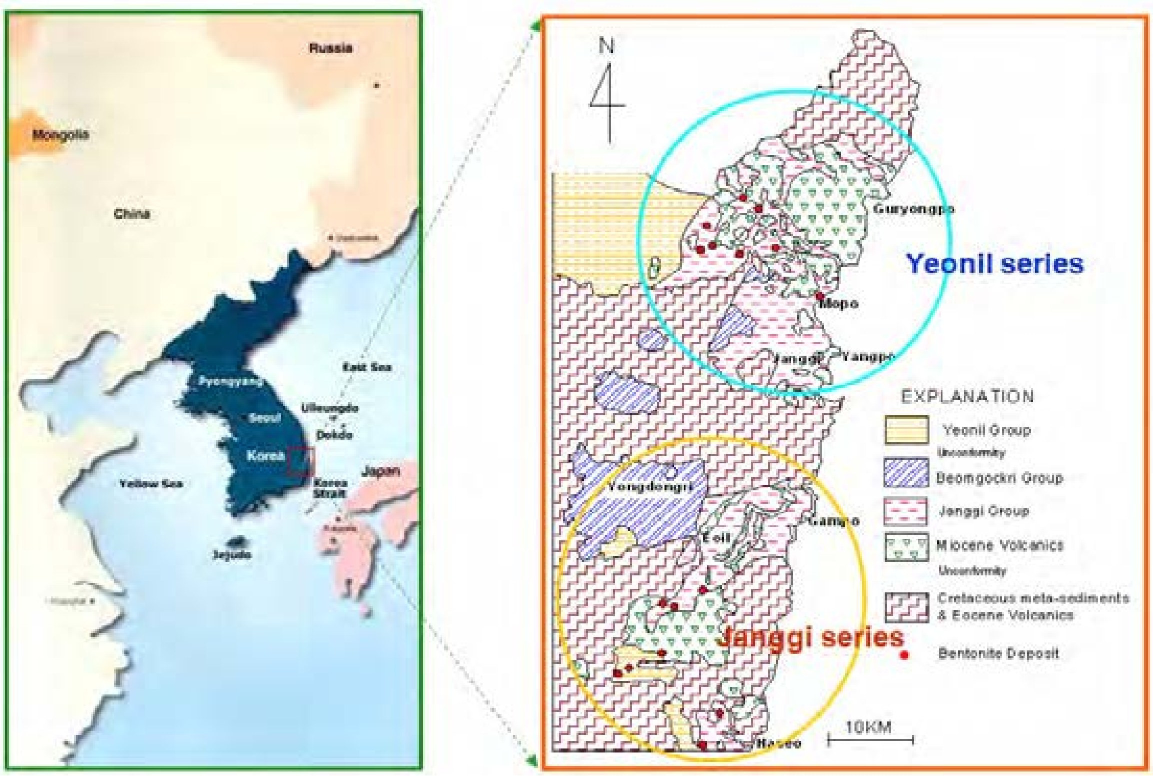

FIG. 5.4. Map of bentonite deposits in the Republic of Korea.

5.1.5. Russian Federation

In comparison to other mineral resources, bentonite resources are not abundant in Russian Federation. Fig. 5.5 shows the major discovered deposits with the estimated reserves listed in Table V-1.

The *Pikalevskoe* bentonite deposit in Leningrad region of north-western Russian Federation, shown in Figure 5.5 as location 16 (circled), was discovered in 1996. The bed of smectitic clays in this deposit is from 0.5 up to 4.0 m thick (mean 1.5 m). It is confined to the Tarusa horizon of the Serpukhovian stage (Lower Carboniferous). The mineral assemblages of bentonite are dominated by alkali-earth dioctahedral smectite (up to 80%), mixed-layer illite-smectite, illite, kaolinite, quartz and calcite [69]. The established bentonite resources of the *Pikalevskoe* deposit are about 4 million tonnes with the expected overall deposit expected to contain about 300 million tonnes. The bentonite in the *Pikalevskoe* deposit belongs to the alkali-earth type and can be utilized in ceramic and foundry industries, for the preparation of drilling fluids, and for iron-ore pelletizing. Given the very limited volume of material in many of the other known bentonite deposits listed in Table 5.1, the *Pikalevskoye* deposits are of particular interest as they contain sufficient material to supply repository requirements. As a result of these considerations, this material was selected for consideration as a candidate bentonite for assessment in this CRP.

Pkialevskoe

Bentonite

Deposit

FIG. 5.5. Location of bentonite deposits in Russian Federation.

TABLE 5.1. ESTIMATED RESERVES OF BENTONITE DEPOSITS IN RUSSIAN FEDERATION [68]

#	Deposit	Estimated Reserves, Millions of tonnes
1	Serpuhov Series	5
2	Mihaylovskoe	40
3	Novo-Pyatkovskoe	10
4	Sherbakovskoe	39
5	Nikol’skoe	1.7
6	Tarasovskoe	25
7	Millerovskoe	17
8	Nurlatskoe	8
9	Biklyanskoe	26
10	Ziryanskoe	28
11	Lyubinskoe	20
12	Chernogorskoe	9
13	Zerkalnoe	0.1
14	Urgalskoe	0.3
15	Nalchikskoe	5
16	Pikalevskoe	300

5.1.6. South Africa

South Africa’s total reserves of economically viable bentonite (formed by the alteration of volcanic material) is 2.4 Mt (Table 5.2) with relatively pure bentonite deposits at Koppies (Free State Province), Heidelberg, Knysna and Woodhouse districts (Cape Province) and an impure deposit at Mbuze (KwaZulu-Natal Province) [70] as indicated by the circled regions in Fig. 5.6.

TABLE 5.2. ECONOMICALLY VIABLE BENTONITE CLAY RESOURCES IN SOUTH AFRICA

Region	District	Geological origin	Estimated reserves (Mt)
Free State	Koppies	*In situ* weathering of volcanic ash	1.7
Southern Cape	Knysna	Weathering of volcanic ash	
	Heidelberg	Weathering of volcanic ash	> 0.7
Eastern Cape	Woodhouse	Alteration of volcanic ash deposited in water	1
Northern Natal	Mbuze	Alteration of perlite, perlitic pillow lava and tuff	large

FIG. 5.6. Bentonite deposits in South Africa.

The bentonite in the Koppies district of the Free State is flat-lying and occurs underneath Ecca shale where it is imbedded in the Swaziland schist. The origin of this deposit is not clear and it has been suggested that the material is related to the weathering of the schist, but samples from one of the deposits clearly indicated that the bentonite was derived from in situ alteration of volcanic ash. The main exchangeable cations are: Ca^{2+} and Mg^{2+} .

The deposit at Heidelberg occurs in the cretaceous beds in the Southern Cape south of the Langeberg mountains (about 250 km from Cape Town) and originates from volcanic ash deposited during the formation of the Langeberg mountains. The clay occurs in large beds dipping in a northern direction towards the mountains. The clay layer is covered by a soft siliceous sandstone layer, consisting mainly of cristobalite and quartz. The bentonites in this area can be described as sodium/magnesium bentonites because unlike calcium bentonites they form a viscous gel in the natural state without the necessity of peptization with sodium carbonate.

The Knysna deposit is located 5 km west of Plettenberg Bay and is part of a succession of clay and sand layers. The clay bodies dip in an easterly direction. The bentonite appears to be similar to that of the Koppies deposit and the main exchangeable cations are Ca^{2+} and Mg^{2+}.

The deposit in the Woodhouse district is located 19 km south of Jamestown. It is underlain by yellowish sandstone and overlain by red mudstone. The mudstone has been altered into a hard and cherty rock that is very resistant to weathering. The exchangeable cations are Ca^{2+} and Mg^{2+}. This deposit seems to vary in quality and appearance and originated via alteration of volcanic ash deposited in water. This reserve is only marginally economic because of the variable quality of the deposit and its location on the steep flanks of a mountain.

An impure deposit of bentonite is found near Mkuze in the Ubombo district in the KwaZulu-Natal province. Little is known about the deposit but it appears to have been formed by the alteration of perlite, peritic pillow lava and tuff.

South Africa has only a small domestic bentonite industry with bentonite from the Koppies and Heidelberg regions currently being used for sealing of dams and other minor applications. These resources were not previously explored with the intention of being used for sealing materials in a radioactive waste repository. In order to study the properties of these bentonites and their potential for use as components of the engineered barrier systems (EBS) in a HLW waste repository, *Eccabond* (from Koppies region) and *Envirobent* (from Heidelberg region) were selected as candidate materials for evaluation in this CRP.

5.1.7. Ukraine

Study of deposits of bentonite clays on the territory of Ukraine has shown that over 100 bentonite clay occurrences of different generic types, mostly with alkaline-earth adsorbed cation complexes [71, 72] have been found in Ukraine. These bentonite clays can be classified into the following four genetic types: dominated by Tufogene-sedimentary (62.9%), sedimentary (30.3%), with less widespread occurrences of post-volcanic (4.5%) and alluvial (2.3%) deposits.

Prychornomorska depression

These clays are characterised by good adsorption properties and occur in Sarmatian sedimentary deposits in the Odesa region. Detailed study of these clays revealed their polymineral character. The montmorillonite content in these deposits is relatively low with hydromica and kaolinite dominating the mineralogy.

Mountain Crimea and epihercynic platform of the flat Crimea

Postvolcanic bentonite clays are found in volcanic tuffs of the Karadag mountains. These bentonites are composed of alkaline (sodium) montmorillonite. The total amount of the adsorbed cations in the clayey rock is 106 milli-equivalents (meq) per 100 g of dry clay. Probable reserves are 2–3 million tonnes; however developing of this deposit is impossible since it is located within a national park.

Donbas deposit

Grygorivsky deposit bentonites are confined to Bilokuzminivska suite of the Early Triassic. The commercial mineral reserve is 260 million tonnes. Grygorivsky deposit is an extended multi-strata flat deposit of alkaline-earth bentonites divided into sand and sandy-carbonate clay strata. Nine 0.5–0.7 m thick strata are of practical importance. Basing on the laboratory-technologic studies, Grygorivsky bentonite can be used in industry in natural and activated state.

