Analogue application to safety assessment and communication of radioactive waste geological disposal. Ilustrative synthesis

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Analogue application to safety assessment and communication of radioactive waste geological disposal. Ilustrative synthesis

Autores: Carmen Ruiz López, Javier Rodríguez* (CSN)
Pedro Hernán (Enresa)
Fernando Recreo, Celsa Ruiz, Pedro Prado (Ciemat)
M. José Gimeno, Luis F. Auqué, Javier Gómez,
Patricia Acero, Álvaro González (U. de Zaragoza)
Javier Samper, Luis Montenegro, Jorge Molinero,
Jordi Delgado (U. de La Coruña)
Antonio Criado, J. Antonio Martínez, Susana Ruiz (UCM)

* Until September 2002, when he joined the Spanish Institute of Geology and Mining

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The study "Application of natural and archaeological analogues to the safety assessment of high level radioactive waste geological disposal and to communication", sponsored by the Spanish Nuclear Safety Council (CSN), with the collaboration of the Spanish national radioactive waste management company (ENRESA), has been carried out by Spanish Research Centre for Energy Environment and Technology (CIEMAT), the University of Zaragoza (UZ), the University of La Coruña (UDC) and the Complutense University of Madrid (UCM).

The initiative and management of the study have been undertaken by the High Level Waste Department of the CSN, which has conducted the study acting as coordinator.

ENRESA has participated in the tracking and technical coordination of the project providing information on the Spanish disposal concepts and its previous analogue studies.

The working team of the CIEMAT Energy Environmental Impact Department has provided support in coordination of the project and revision of the document and has collaborated in issues relating to methodologies for performance assessment of disposal systems.

The Geochemical Modelling group of the UZ Earth Sciences Department has contributed its knowledge and experience in the fields of geochemistry and hydro-geochemistry.

The Underground Hydrology Group of the UDC Civil Engineering School has contributed its knowledge and experience of aspects relating to hydrogeology and flow and transport models.

The Material Science and Metallurgical Engineering Department of the UCM Faculty of Chemistry has provided the description of the most significant archaeological analogues selected, including the corresponding summary dockets.

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peticiones@csn.es

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Contents

PROLOGUE

There is wide consensus among the scientific community that the disposal of long-lived radioactive wastes in engineered facilities located in deep geological formations is the technically available solution for the isolation of such wastes from mankind and the environment for the period required to ensure that they do not pose an unacceptable risk. It is also recognised that decisions relating to the implementation of this solution have not only a technical component but also a social one, this requiring the application of a transparent and open process.

The strategy contemplated to date in the Spanish national programme for high level long-lived waste management, as set out in successive General Radioactive Waste Plans (GRWP) approved by the Government, consists of their disposal in a deep geological facility, also named repository, although, in accordance with the provisions of the GRWP in force[1] no decision will be taken in this respect until the year 2010.

Assessment of the safety of these disposal systems requires knowledge of their performance over long periods of time. Although a large part of the necessary knowledge may be obtained from field and laboratory studies, the use of natural and archaeological evidences and phenomena that have occurred over timescales comparable to the periods of time under consideration may serve as a support for the technical arguments used in assessing the safety of such systems and for communication to technical and non-technical audiences.

Natural, archaeological and anthropogenic or industrial analogues, known as systems having some similarity to the components, events or processes of disposal systems, have been the subject of numerous studies and research efforts in recent decades, and have led to a large and widely disseminated corpus of documentation.

One of the functions of the Nuclear Safety Council (CSN)[2], as the organisation responsible for nuclear safety and radiological protection, is the performance of safety assessments for any installation implying the use, handling or storage of radioactive substances and nuclear materials prior to their authorisation, including spent nuclear fuel and radioactive waste disposal facilities. In addition, the CSN is responsible for carrying out studies, assessments and inspections of the plans, programmes and projects required for all the different phases of radioactive waste management[3] (www.csn.es).