Central and near Dnieper areas of the Ukrainian shield

In this region the Cherkassy sedimentary clay deposit was examined. There are montmorillonite-rich horizons, which were generated by weathering of gabbro-anorthosites, amphibolites and gneisses. The 100–120 m thick well-developed area of crustal weathering includes a 15–40 m thick montmorillonite-rich zone in some places of the lower part of the deposit. Montmorillonite-nontronite clays are also found in Mesozoic crustal weathering in the Volyn region. Their thickness is up to 10 m. Clays are characterized by high content of exchange cations: up to 124.5 mg-equivalent per 100 g of dry clay.

The *Cherkassy* deposit of bentonite and palygorskite clays is one of the largest in Ukraine located in the centre of the Ukrainian shield and is connected with continguous deposits of the Early and Middle Miocene [73–75]. The area of the layer is 195 km^2, the total reserve is 825 million tonnes and in Dashukovsky site, it is 5.3 million tonnes. The productive strata of Cherkassy bentonite and palygorskite clay deposit consists of 5 layers distinguished by macroscopic features (top-down). The clays are commercially available in crushed and dried form from Dashukovskiy plant, which produce bentonite powder on the market. In some cases additives such as soda ash are added to bentonite to induce some desired changes in the swelling or rheological properties of the bentonite. This material was forward as candidate material for studying in this CRP.

FIG. 5.7. Main deposits of bentonite clays on the territory of Ukraine.

5.1.8. Germany (Friedland clay)

Although not a participant in this CRP, Germany is an important source of commercial smectite-rich material. A very homogeneous clay body of Tertiary age with a volume of at least 200 million cubic meters is located in the Friedland area close to Neubrandenburg in north-eastern Germany (Fig. 5.8) [51]. The clay is locally named *Friedland* Ton and provides an example of a commercially available, relatively smectite-poor European clay that may be appropriate for use in repository sealing. This clay is commercially produced by the company DURTEC based in Neubrandenburg, Germany. This smectitic clay is presently used for isolation of shallow domestic waste landfills and is presently being considered as a possible material for backfilling of drifts and shafts in repositories for radioactive and hazardous chemical waste. This clay, which is of Tertiary age, has an average content of expandable minerals of 50–60%, (montmorillonite in Na-form and mixed-layer mica/montmorillonite), and 24% quartz, 5% feldspars, 13% mica, 11% chlorite, and 2% carbonates with very little sulphur and potassium content [51].

As Friedland clay is being considered by some repository programmes as a potential component of the EBS it was decided to treat this material as candidate material and compare it to the reference and sample clays used in this CRP.

FIG. 5.8. Friedland bentonite clay deposit near Neubrandenburg in Germany.

5.2. Reference materials used by CRP

An important aspect of this CRP is the comparison of relatively unknown swelling materials to more fully characterized swelling material proposed for HLW repository use. This comparison provides a basis for assessing the likely suitability of these lesser-known materials as candidate components in a repository sealing system. Extensively characterized materials (Wyoming (MX 80), Kunigel-V1 and Saskatchewan) were supplied to the Member States participating in this CRP to be used as reference materials for the purpose of the CRP. As these materials are already part of well-established and extensively documented national programmes, the screening methodology will increases the confidence of the member states considering their use of their own swelling material at waste disposal sites. A short description of the reference clays used in this CRP is provided in Sections 5.2.1 through 5.2.3.

5.2.1. Clay material from Wyoming (MX 80)

MX-80 is a specific bentonite clay product mined and processed from the Clay Spur bed, Wyoming, USA [76]. It is commonly referred to as having 65 to 75 wt.% Na-rich montmorillonite and is a commonly referenced candidate material for use in engineered barriers within underground nuclear waste disposal sites in Switzerland and Sweden. A variety of other similar bentonite products are available from Wyoming-type sources of clay originating from the same region of the USA [77, 78].

Mining of bentonite started in the area along the borders of Wyoming (WY), Montana (MT), and South Dakota (SD) (Fig. 5.9), USA., in 1888 and in 1898 "bentonite" was assigned as a mineral name after the Ft. Benton formation[2]. Production of bentonite continues in these three states and elsewhere in the United States of America but materials from the WY, MT and SD regions are the major sources of bentonite for North America [77, 78]. Currently, bentonite deposits in Wyoming make up 70% of the world's proven supply. Being of very high quality it is economical to mine bentonite as deep as 50 feet in these deposits.

There are three major economic geologic areas of production of swelling bentonite in Wyoming:

- The Clay Spur areas around Upton and Newcastle;
- The Colony Area in the north east corner of Wyoming;
- Along the east and west flanks of the Big Horn Range.

Wyoming (MX 80) bentonite clay is derived from volcanic ash deposited in a shallow marine environment. It contains 65–75% of the phyllosilicate montmorillonite, 10–14% quartz, 5–9% feldspars, 2–4% mica, 3–5% carbonates and chlorite and 1–3% heavy metals. In addition to smectite, bentonite contains quartz, chalcedony, feldspar, plagioclase, calcite, dolomite, analcite, pyrite, etc. as accessory minerals. The chemical composition of bentonite is SiO_2 61-65%, Al_2O_3 22–25%, Fe_2O_3 1–7%, MgO 1–2%, CaO 0-0.6%, Na_2O 0-1% and K_2O 0–3% but is often chemically treated to obtain desirable properties for special applications [78]. Being a sodium-dominated montmorillonite it has very high free-swell capacity and exhibits other very desirable physical and chemical characteristics for application in a nuclear waste repository. The basic mineralogical, chemical and geotechnical properties of Wyoming bentonite are presented in Section 5.3.

FIG. 5.9. Wyoming bentonite deposit near Wyoming in United States of America [79].

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[2] See US Patent Number 5.292,908, Modified bentonite, Kondo, M., 1994.

5.2.2. *Bentonite from Saskatchewan from Canada*

Canada has considerable quantities of bentonite within its borders, particularly in the Prairie Provinces (Manitoba, Saskatchewan and Alberta) (Fig. 5.10) [77]. These bentonites, which are similar in origin and structure to those in Wyoming and South Dakota, are of Cretaceous and lower Tertiary ages. They occur in marine shales and limestones and their thickness ranges between a centimetre and a few metres. Alteration of volcanic ash is the generally accepted mode of formation. The bentonites found in the Canadian prairie region range from calcium-dominated through mixed Na-Ca and Ca-Na to sodium-bentonites depending on locality and geologic horizon. The region between Avonlea Creek and Moose Jaw Creek south of Wilcox Saskatchewan contains large reserves of swelling bentonite (Na-Ca-type) in the marine Bearpaw Formation and is shown in the circled region of Fig. 5.10. Saskatchewan's only operating bentonite quarry is operated by Canadian Clay Products Inc. near Truax. The processing plant, located at Wilcox, has an annual production capacity of about 80 000 tonnes per year. Historically other mining operations have been active in the provinces of Alberta and Manitoba but these operations have ceased. The basic geotechnical properties of Saskatchewan clay are presented in Section 5.3.

Being a domestic resource Atomic Energy of Canada undertook an extensive physical, chemical, mineralogical and geotechnical characterization programme on the *Saskatchewan* materials and other potential North American supplies of bentonite between 1980 and 1994 [77, 78, 80] and qualified a number of them as being potentially suitable for repository applications.

FIG. 5.10. Saskatchewan clay deposit in Canada.

5.2.3. *Kunigel-V1 bentonite from Japan*

Bentonites formed in Japan from volcanic ash and by in situ alteration through hydrothermal processes [81]. Tertiary liparites and liparitic tuffs are present on several sites in Honshu and Hokkaido. At Yamagata and in the Tsukinuno area on the main island, the deposits were formed in Early Miocene mostly by hydrothermal processes that gave large clay bodies that are irregular in shape (Fig. 5.11). Depending on the composition of the parent rock, zeolites have also been formed in large amounts. In the Gumma Prefecture of the Kwanto district, there are several beds with a total thickness of up to 5 m, interbedded with mudstones and hence of heterogeneous and relatively poor quality.

Bentonite deposits in Japan are mostly embedded in the Neogene sedimentary and pyroclatic rocks. Parent rocks of bentonite are mostly silicic vitric tuff, and bentonitization of the tuff generally occurred due to diagenesis or low-temperature hydrothermal alteration. Types of clay deposits that are mined are: 48% sedimentary clay (e.g. kaolin deposits in Neogene sediments), 38% hydrothermal clay (e.g. pyrophyllite, kaolin, and sericite deposits with igneous rocks) and 14% clays formed by mainly diagenesis (bentonite and fuller’s earth).

FIG. 5.11. Kunigel clay mined at Tsukinuno mine near Yamagata.

The bentonite deposits can be divided into Na or Na-Ca bentonite deposits and Ca bentonite deposits based on the composition of their interlayer exchangeable cations. The Na or Na-Ca bentonite deposits generally occur as large- to medium- scale stratified ore bodies and are mostly distributed in the Yamagata-Miyagi and Gunma-Niigata regions. A representative Na bentonite deposit from this region is the Tsukinuno deposit. This deposit consists of large-scale stratified ore bodies and the Tsukinuno bentonite is composed of Na smectite with subordinate quartz, feldspars, illite, calcite, and zeolite. Calcite is commonly concentrated into elliptical nodules up to several tens of centimetres. Parent rocks of the bentonite are felsic tuff beds intercalated in Middle Miocene hard shale.

Kunigel is produced at the Tsukinuno Mine in Yamagata in Japan. This is sodium-type bentonite containing nearly 48% montmorillonite, and is frequently used in the studies on the material for artificial barriers against nuclear waste in Japan Kunigel-V1 is a crude bentonite, and chalcedony, quartz, plagioclase, calcite, dolomite, analcite and pyrite are contained as impurities. The basic geotechnical properties of Kunigel-V1 bentonite are presented in Section 5.3.

5.3. Results of preliminary screening of test case materials

5.3.1. Clay mineralogy estimation of test case materials

5.3.1.1. X ray diffraction analyses

As discussed in Section 3.1, the use of X r ay diffraction analysis to determine the mineralogical composition of clay materials is perhaps the most definitive analytical tool available. It will identify the specific minerals present and an estimate of the relative proportions can be made by comparing the traces obtained using untreated material with those for material that have undergone pre-treatment. An example of the type of change observed for bentonites having undergone pre-treatment (glycolation) is provided in Fig. 5.12. It should be remembered however than X rays do not provide a measure of exchangeable cation compositions or non-crystalline minerals present, both of which can have a major influence on determining the suitability of the clay. Conducting these tests also requires the use of a mineralogy laboratory equipped with an x ray diffractometer, an operator experienced with its operation and how to pre-treat and prepare the specimens for analysis. An experienced clay mineralogist can further assess x ray patterns to identify mixed layer systems and slight variation in mineral structure as well as provide a semi-quantitative determination of relative mineral abundances.

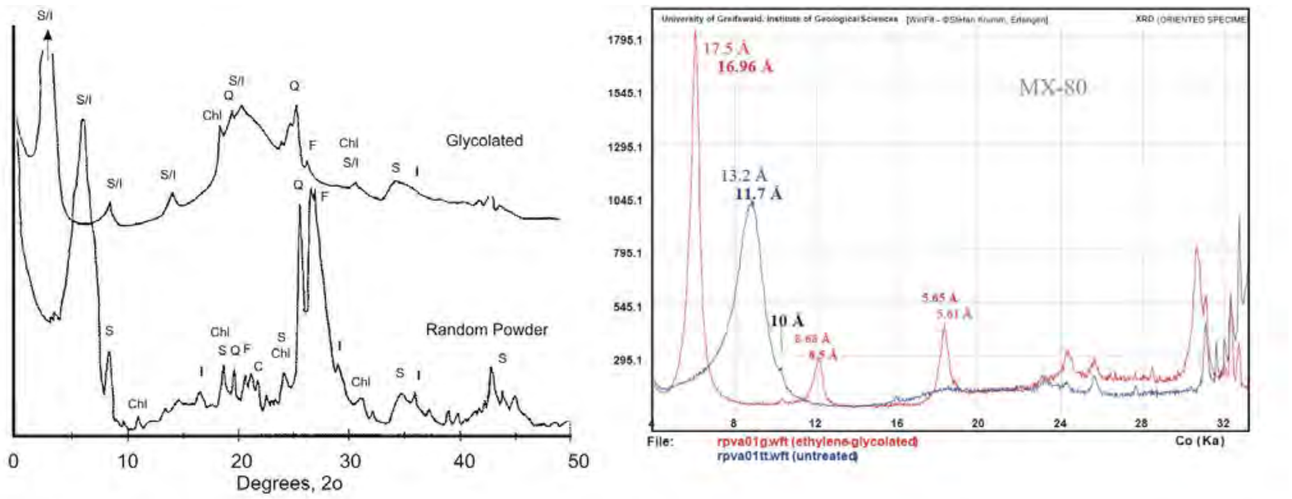


FIG. 5.12. Examples of change in X ray diffraction patterns resulting from pre-treatment (left: Saskatchewan Bentonite [82]; Right: Wyoming MX-80 bentonite [51]).

The results of X ray diffraction patterns collected for several of the materials examined in the course of this CRP have been compiled and are shown in Fig. 5.13. Included in the traces in Fig. 5.13 is a material of previously unknown mineralogical composition originating from South Africa. This material provided a very different trace from the other materials examined, lacking the characteristic smectite peaks (e.g. ~60 and ~180 in Fig. 5.13), found in swelling clays. Therefore based solely on an X ray trace a material of previously unknown composition could be eliminated as a potential source of clay for use as a buffer in a repository. All of the remaining traces indicate a high proportion of montmorillonite is present with quartz being the most common accessory mineral. These data, combined with results of analyses previously conducted are tabulated in Table 5.3, which provide a means of comparing their mineralogical composition.

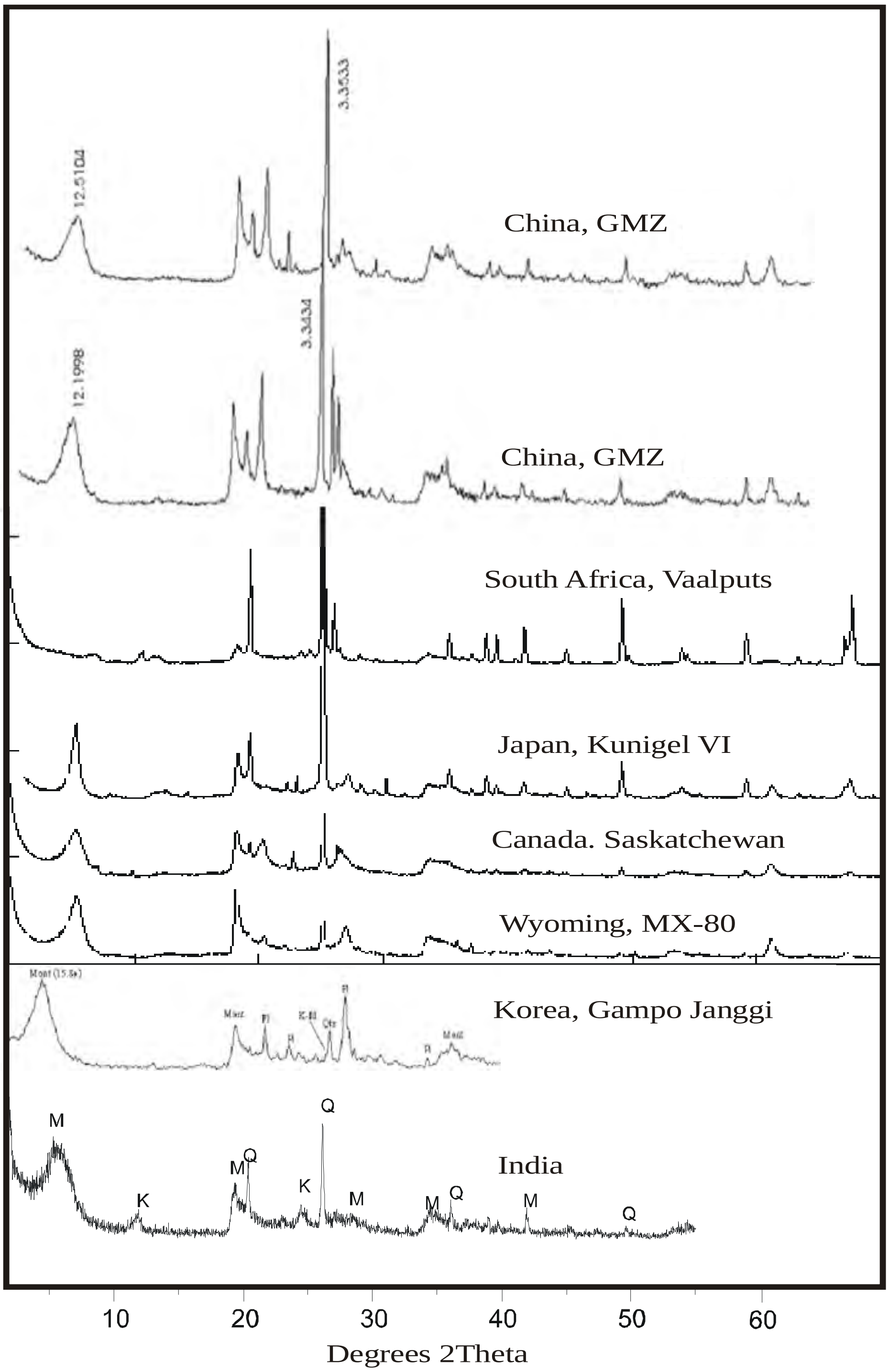


FIG. 5.13. X ray diffraction patterns for some of the materials examined by CRP.

TABLE 5.3. SUMMARY OF MINERALOGICAL ANALYSES FOR CANDIDATE CLAY SAMPLES AND ESTIMATED WEIGHT PERCENTAGE OF MINERALS PRESENT

Country, Material	Montmorillonite	Kaolinite	Illite	Mica and hydromica	Quartz	Feldspar	Pyrite
Canada, Saskatchewan [82]	~80	Nil to <5	Minor phase <10	Trace - Minor phase	Minor phase, 5-10	Minor phase <5	Nil
China, Gaomiaozi	75-90	No data	No data	No data	8-15	Minor phase <5	No data
Czech Republic, Rokle	90	No data	No data	<1	2	No data	No data
Germany, Friedland Clay [83, 84]	45 30	No data present	No data present	13 present	24 present	5 present	- trace
India	Main phase	Minor phase	-	-	Minor phase	-	-
Japan, Kunigel-V1	46-49	-	-	-	29-38	3-6	1
Republic of Korea, Janggi Series	50-90	-	-	Minor phase	Minor phase	Minor phase	-
Russian Federation, Leningrad region, Picalevskoe	80	-	Minor phase	-	Minor phase	Minor phase	-
South Africa, Eccabond N	94	-	-	-	5.5	0.5	
South Africa, Envirobent 0104	85	15	-	Minor phase	Minor phase	-	-
Ukraine, Cherkassy	90-95	-	-	-	3-5	Minor phase	Impurity?
USA, MX-80 [82]	75	-	Incl. In Mica	<3	10-15	3-10	Impurity?
USA, Wyoming-type [82]	88-92	-	-	Trace	4-16	2.3-4	-