1 5th GRWP currently in force is available via the Enresa web site (www.enresa.es)
2 Law 15/1980, of 22nd April, creating the Nuclear Safety Council
3 Law 14/1999, of 4th May, on Tariffs and Public Prices for services rendered by the Nuclear Safety Council

The strategic objectives of the CSN[4] as regards safety of spent fuel and high level waste management include the need for tracking of the activities performed at international level, in order to assess their potential for application to the Spanish case and participate in the development of safety assessment methodologies, in accordance with the schedule and objectives of the General Radioactive Waste Plans.

In keeping with the above, the CSN has made important efforts in the tracking of and participation in international activities relating to long-term spent fuel and high level radioactive waste management, especially since the mid nineties, and has carried out a series of studies on safety assessment of deep geological disposal systems, with a view to making available the scientific and technical bases for future evaluation of whatever requests might be filed, in accordance with the strategies contemplated in the national programme.

Within this framework, the CSN has promoted and conducted a study on the "Application of natural and archaeological analogues to the safety assessment of high level waste geological disposal and to communication".

The general objective of this study, known in short as "Natural Analogues", has been the compilation and review of the most relevant work and research carried out in recent decades and published to date, in order to gain insight into the state of the art as regards the potential of analogues and the use that is being made of them in the aforementioned areas. The study has been performed with the collaboration of Enresa[5] and has been undertaken by a multidisciplinary team from Ciemat and the Universities of Zaragoza (UZ), La Coruña (UDC) and Madrid (Complutense University - UCM).

The reason underlying the performance of the Natural Analogues study by the CSN arose in 1997 as a result of increasing interest in natural analogues at international level, manifested in the technical document issued by the International Atomic Energy Agency on regulatory decision-making in the presence of uncertainty[6] and in the document by the OECD Nuclear Energy Agency on confidence in the long-term safety of deep geological disposal systems[7].

4 P CSN Strategic Orientation Plan, approved by the plenary assembly on 5th February 1998

5 By virtue of the CSN-ENRESA Collaboration Framework Agreement of 2nd June 1998 for carrying out research of common interest without interfering with the functions of both organisations in licensing processes.

6 Regulatory decision making in the presence of uncertainty in the context of the disposal of long lived radioactive wastes. Third report of the Working Group on Principles and Criteria for Radioactive Waste Disposal. TECDOC-975, International Atomic Energy Agency, Vienna, Austria, October 1997.

7 Confidence in the Long-term Safety of Deep Geological Repositories. Its Development and Communication. OECD Nuclear Energy Agency, Paris, France, 1999.

As indicated below, the results of the Natural Analogues study, presented in part in different national and international forums, have been structured in various documents of a varying degree of detail and technical content aimed at different target groups.

The present publication is an *illustrative synthesis* of the study on the "Application of natural and archaeological analogues to the safety assessment of high level waste geological disposal and to communication". This synthesis, based on a general description of geological disposal and its safety assessment, defines the concept of analogues and its historic evolution and provides a summary of the potential and actual contribution made by the analogues analysed to safety assessment and communication. These are described through dockets to make the subject accessible to a wider audience.

Introduction

1. INTRODUCTION

High level radioactive wastes (HLW) consist fundamentally of the spent fuel generated by nuclear power plants, when declared a waste, and high level vitrified wastes from the reprocessing of this nuclear fuel. They are characterised by their high concentrations of medium and short-lived radionuclides and considerable concentrations of long-lived radionuclides, and also generate large amounts of heat.

These wastes may be temporarily stored under safe conditions in facilities that are monitored and controlled for several decades. However, given that they may imply a risk for human health and environment during time periods of hundreds of thousands of years, the management strategy with the greatest international consensus is disposal in facilities constructed in geological formations at depths of several hundred metres.

Deep geological disposal systems are based on placing a passive system of multiple engineered and natural barriers between the wastes and the biosphere, providing the isolation and retention of the radionuclides disposed of in such a way that they do not encompass an unacceptable risk for human health and the environment now or in the future.

Assessment of the safety of disposal systems requires forecasting of the performance of the different materials and barriers that make up the disposal system and of the total system in response to the events and processes that might take place over periods of hundreds of thousands of years, as well as evaluation of the long-term radiological consequences. The ultimate objective is to achieve *reasonable assurance* that the consequences of the events and processes that might affect the disposal system will not exceed the regulatory criteria established.