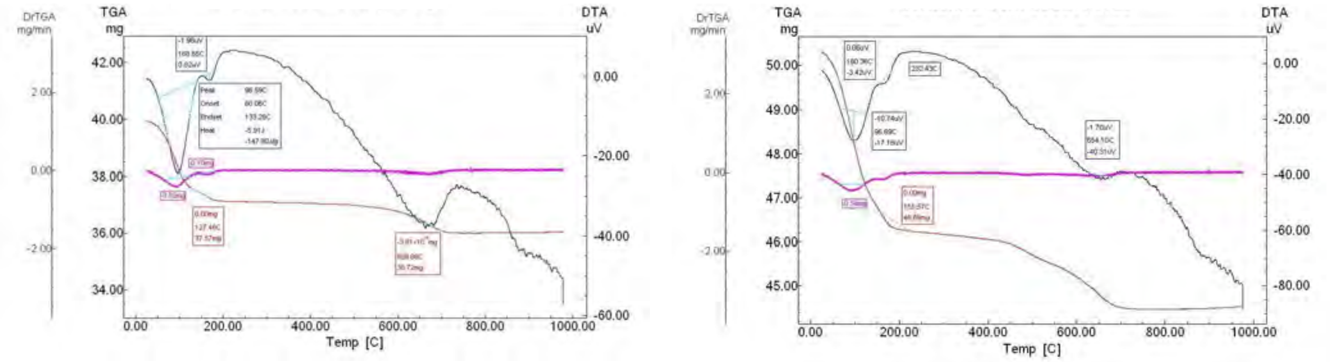
* Calcite and/or gypsum also present in small quantities

Most of the materials examined by the CRP contain a very high swelling clay component (>80 % present in most of the materials examined). Only the materials from Germany (Friedland), Japan (Kunigel-V1) and the Republic of Korea (Janggi Series) show montmorillonite contents that range substantially lower than this. It should be noted that the measurements of montmorillonite content presented in Table V-3 show the range in the quantity of smectite present, often of more than 10% variation in individual component presence is reported. This type of variability (and often higher) is commonly encountered within bentonite deposits and is a feature of the natural and sedimentary nature of most bentonites. The source material from which the bentonite was formed will vary with distance and depth and as a result the bentonite will exhibit similar variability. Another source of variation is localized geochemical influences (e.g. presence of K) that may have resulted in alteration of the mineralogy within the deposit. The presence of potentially substantial variations in the composition of the bentonite in a given deposit has an influence in the subsequent long term use of the material being excavated from a single locality. Constant quality checking is necessary for any commercial production of bentonite material, and commercial producers of bentonite currently undertake such checks as part of routine quality control, although XRD testing is not usually part of this process.

5.3.1.2. DTGS assessment

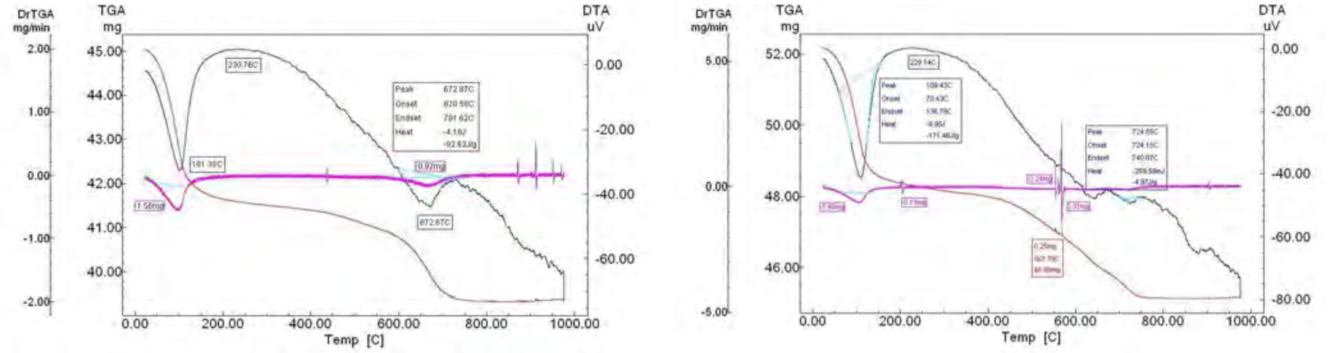
Differential Thermal Gravimetric analysis provides another means of identifying what minerals may be present within a specimen and also if analysed by someone familiar with the technique it can be used to estimate the relative mineral abundances. It is however only at best only semi-quantitative but provides a means of confirming the results of other mineralogical assessment techniques such as X ray diffraction. The results of the DTGS analyses presented in Fig. 5.14 provide indications of the relative abundance of the minerals present in each of the clay samples examined. They do not however provide a quantitative measure of proportion an in some cases the results of the DTGS and X ray diffraction analyses are difficult to rationalize. They require an experienced clay mineralogist who is familiar with layer silicates in order to evaluate the details of sample composition, especially where mixed layer minerals are present.

The Centre for Experimental Geotechnics (CEG) of the Czech Technical University in Prague provided the DTGS spectra for several of the raw clay and bentonite materials being examined as potential candidate materials for use in sealing applications. The DTA curves and the TGA curves produced at the CEG for the Gaomiaozi, Gampo, Eccabond N, Envirobent 0104, Cherkasy, RMN and Picalevskoe materials described in Section 5.2 are shown in Figs 5.14(a)-(g), as well as in Fig. 5.14(h) the trace for Kungel V1 bentonite provided at that time by JNC (now JAEA).



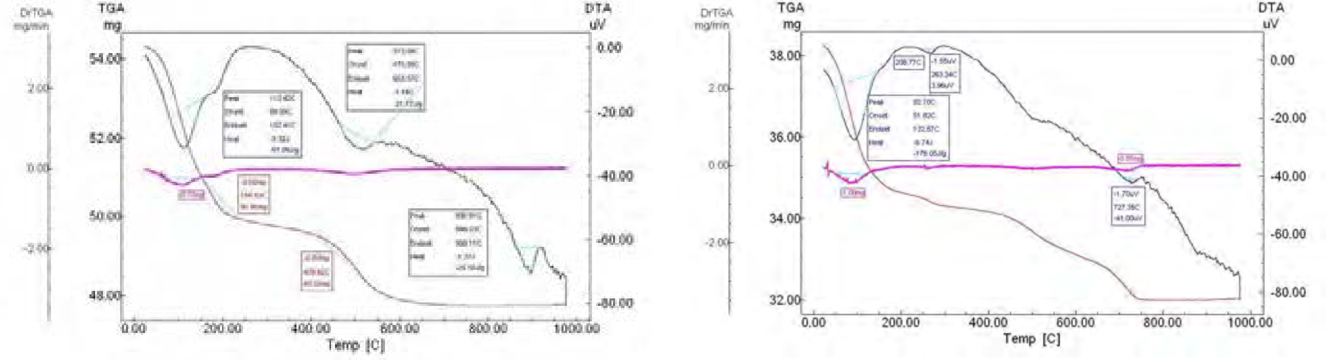
(a) GMZ01 (China)

(b) Gampo (Republic of Korea)



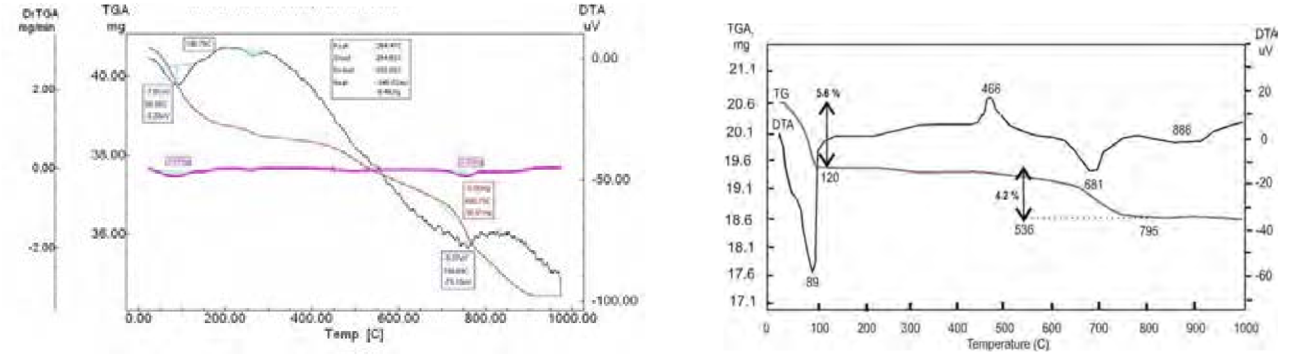
(c) Eccabond N (South Africa)

(d) Envirobent 0104 (South Africa)



(e) Cherkassy (Ukraine)

(f) RMN (Czech Republic)



(g) Picalevskoe (Russian Federation)

(h) Kunigel V1 (Japan)

FIG. 5.14. DGTS spectra of clays.

The curves provided as Fig. 5.14 show that the materials all have a low endothermic peak temperature and dehydroxylation peak temperatures at approximately 80–110ºC and 600–700ºC respectively in the DTA curve. The locations of these peaks with regard to temperature vary slightly as the result of factors such as mineralogical composition and the degree of clay mineral inter-layering. The magnitudes of these peaks also provide an indication of the proportion of layer silicates (clays) present in the specimen as non-clay minerals have exotherms different than those of clays.

The TGA curves for these materials indicate that rapid initial loss of mass (water) continues to about 190ºC with plateau region following up to about 600ºC, which reflects the loss of cation coordinated and hydroxyl water, typically associated with swelling clays. These DTA and TGA curves are consistent with those previously observed for MX-80 (Wyoming) and Saskatchewan bentonite [85]. The presence of minor bumps in the curves at temperatures not close to the 190ºC and 600ºC water losses (for example in Fig. 5.14 (traces f and g) where ongoing loss of mass occurs over the temperature range of 200-600ºC), can be attributed to other water-containing minerals and most likely mixed layer swelling and non-swelling clays (e.g. illite-smectite, chlorite, vermiculite-chlorite). Similarly the lack of a strongly defined mass plateau in the 200-600ºC range is an indication of a substantial quantity of mixed layer clay that gradually loses its structural water. These minor components may be significant with respect to the suitability of these materials for use in repository applications and provide an indication that more detailed assessment is needed before they can be accepted for use.

5.3.1.3. Exchangeable cations and cation exchange capacity

As discussed previously, two of the key parameters that affects the swelling, hydraulic and ion-exchange properties of a clay material are its ion-exchange capacity and the composition of these ions. Although not an absolute measure of the mineralogical composition or purity, swelling clay materials characteristically have high cation exchange capacities. It should be noted that the measure of this property can also be affected by minor constituents such as organics (either naturally present, or added to improve swelling or rheological properties of clay). Associated with cation exchange capacity is a measure of what cations are naturally present in the ion exchange sites. Measured values for the clays examined by this CRP are provided in Table 5.4.

Strongly charged ions such as Ca, Mg, Fe in the exchange sites will result in a material that has lower free-swell capacity and a tendency not to as readily exchange cations from its surface. This reduced exchangeability may have an influence in the ability of the bentonite to sorb contaminants and such material would certainly have a reduced ability to generate a swelling pressure on its confinement, especially at lower densities. With a lower swelling pressure, higher hydraulic conductivity would be expected to be higher for materials of similar density that have mono-valent cations associated with its exchange surfaces. It should be noted however that systems having divalent cations will be less affected by changes in groundwater chemistry than systems initially dominated by monovalent cations such as Na and may be more resistant of removal of colloidal mass over the long term. Additionally, as noted previously, the presence of certain cations (e.g. K^+) may under elevated temperature conditions result in an accelerated rate of montmorillonite mineral conversion to an essentially non-swelling illitic clay over relatively short timescales (100s to 1000s of years depending on specific conditions). As the repository needs the clay to retain its swelling capacity over a longer timescale then care should be taken to ensure that the bentonite used is chemically compatible with the repository application.

TABLE 5.4. EXCHANGEABLE CATIONS AND CATION EXCHANGE CAPACITY

Country, Deposit	Exchangeable Cations (meq/100 g)				Cation Exchange Capacity (meq/100 g)	pH
	Na	Ca	Mg	K		
Canada, Saskatchewan	45	35	5	3	50–85	9.0
China, Gaomiaozi	32–58	14–30	6–12	3–6	71–78	8.7–9.2
Czech Republic, Rokle		46	47	12	62	
Germany, Friedland [84]	11 22.2	4 11	2 6.9	34 2.2	50–60 42.3	8.3
India	10–16	1–3		ND	50–65	7.0–8.7
Japan, Kunigel-V1	55–63	42–46	2–7	1–3	52	
Korea, Janggi Series	1–7	24–57	18–33	1–2	50–92	9.5
Russian Federation, Leningrad reg., Picalevskoe	0	32	10	3	40	
South Africa, Eccabond	46	29	12	1.3		10
South Africa, Envirobent 0104	32	23	12	2.1		10
Ukraine, Cherkassy	60–63	60–70	ND	1–2	72-93	
USA MX-80 [84]	56–71	30	15	2	71 84–104-	8.5–10
USA, Wyoming [78]	49	29	5	<1	55-90	8–10

In Table 5.4 it can be seen that the bentonite from the Czech Republic and Germany apparently have higher proportions of K in the exchange sites of the clay than the remainder of the materials examined. The Czech material is unique within this study in that it is a high-purity bentonite that is Ca-Mg dominated and so will not likely readily exchange these cations for K and so may be less susceptible to mineralogical alteration. The potassium results for the material from China are unexpected given that the material has a very low concentration of other exchangeable cations and apparently very little in the way of alteration products (illite) within the deposits (see Table 5.3, Fig. 5.13 and Fig. 5.14) and a low presence of K in its solid phase (see Section 5.4). The Friedland clay has a substantial K content and this is apparently variable within the deposit (as is smectite content), this can be attributed to its source (sedimentary – erosional) and relative youth. The Indian bentonite is also somewhat problematic with respect to exchange properties reported for it, the data does not provide sufficient information to assess its cation exchange behaviour or potential for mineralogical alteration. It will be necessary to do more examinations on these materials in order to develop a better understanding of their composition, together with what effects their exchangeable cations may have on their suitability for repository use.

5.3.2. Geotechnical characteristics

As was discussed in Sections 3 and 4 the basic geotechnical characterization of materials being screened as candidate swelling clay sources can provide valuable information on their potential suitability without needed to begin more expensive and time-consuming chemical and mineralogical assessments. Swelling clays are known to have extremely high Liquid Limits and plasticity indices and this is perhaps the quickest and easiest way to begin a screening process. If the sample does not have these basic characteristics it is unlikely that it will be appropriate for use as a repository sealing material. The second basic geotechnical screening test is particle-size analysis. Swelling clays are typically found as small particles that tend to be in the clay to fine silt size range (see Section 4.3.2). As a result if a quick particle-size analysis finds that most of the particles are in the coarse silt and larger range, it is unlikely that the material being examined will be suitable as a source for swelling clay.

As an exercise to evaluate a range of “candidate” clay materials for their potential usefulness as sources of swelling clay for sealing of a waste repository they were tested to determine their basic geotechnical properties. Table 5.5 presents the results of these screening tests undertaken by the participants of this project. From these data it is difficult to distinguish the material behaviour based on grain-size analysis, well characterized and high swelling capacity bentonites such as MX-80, Saskatchewan and Wyoming materials and lower swelling capacity Kunigel-V1 materials all show similar grain-size distributions. Comparing these materials with other candidate clays in Table 5.5 does not result in any clear differentiation between these materials and their subsequently determined swelling or other geotechnical properties based on particle size distribution. Care should therefore be used in relating grain size distribution to swelling or other geotechnical properties.

5.3.3. Free swell

Free swelling of bentonite materials is a quick and easy diagnostic feature. The montmorillonite-type clays have a very high swelling index. This mineral type will, in the presence of fresh water, take on large quantities of water and expand to many times its initial volume. The amount of swelling is predominantly dependant on the content of swelling clay, the type of cations present on its exchange surfaces and the chemical composition of the water used to do the swelling test. As a result it is necessary to look at the results of free swell tests in conjunction with some of the other basic screening tests before deciding on the potential suitability of a candidate material.

Table 5.5 contains the free-swell measurements made on a variety of materials, including those examined as part of this study. In all of the sodium-dominated “bentonite” materials a substantial free-swell capacity was observed (>8 cc/2 g of clay in fresh water). Materials having a lesser swelling clay content or whose exchange sites were dominated by divalent cations (e.g. Ca, Mg, Fe) rather than sodium exhibited a discernibly lower free swell capacity (e.g. Friedland, Cherkassy and Picalevskoe). The use of free swell measurements to assess a clay provides only a crude measure of the relative hydroscopic character of the clay and is strongly affected by the mineralogic and surface exchange characteristics of the material being tested. It does however provide a quick means of telling if the material has the potential to swell under fresh-water conditions and is worth further investigation.

Recognizing the above-listed limitations with respect to use of free-swell capacity as an indicator, it was determined that all of the materials examined in this CRP have the capacity to swell substantially when unconfined and exposed to freshwater.

TABLE 5.5. BASIC PROPERTIES OF CANDIDATE CLAYS

Sample Source & Name	Liquid Limit (%)	Plastic Limit (%)	Plasticity Index	Specific Density (kg/m^3)	Grain Size Distribution			Free Swell Index (cc/2 g)	Surface Area (m^2/g)
					Clay (%)	Silt (%)	Sand (%)		
Canada, Saskatchewan**	150–385	33–64	115–330	2700	19	81	<1	>9	500–650
China, GMZ-01	336	46	290	2771	55	41	4	13.4	
Czech Republic	132	49	83	2831	57	38	5	8.5	
Germany, Friedland	90–130	29–34	56–101	2710	70	20–25	3–4		150–200
India	~425	53–55	367	2040–2580	84–86	13–15	3–4	20-30	
Japan, Kunigel-V1**	>280	<50	>230	2700	13	83	4	7–16	
Republic of Korea, Gampo	162	39	123	2756	41	50	9	10.1	
South Africa, Eccabond	478	56	422	2694	49	45	6	28.1	
South Africa, Envirobent	553	51	502	2808	57	41	2	32.2	
Russian Federation, Picalevskoe	114	33	81	2690	21	73	6	5.0	
Ukraine, Cherkassy	125	45	80	2707	19	78	3	6.8	
USA, MX-80*	400	70	330	2700	12	84	4	12–26	562
USA, Wyoming-type**	>350	~40	>310		13–22	76–84	0-5	8–26	750–800

* This has been the reference material for a number of nuclear fuel waste disposal concepts (e.g. SKB, NAGRA, POSIVA).

** Commercial Bentonite Materials from a variety of sources [52, 77, 78]

5.4. Performance verification of test materials

5.4.1. Swelling pressure

Swelling pressure developed by a confined bentonite given access to free-water provides a key measure of the ability of the material to potentially act as an effective seal in a repository environment. As outlined in Section 4.6 and shown in Fig. 4.8, the absolute swelling pressure developed will depend on a variety of factors, perhaps the most important of which are EMDD, pore fluid chemistry and the nature of the cations in the exchange sites on the clay mineral surfaces. Fig. 5.15 shows swelling pressures developed by several of the reference and candidate clays examined by the CRP. To illustrate the importance of accurately determining the montmorillonite content of the candidate material when developing estimates of the swelling pressure it can develop, the Friedland clay data is plotted twice, once for the upper limit of smectite (45%) and again for the lower limit (30%) as noted in Table 5.3. The shift in the data is quite substantial and provides a possible explanation for the scatter present in the remainder of the data and literature data in general.

The data presented for generic Wyoming or Saskatchewan materials is actually for materials from different locations or obtained at different times from these deposits and so are subject to some degree of mineralogical variability. The result is a set of swelling pressure data that while producing a consistent EMDD-swelling pressure relationship will exhibit some degree of scatter as the result of minor compositional variability. This means that unless the mineralogy of each of the clays is very well known, there will exist a degree of uncertainty with respect to the swelling pressure a particular material can develop under a given geochemical environment. This also highlights the need to maintain quality control checks on materials ultimately selected for use in a repository so that performance is maintained within limits that are deemed acceptable by the repository designers and safety assessment specialists.