A large part of the knowledge on the performance of the materials, components and processes that may occur in the disposal system may be obtained from field and laboratory experiments. Nevertheless, due fundamentally to the long time periods to be considered, long-term safety assessment of geological disposal facility requires the use of performance predictive modelling, along with rigorous analysis of the uncertainties in predictions.

In order to address existing uncertainties, it is recognised that the quantitative safety assessment should be completed with other additional arguments (what is known as the use of "*multiple lines of reasoning*"), to increase confidence in safe disposal performance and to communicate and explain it to the stake-holders involved in the disposal decision-making. This gives rise to the so-called "*Safety Case*", which includes in addition to the quantitative safety assessment being the core of the safety demonstration, a statement of confidence in the safety indicated by the assessment (NEA, 1997a; IAEA, 1997, 2002).

The study of *natural analogues*, defined as "the presence in the environment of materials or process similar to, or that may be related to, those that are predicted to occur in some component of the disposal system" (IAEA, 1989), constitute one of the "multiple lines of reasoning" aimed at increasing confidence in the disposal safety, since they can help to understand and explain long-term performance of materials similar to those that will form part of the waste disposal system, to identify the events and processes that may affect its evolution and to develop more adequate models for its representation.

Most of the studies on natural and archaeological analogues have been promoted by waste management agencies and carried out through international collaboration in the different Framework Programmes of the European Commission (EC) and of international organisations such as the International Atomic Energy Agency (IAEA) and the Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development (OECD), among others. Furthermore, some regulatory bodies, such as those of Canada (AECB), Finland (STUK), Great Britain (HMIP), Sweden (SKI) and the United States (NRC), have undertaken their own projects or have collaborated in the most significant international projects, generally relating to the disposal concepts considered in their respective countries, with a view to developing their own bases and judgements. Thus:

- AECB has performed its own analogue studies in uranium deposits in the Canadian shield, aimed mainly at gaining insight into the behaviour of fuel and radionuclide transport in fractured media.
- HMIP (now the UKAEA) has participated in the international projects at Poços de Caldas (Brazil) and Maqarin (Jordan) after having carried out a series of projects on analogues in the British Isles, such as Needles Eye, Broubster and South Terras at the end of the 1980's.
- The NRC, which carried out an analogues project in a series of uranium deposits during the period 1981-1987 and participated in the international project at Koongarra (Australia) financed by the NEA (1987-1992), has recently focused a part of its research on the study of the Peña Blanca (Mexico) and Santorini (Greece) analogues.
- SKI has participated in the international project of the Koongarra analogue, where it has attempted to check the applicability to the study of these natural systems of its own methodologies developed in the field of geological disposal safety assessment, such as the methodology for scenario development.
- STUK has participated in the international project performed at Palmottu (Finland) since 1988, the aim being to gain insight into radionuclide transport processes in fractured media similar to those considered for geological repository siting within the Finnish programme.

The interest and potential of analogues from the regulatory point of view has also been recognised in recent regulations, such as the safety requirements for geological disposal of the IAEA[8] and 10 CFR Part 63 of the USNRC[9].

In view of the growing internationally recognised interest in natural analogues, as an approach to increasing confidence in geological disposal safety assessment, in 1997 the CSN carried out a preliminary study on the role of natural analogues in safety assessment for deep geological disposal of high level radioactive wastes (CSN, 1997). This study underlined the large number of analogue studies performed and the wide spread of the available information, strengthening the interest in performing more detailed studies.

Considering all the above mentioned, in 1999 the CSN decided to carry out the "Natural Analogues" study in order to review and analyse the results of the most significant analogue studies and identify its potential and actual contribution to safety assessment of high level waste repositories and for communication purposes, gaining, in this way, an overall, updated view of the subject. This study is described and summarized in this publication.

1.1. Description of the Natural Analogues study

The *objective* of the study on the "Application of natural and archaeological analogues to high level waste repository safety assessment and communication", promoted by the CSN and known briefly as the "Natural Analogues" study, has been to obtain a better understanding of the potential of such analogues for application to waste disposal safety assessment and for communication to non-technical audiences, as well as of the actual use made of them to date in these two areas. The general aim was to obtain a view of the state of the art as a basis for the definition of future positions and courses of action in this field.