The data presented in Fig. 5.15 highlights the similarity in the swelling pressure behaviour of five of the materials considered in this CRP. Despite clear differences in their cation exchange and other geotechnical characteristics these materials behave in a similar manner, especially at EMDD values in excess of approximately 1000 kg/m^3. Although the data is limited, it would appear that the Czech and Friedland materials begin to exhibit slightly below average swelling pressures when their densities drop below approximately 1000 kg/m^3. This is not unexpected as lower density materials will be more strongly affected by their exchangeable cation composition and both of these materials are non-sodic smectites. At densities of interest for repository applications, the clay materials examined in this study can therefore all be hydraulically characterized in terms of the compacted density of their montmorillonite content and from this density specifications can be developed when repository design is undertaken. The data also shows that it is possible to use a normalizing parameter (EMDD) to describe or predict the swelling pressure behaviour of clays from very different sources.

It should be noted that data from candidate clays that matches the EMDD vs. swelling pressure characteristic curve shown in Fig. 5.15 does not mean that they are necessarily suitable for use as a buffer. This is especially true in materials having low swelling clay content since in order to meet the swelling pressure requirements of the SKB concept they would need to be very densely compacted (see Fig. 4.8). Depending on the clay considered such densification may not be practical. The decision as to the suitability of a candidate clay will be based on the specific geological, geochemical, geometry and other conditions relevant to each national programme. It may be that local clays will not meet the specifications for a buffer clay and that local clays that are of insufficient quality for use as a buffer may still be suitable for engineered barrier materials further from the waste canister (e.g. tunnel, room and shaft backfills). This is the type of approach currently being considered by Sweden, Finland where Friedland clay is being considered as a tunnel backfill and Canada where glacial lake clay is being considered as a component in tunnel backfill.

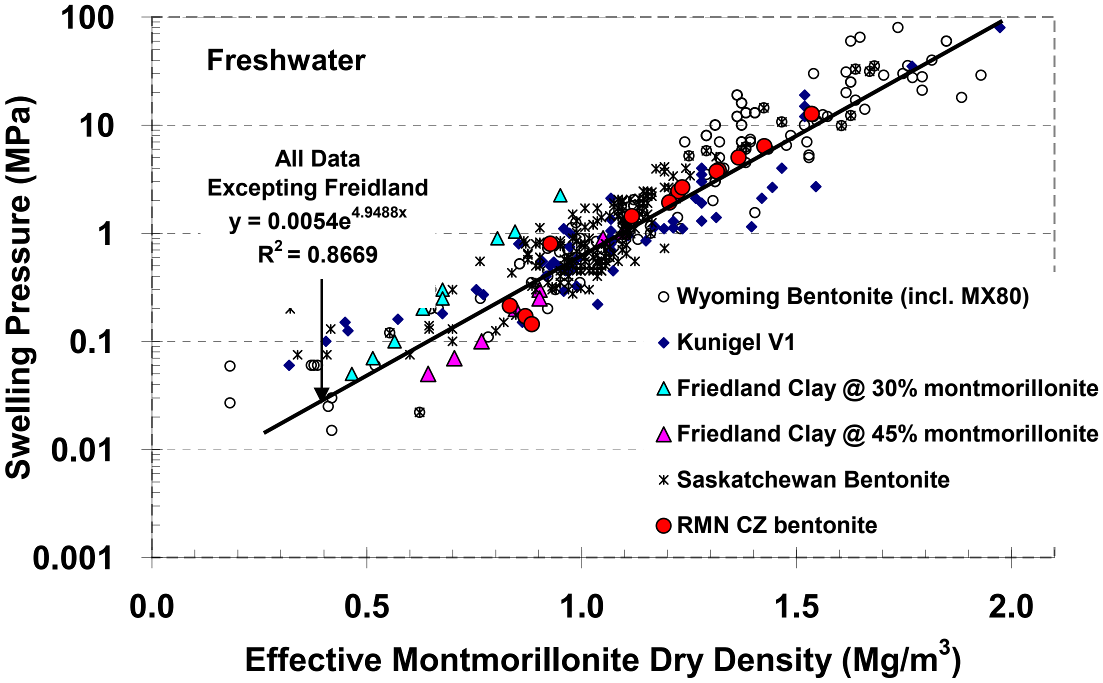


FIG. 5.15. Swelling pressure measured in smectite-rich clays examined in the CRP. (The importance of accurately knowing the smectite content of the clays is illustrated by the two plots for Friedland, EMDDs for 30% and 45% smectite are plotted.)

5.4.2. Hydraulic conductivity

As was discussed in Sections 3 and 4, it is known that the hydraulic conductivity of a clay-based swelling material will be dependent on a large number of factors, of which EMDD, pore fluid and exchangeable cation compositions will dominate. Testing of the hydraulic conductivity, like the measurement of swelling pressure is not a measure that would normally be included in initial screening of materials as part of routine quality checking of materials. Conduct of this type of test requires specialized equipment and is very time consuming so hydraulic characterization would normally be done once preliminary screening has been accomplished. Figure 5.16 shows a plot of hydraulic conductivity of several of the materials examined in this CRP as it relates to the normalizing EMDD parameter and despite their very different smectite contents the data for the Wyoming, Saskatchewan, Czech and Friedland clays can all be described using the EMDD relationship. Although data was not available for the other candidate materials, it might be expected that they would also fit the curve presented in Fig. 5.16.

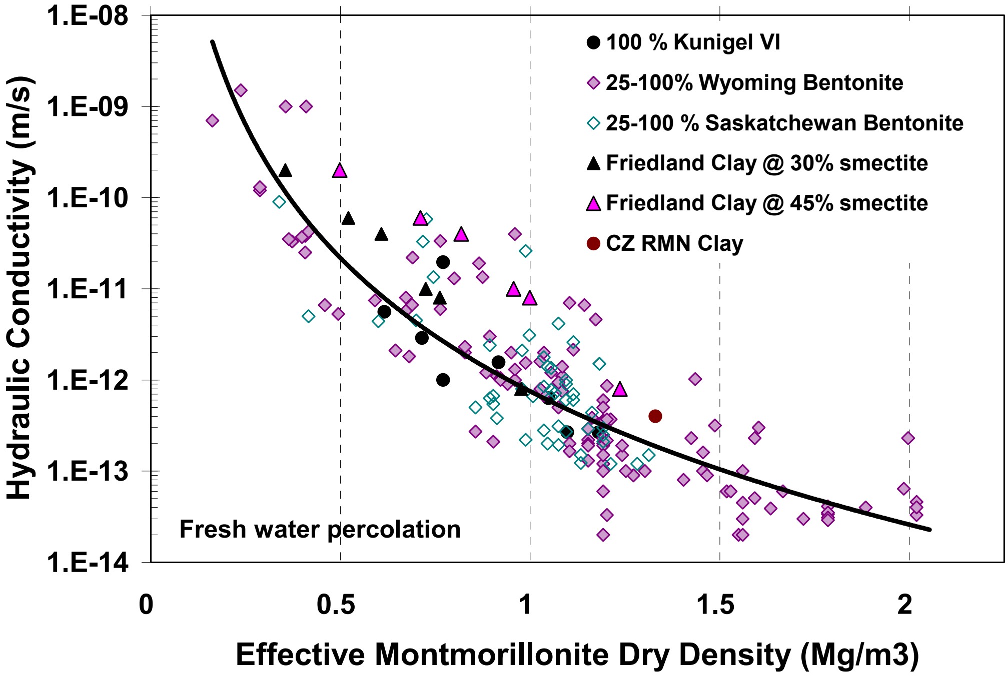


FIG. 5.16. Hydraulic conductivity measured for smectite-rich clays examined in the CRP.

As with Fig. 5.15, Fig. 5.16 includes a dual plot of the hydraulic conductivity versus EMDD plot where the extreme smectite content limits reported in literature are used to calculate EMDD for Friedland clay. The result of changing smectite content from 45% to 30% is a set of data that shifts from sensitive higher k for a given EMDD to one that falls within the scatter for other more extensively characterized materials. This again illustrates the need to accurately know the smectite content of candidate materials and the ability to predict hydraulic conductivity using EMDD normalization if mineralogical composition and dry density are known. It also highlights the effect of deposit variability on the achievable performance of swelling clays.

The same issues and limitations regarding the density needed to be achieved by low swelling capacity materials that were identified for swelling pressure exist for the hydraulic conductivity characteristics of swelling clays having relatively low smectite content. As a result careful consideration needs to be given regarding selection of materials for use as buffer.

5.4.3. Presence of potassium and iron

As discussed previously, potassium and iron are two elements that under certain thermal and chemical conditions, can adversely affect the longer-term performance of montmorillonitic clays by facilitating their conversion to illite or generation of cementitious materials that may prevent clay swelling. While the presence of these elements is expected in any natural material, excessive amounts may prove problematic depending on the repository concept considered. What is equally important to their quantity in the candidate material is their location. Potassium or iron in relatively insoluble crystalline materials or within existing illitic materials is not likely to be of the same significance as if they were present in amorphous or soluble mineral forms. It is therefore important to assess the measurements of total potassium and iron in context and look more carefully at other more complex evaluation tools if high concentrations are detected.

Table 5.6 presents the results of the chemical digestion results for the clay minerals examined in this study. Most of the materials examined in this study have chemical compositions that fall within a relatively consistent range. Notable exceptions to this are:

(1) The relatively higher SiO_2 values (can be in excess of 70%) for the materials from China, Japan and the Republic of Korea. The higher SiO_2 is likely due to the substantial quartz content known to exist in the materials from Japan and the Republic of Korea. The material from China also likely contains a higher than average quartz content.
(2) The higher Fe_2O_3 and higher than average CaO content for the Czech clay has been previously identified and is expected in an iron-rich smectite.
(3) The higher K_2O concentration in the solid component of the Russian and Friedland clays are from an unknown source and may be due to minor accessory minerals. These materials are both of erosional origin and so the presence of potassium-containing accessory minerals is not unexpected.