The study has included the review and analysis of most relevant natural and archaeological analogues at national and international level, well documented in the existing literature, with especial emphasis on those most relevant for the disposal concept in granite and clay formations, as they are specifically considered in the current Spanish disposal concepts. The scope of the project has not initially included the study of analogues of near surface disposal of low and intermediate level radioactive wastes (although consideration has been given in the study to analogues of materials forming part of such facilities), nor of anthropogenic analogues, which are mainly applicable in the field of radionuclide transfer in the biosphere.

8 Geological Disposal of Radioactive Waste. IAEA Safety Standard Series-Draft Safety Requirements DS 154, 2002.10-14. OIEA, 2003. (to be approved by the Commission for Safety Standards)

9 U.S. Nuclear Regulatory Commission. 10 CFR Part 63, Disposal of High-Level Radioactive Wastes in a Proposed Geologic Repository at Yucca Mountain, Nevada. Proposed rule 10 CFR Part 63.

The *working teams participating* in the study have been the Environmental Impact of Energy Department of CIEMAT, the Geochemical Modelling group of the Department of Earth Sciences of the University of Zaragoza (UZ), the Underground Hydrology group of Civil Engineering School of the University of La Coruña (UDC) and the Material Science & Metallurgical Engineering Department of the Madrid Complutense University (UCM). These teams have experience in disposal performance assessment methodologies (CIEMAT); the study and modelling of hydro-geochemical processes (UZ); the study and modelling of hydro-geological and thermo-hydro-geochemical processes (UDC); and research into corrosion processes and microstructural and mechanical changes in metallic parts (UCM). Enresa showed its interest in the study, offering its participation through the CSN-Enresa Collaboration Framework Agreement for research purposes.

The study has encompassed the performance of the following generic activities:

- *Bibliographic review, general analysis and classification of the information from existing analogue studies* and *selection* of the most relevant according with the objectives and scope of the study. There were also identified the components and processes of a geological disposal system and the *areas of safety assessment* that might benefit from the information from analogues.
- *Detailed description of each analogue selected*, taking into account the objectives and scope of this study.
- *Detailed analysis of the application of analogue studies to the safety assessment of geological disposal repositories.* An ample set of disposal performance and safety assessments performed by waste agencies and regulatory bodies of some countries was studied in order to determine the actual use made of the information on analogues in safety assessment, as regards the definition of the disposal concept and repository design, scenarios and conceptual model development, as well as for enhancing confidence in the safety of this solution.
- *Analysis of existing strategies and information on the use of analogues for public communication of the geological disposal concept and relevant aspects to repository safety*, through the compilation and review of the material reported by waste management agencies, regulatory bodies, and other organisations (articles, brochures, videos, web sites, etc.), in which information on natural analogues has been used for illustrative purposes.

Given the nature of the review and analysis of the state of the art and the large volume and dispersion of the analogue studies published to date, in performing the study it was necessary to undertake an important effort in *bibliographic* *compilation,* in relation both to the analogue studies and research projects themselves and to the geological disposal performance and safety assessment exercises. This bibliographic review has covered the following issues:

1. Information on natural analogues existing in summary publications, results of analogue research projects, and information arising from the work performed by waste management agencies, regulatory bodies and international organisations such as the IAEA, the NEA and the EC (in particular the publications of the working group on natural analogues, NAWG).
2. Safety assessment reports, documents on model verification and validation, including international intercomparison and confidence building studies, specific technical documentation on different processes of the disposal system, etc., in order to identify the application of analogues to the different areas of safety assessment.
   The safety assessment exercises reviewed have been those performed most recently in countries with the most advanced waste disposal programmes (Canada, Finland, Japan, Sweden, Switzerland, USA), along with the Spanish exercises, with special attention paid to those carried out in granite and clay formations.
3. In relation to the objective of identifying the use of analogues in communication, the existing illustrative support materials (brochures, videos, web sites, etc.) including references to analogues for public communication of the relevant aspects of disposal systems and their safety were analysed and reviewed.