TABLE 5.6. CHEMICAL COMPOSITION OF REFERENCE AND CANDIDATE MATERIALS (IN WT.%)

Country, Deposit	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	FeO	MnO	CaO	MgO	K_2O	Na_2O	P_2O_5	H_2O*
Canada, Saskatchewan Bentonite [78]	59–65	0.1–1	16-20	3-5	No data	No data	1–2	1–2	0.1-2	1–2	<0.3	-
China, Gaomiaozi, clayey bentonite	63-72 69.2	0.0-0.1 0.13	1320 14.4	1–3 3.1	- 0.02	0-0.1 0.04	1-2 1.3	3-4 3.3	0-1 0.83	1-2 2.0	No data nil	- 5.4
Czech Republic, Rokle	54 53.7	No data 1.35	19 19.5	14 13.9	No data No data	No data No data	3 3.4	3 2.8	1 1.2	1 0.9	No data No data	5 3.2
Germany, Friedland [84]	57 60	No data 1	18 18	5.5 7	No data	No data	0 0.4	2 2	3.1 3	0.9 1	No data	- -
India	48–56	No data	15–20	3–6	No data	No data	0.1–0.4	3–4	-	1–2	No data	
Japan, Kunigel-V1	71	0.2	14	2	0.6	0.2	2	2	0.3	3	No data	4.7
Republic of Korea, Janggi Series	54–79 56.8	0.1–1 0.8	11–18 20	1–9 6	No data 0.2	0–0.2 No data	1–4 2.6	1–4 0.8	0.1–3 0.9	0.1–2 1.3	0.0-0.3 No data	- 10.6
Russian Federation Leningrad reg., Picalevskoe	60 64.8	1 1.14	17 15.1	6 5	No data 0.33	- 0.02	1 2.3	2 1.5	4 2.2	- 0.12	- 0.05	- 2.36
South Africa; Envirobent	58	0.3	18	6	-	-	2	3	0.6	0.2	0.7	-
South Africa; Eccabond	62	0.2	18	3.5	0.2	No data	0.6	3.9	0.7	2.4	No data	6.5
Ukraine; Cherkassy	52–54	0.2–1	15–18	4–5	0.3	0.01	2	2–3	0.1-1	0.2	0.1nt.	-
USA MX-80*	58–64	0.1–0.2	18–21	3	No data	No data	0.1-1	3	0.2–-0.4	2–3	No data	6
USA Wyoming+ [78]	56–67	0.1–1	16–21	2–4	No data	No data	0.4–4	1–3	0.1-1	2–3	No data	6–8

[*] Structural or strongly adsorbed water, either measured or inferred due to unaccounted for specimen mass. May also include some organic materials

[+] Compilation of various commercially available sodium bentonites from the Wyoming region of the USA.

These deviations from the “average” observed for the other bentonites are therefore largely explained by known mineralogical variations within these materials. These chemical analyses are consistent with and support the mineralogical and other materials properties assessments for these clays and serve to provide confidence in the mineralogical assessments produced.

5.5. Summary of screening of test case materials

5.5.1. General

The data related to the basic properties of the clays examined in the course of this CRP were evaluated and these evaluations were summarized in Table 5.7. These results indicate that based on current screening, most of the materials submitted for evaluation as part of the CRP can be considered for further evaluation either as buffer material for engineered barrier systems (EBS) in waste repositories or else as a component of backfilling materials used further away from the waste canisters. Further studies would need to include a repeat of the preliminary screening tests done in this CRP according to internationally acceptable methodologies, as well as further tests such as organic content, radiation effects, chemical stability, thermal conductivity, sorption properties, hydrothermal stability and others.

The mineralogical, chemical, physical, hydraulic and swelling characteristics of materials provided to this CRP were determined through a number of cooperative work packages involving and organized by the participants. This work allowed the technical expertise of several CRP participants to be utilized and provided an opportunity for those not familiar with these processes to gain an improved understanding of what would be needed to qualify/screen materials for use in a repository.

Measurements made on the raw clays provided by the CRP participants were compared to the very well-characterised reference bentonite clay MX-80 in order to provide a first screening of these materials. Using the well documented properties and apparently suitable characteristics of the MX-80 material as a baseline reference the remaining materials have been tabulated with respect to their comparability to this reference material. It should also be noted that the materials from Canada and Japan have also been extensively characterised and selected as candidate materials for these national programmes, however for the purposes of discussion, all materials were compared to MX-80 bentonite.

TABLE 5.7. CHARACTERISTICS OF CANDIDATE MATERIALS RELATIVE TO REFERENCE CLAY (MX-80)

Screening test	Canada Sask	China GMZ	Czech Republic Rokle	India	Japan Kunigel	Republic of Korea Gampo	South Africa Eccabond	South Africa Envirobent	Germany Friedland	Russian Federation Picalevskae	Ukraine Cherkassy
Physical properties^{++}											
Fines Content (% clay)	OK	>	>	>	OK	>>	>>	>>	>>	OK	OK
Plasticity Index	OK	OK	<<	OK	<	<	>	>	<<	<<	<<
Free swelling	<	OK	<	OK	<	<	>	>	<<	<<	<<
Swelling pressure, EMDD $^{+}$	OK	-	OK	-	OK	-	-	-	OK	-	-
K, EMDD^{+}	OK	-	OK	-	OK	-	-	-	OK	-	-
Mineralogy^{++}											
Swelling clay content	OK	OK	>	-	<	<	>	OK	<	OK	>
TG (smectite purity)	OK	OK	<	-	OK	<	OK	OK	<	<	<
XRD (% smectite)	OK?	OK	OK	OK	<	OK	-	-	<	<	-
Chemical Properties^{++}											
Exchangeable cations											
Na : Ca-Mg ratio^{x}	1	1-2	0	-	1	0	1	1	1	0	1
Potassium	OK	>	>>	-	OK	OK	OK	OK	>> , ?	OK	OK
CEC	OK	-	<	OK	<	< ?	-	-	<<	<<	OK
Chemical Composition^{++}											
Potassium	OK	OK	OK	OK	OK	> ?	OK	OK	>	>	OK
Iron	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK
Silica	OK	>	OK, <	<	OK, >	OK	OK	OK	OK	OK	OK

OK Material exhibits properties comparable to the reference MX-80 bentonite.

\> *Material exhibits content/concentrations that are higher than the reference MX-80 bentonite (this is not necessarily a positive characteristic)*

< *Material exhibits content/concentrations that are discernibly higher than reference MX-80 bentonite (this is not necessarily a negative feature)*

^{x} *MX-80 has an exchangeable Na : Ca-Mg ratio of 1.25–1.6.*

$^{+}$ *Swelling pressure and hydraulic properties demonstrated as being able to be described using EMDD parameter.*

$^{++}$ *See additional text for discussion of properties that are discernibly different than reference material.*

? *A range of measured values exist and so comparability to MX-80 is not clearly established.*

\- *Parameter not available or not measured in course of this study.*

The results provided in the summary of the individual materials examined are for raw, untreated clays rather than industrially modified products. As has been noted in this document and elsewhere, it is possible to improve the swelling and hydraulic properties of non-sodium bentonite materials through altering the exchangeable cation composition via chemical treatment. If this process does not adversely affect the other properties of the clay, then this is a means that might allow high montmorillonite clay content but low swelling capacity bentonites to be improved with regards to their swelling behaviour. Assessing this type of treatment on the behaviour of the candidate materials examined in this CRP would require a considerable effort and is beyond the scope of this project.

5.5.2. Summary of material characterizations

Canada: Saskatchewan bentonite

The screening tests (included physical characteristics, mineralogical composition, clay composition, TG analysis, X ray diffraction and swelling studies) done on the material from Canada indicates that this material is very similar in most respects to MX-80 (see Table 5.7). It exhibits a slightly lower CEC and free swell capacity, which can be attributed to a lower Na : Ca-Mg ratio on its exchange sites. It may also have slightly lower swelling clay content than the Wyoming material. In all other respects it is comparable and can be described hydraulically and in terms of swelling pressure using the normalizing EMDD parameter. Given its very similar nature to MX-80 it can be expected that it could be prepared to the type of dry densities specified for MX-80 buffer and would behave very similarly. The range in duplicated measurements done over many years on different batches indicates that quality control of the product may be somewhat problematic.

China: GMZ bentonite

The GMZ bentonite provided for evaluation by the CRP appears to in most respects, equal to or exceeds the quality of the MX-80 reference material (See Table 5.7). This material apparently has a higher Na: Ca-Mg ratio than the reference MX-80 clay. It also represents a very large deposit of bentonite that would certainly be large enough for a long term repository sealing programme. A more thorough evaluation in order to confirm these properties and its swelling and sealing characteristics would be useful.

Czech Republic: Rokle bentonite

The Rokle bentonite was recognized before the start of the CRP as being a material that will have different properties and behaviour than the sodium bentonites typically evaluated as buffer material. This is primarily associated with the Ca-Mg domination of the exchange sites of the clay and the elevated presence of interlayered smectite-illite clays relative to the reference MX-80 bentonite. The results of comparisons are provided in Table 5.7 and show that this is a material that has a lower swelling capacity and lower plasticity index than MX-80, potentially making its ability to self-seal problematic. This is balanced to some degree by its comparable swelling pressure and hydraulic properties at elevated EMDD. Given its high swelling clay content the dry density required to achieve substantial swelling pressure and low hydraulic conductivity is also quite comparable to the reference clay. Also of concern is the apparently elevated potassium content in the cation exchange sites, which may pose a risk for more rapid illitization.

India

The information regarding the candidate bentonite provided to the CRP by India was incomplete with regards to its geotechnical and surface chemistry and in some cases the results provided are conflicting (e.g. low exchangeable cation content but high CEC). The results of this clay's properties measurements relative to MX-80 are provided in Table 5.7. As a result of missing properties information it is difficult to provide a definitive assessment of this material's potential suitability for repository use. From the limited data available it would appear that this material has a high swelling clay content and therefore its exchange sites are likely sodium-dominated. However, before any conclusions can be drawn regarding its potential for use in a repository sealing application, much more characterisation will need to be done.

Japan: Kunigel-V1

Kunigel-V1 is a well-characterised bentonite product having been extensively studied in Japan for its potential as a sealing material in the Japanese repository concept. It was recognized before being evaluated as part of the CRP as being a material that has a lower swelling clay content than the MX-80 reference material. It is a material whose swelling and hydraulic properties can be described quite accurately using the EMDD concept and has been successfully used in large-scale sealing experiments, providing confidence in its potential for repository use. The Kunigel-V1 material has its cation exchange properties dominated by sodium and calcium with very little potassium, which should make it more resistant to illitization.

Republic of Korea: Janggi bentonite

This bentonite has been examined for many years as a potential material for repository sealing applications in the Republic of Korea. Like the Czech bentonite, it has a lower swelling clay content and likely more mixed smectite-illite minerals than MX-80. Also, like the Czech material, it has a lower ion-exchange capacity that is dominated by Ca and Mg, but — unlike the Czech material, it does not have an elevated potassium content on the exchange sites. This will give this clay a lower swelling potential than the MX-80 reference clay but potentially more mineralogically stable due to the lack of potassium in its exchange sites.

South Africa: Eccabond and envirobent

South Africa submitted two commercial bentonite materials for consideration by the CRP, eccabond and envirobent. These materials both have very high swelling clay content and analyses showed that these materials were of comparable quality to the MX-80 reference material (eccabond of slightly higher smectite content and purity than envirobent). These clays have their cation exchange sites dominated by Na and Ca with only small quantities of Mg and trace potassium. It could therefore be expected that these materials may perform comparably to the MX-80 bentonite although more extensive characterisation will be required before a decision can be made regarding their suitability.

Russian Federation: Picalevskae

The material provided to the CRP for evaluation shows a mineralogical character that is similar to that of the Czech bentonite, but apparently at a lower swelling clay content. Its geotechnical properties are summarized in Table 5.7 (low plasiticity, low swelling capacity…) most closely match those of the Cherkassy material from the Ukraine. Its clay mineralogy seems to be dominated by mixed layer clays resulting is low CEC and the exchange sites are occupied by Ca-Mg resulting in a lower swelling capacity. Comparison of this clay to MX-80 shows that it could not be expected to achieve the swelling, hydraulic or other properties that have been exhibited for the reference material. It does have some capacity to swell and might be suitable for use as a sealing material for use in backfilling, much as the Friedland material is.

Sweden: MX-80 and Friedland clay

Based on more than 30 years of ongoing research, SKB is proposing the use of MX-80 (or equivalent) bentonite as the source material for its buffer. This material has been extensively characterised and the results provided in open literature, as a result it has been used as the baseline bentonite clay for this CRP. Other materials of lower swelling capacity are being considered for use in filling the excavations more distant from the canister. One of these materials is the Friedland swelling clay excavated in North-Eastern Germany. The Friedland clay is not comparable to the bentonite materials considered as buffer materials in this CRP as it contains a much lower swelling clay component and although the cation exchange sites are sodium-dominated, the material has only limited swelling capacity. This clay could not be compacted sufficiently to exhibit the magnitude of swelling pressure prescribed for a buffer material but could be used in a less-demanding sealing function further away from the canister (as backfill). The data for this material also indicates that it is quite variable in quality and given its low swelling clay content this will make quality control more challenging and important.

Ukraine: Cherkassy

The Cherkassy clay provided to the CRP was found to consist of fine grained materials initially identified as montmorillonite but more likely consisting of mixed layer montmorillonite-illite minerals (see DGTS traces and cation exchange data). This clay has a swelling capacity and geotechnical characteristics intermediate to the Russian Picalevskae and the German Friedland clays. A greater proportion of its cation exchange sites are occupied by sodium than in the Russian or German clays but this does not discernibly alter its general geotechnical properties. It would seem that this material, like those from Russian Federation and Germany could not be expected to perform comparably to the reference MX-80 clay. It might be more suitable for use in backfill if the same buffer performance characteristics (especially swelling pressure) as defined for an SKB-type buffer are defined as being required for the buffer in a repository located in the the Ukraine.

6. CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE IAEA SPONSORED ACTIVITIES

6.1. Conclusions

Most of the existing multi-barrier concepts for geological repositories of HLW require the use of swelling clay materials as part of the engineered barrier system. A commonly proposed material for use as the swelling clay component is montmorillonite-rich bentonite and it is commonly considered for use in filling the volume between the excavated rock and the waste canister. The suitability of a particular swelling clay for repository application is determined by its basic mineralogic composition, chemical properties, swelling and sorption properties as well as the environment into which it is installed. It must be able to fill and maintain an ability to swell to close any open space remaining between the canister and the surrounding geologic medium (swelling properties). Its swelling properties will also act to limit oxygenation of the region adjacent to the canister once water saturation is achieved and thereby limit canister corrosion. The sealing material also needs to provide a degree of radionuclide retardation through its sorption properties and maintaining of low rate of water movement through it (hydraulic properties). These properties all need to be maintained over many thousands of years and potentially under a wide variety of groundwater conditions.

The primary purpose of this CRP was to transfer knowledge regarding approaches to preliminary characterization of candidate clay materials from member states with well-developed nuclear fuel waste management programmes to those in earlier stages of programme development. Most member states with advanced programmes and concepts requiring engineered barriers have already identified potentially suitable swelling clay materials for use in their repositories. They have characterized and evaluated a variety of bentonite products and at present have found no fundamental reasons why they are not suitable for use although work is ongoing to further detail their performance under repository conditions. These programmes are also involved in ongoing studies to evaluate alternative buffer clays for repository use as part of ongoing materials and cost optimization. For Member States with less advanced waste management programmes, and considering the volume of clay that would be required in a repository, it would in many cases be very cost-inefficient to import already characterized materials from another country. On the other hand, using inappropriate material would reduce the overall safety of the system. Hence, in order to assist in repository concept development and aid in screening of potentially suitable materials this CRP was initiated.

The CRP participants went through the stepwise process of characterizing samples and compiling information available on these swelling clays as part of an initial screening exercise to determine the potential suitability of these materials for repository use. This process allowed the participants from the member states to compare their sample materials with the characteristics of well-known swelling clays that have been selected as potentially suitable repository materials. As part of the evaluation process, the clays examined in this CRP were compared using both standard characterization tools and specialized analytical techniques that have been developed to measure and predict the swelling and hydraulic properties of swelling clays. In the course of this study several of these previously un- or incompletely-characterized materials were successfully integrated into generic swelling and hydraulic performance models.

A number of potentially suitable materials were identified in the course of completing this CRP but all will require more comprehensive analyses before they could be selected for use in a repository. The importance of defining the performance requirements for a repository under the geological and geochemical regimes actually present in the candidate host geological environments means that ultimately material suitability will need to be further defined based on local conditions and the repository concept selected. The tools developed to determine the basic properties of swelling clays are transferable from one programme to another, thereby providing a valuable international resource related to repository design. The identification of large deposits of potentially suitable sources of high-quality bentonite has also provided alternative supplies of material for consideration by international programmes.

The three most important accomplishments of this CRP were:

- The development of a network of linkages between the various national representatives involved with repository sealing materials and repository development programmes. This network of contacts and co-operation has persisted beyond the duration of the CRP and has allowed for ongoing information exchange related to qualification of swelling clays for use of swelling clay materials in repository applications. This has provided member states with less-well developed programmes with sources of ongoing technical information and assistance. Participants associated with more mature repository development programmes have in return had access to materials information developed in the course of the growth of these newer programmes, providing options for materials acquisition that may be of considerable value in the future.
- Several MS with little experience with development of an EBS system for a HLW repository have had the opportunity to improve their knowledge base regarding the DGR concept and how to qualify materials for potential use. The CRP also saw three IAEA fellowships awarded to participants from nations with lesser-developed programmes (Czech Republic, Republic of South Africa and Ukraine) to AECL’s URL. During this time they were able to observe and participate in the dismantling of a large swelling clay-based EBS simulation (tunnel plug) and observe the practicalities associated with conduct of repository-scale simulations.
- MS were given the opportunity to compare potentially suitable swelling clays from their own countries against well-characterised reference materials from several nations. The development of a preliminary screening process by the CRP participants has facilitated a process that will allow for cost-effective evaluation of previously uncharacterised materials that may be identified in the future.

6.2. Suggestions for future activities related to swelling clays

The initially proposed topic for this CRP was “Characterisation and performance studies and demonstration in underground research laboratories of swelling clays as engineered barriers of geological repositories”. As noted at the beginning of this report many of the Member States involved in this CRP were not sufficiently advanced with respect to identification and qualification of their own domestic supplies or how to undertake performance studies of swelling clays to be comfortable with undertaking or assessing demonstrations of EBS systems. The CRP therefore focussed on preliminary characterisation of key performance factors for swelling clays in the laboratory. At the end of this CRP the participants had gained considerable confidence in the basic materials behaviour factors that will control performance of swelling clays and were ready to examine how they would perform under larger scale (either through larger-scale laboratory or intermediate-scale field constructions).

As EBS demonstrations at-or-near field scale have been done at several existing URLs (e.g. Sweden, Canada, Switzerland, Belgium), and facilities are under construction in France and

Japan, there is a body of EBS-related experience available from Member States that possess such facilities but much of this skill set has not been transferred to other national programmes considering construction of a DGR for HLW disposal. There would be considerable value in the development of a relatively basic EBS demonstration under field conditions at a facility where participants from programmes with well-developed expertise could work with others to transfer experience and expertise more broadly. For those Member States with limitations in resources regarding their management of HLW, construction of domestic URLs is likely prohibitively expensive and unnecessary if access and training at existing facilities can be facilitated.

A CRP where participants work through the process from material selection and qualification to installation and monitoring of an EBS simulation and ultimately its dismantling and evaluation might be an appropriate follow-up project. The conduct of such an activity would however require careful definition of scope in order to limit the costs of such an undertaking and would require a number of years of operation before its conclusion.

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