In order to compile and organise the information used in the study in a systematic manner and given the interest for the study and for subsequent uses a Database was drawn up able of establishing relationships between different information fields (organisations involved and analogue projects, authors and their fields of speciality, materials, processes and areas of geological disposal system safety assessment and potential analogues). This data base, with 1,800 references, was developed and fed by the University of Zaragoza.

The *results* of the study have been structured in documents having different degrees of detail and technical content, aimed at different audiences. These documents are indicated below:

- A general *illustrative summary* of the analogue study, aimed at a wide technical audience not necessarily familiar with the subject. This is the present publication.

- A *catalogue* of the analogues selected, with a detailed description of each analogue, aimed at a specialist technical audience. This document constitutes a second volume completing this publication.
- A detailed study on the state of the art of the potential and real application of analogues to safety assessment and communication of high level waste geological disposal, which constitutes the most important result from the technical point of view. It is edited as internal document, pending of review and publication.
- A *bibliographic database* containing summarised and inter-related information on all the documents analysed during the study including some 1,800 references in the area of analogues and safety assessment.

Some of these results have been presented at national meetings and conferences *(CSN R&D Seminaries*, 2002; *29th Meeting of the Spanish Nuclear Society*, 2003) and at international forums (*Migration'03*: 9th International Conference on actinide and fission product chemistry and migration in the geosphere). In addition, the study was presented at the *1st Meeting of the EC NANet Project*[10], where CSN is participating thanks to the experience acquired from this study, and at the EPRI Workshop "Application of natural and archaeological analogues to evaluate the adequacy of models used in radioactive waste repository performance assessment (October, 2003).

2. OBJETIVE AND STRUCTURE

The objective of this publication is to present briefly and simply to a wide and not necessarily technical audience the results of the CSN Study on Natural Analogues. The document explains the radioactive waste geological disposal concept and its analogy to natural and archaeological systems and phenomena facilitating its understanding. It also includes a summary description in the form of illustrative dockets of the analogues selected and analysed in the study, and a brief presentation of their potential and real contribution to safety assessment and public communication.

This summary document contains 7 chapters, in addition to the Introduction (chapter 1) and the present chapter (chapter 2).

Chapter 3 explains the concept of deep geological disposal for high level radioactive wastes (HLW), including the general characteristics of the repository systems currently considered in different countries' waste programmes and in

10 NANET: "Network to review natural analogue studies and their applications to repository safety assessment and public communication"

Spain, the strategies for long-term safety assessment of these disposal systems and approaches to increasing confidence in disposal safety.

Chapter 4 presents the concept of "analogue" and the historic evolution of analogue studies.

Chapter 5 describes the analogies between the different components and processes operating in a radioactive waste geological repository and the natural and archaeological systems and the processes taking place in them.

Chapter 6 identifies the different areas of safety assessment that might receive support from natural and archaeological analogues and includes a brief description of the contribution that analogue studies might make to these areas.

Chapter 7 describes the potential contribution of analogues to public communication and illustration of important aspects of geological disposal and its safety.

Chapter 8 presents the analogue selection and classification approach adopted in this study and describes the analogues finally analysed and their contribution to safety assessment and communication. These analogues are grouped into natural systems (depending on the type of geological medium) and archaeological systems. The description of the analogues is presented in summary form, through dockets, in order to facilitate the understanding by readers not necessarily familiar with the subject.

Finally, chapter 9 presents the conclusions drawn and aspects of interest identified during the development of the study.

In addition, Annex A includes a table of the most significant recent performance and safety assessment exercises of different disposal concepts, indicating the performing organisation and the objectives of the study, since many of them are mentioned when describing the actual contribution made to date by analogue information to safety assessment. In this publication is also included a more detailed description of the processes that might affect the evolution of the disposal system and their potential analogies (Annex B), completing in this way the related part in chapter 5. Furthermore, there is an index of abbreviations (Annex C), aimed at facilitating understanding of the concepts and abbreviations used in the text.

The cover of the document contains a CD-ROM with the bibliographic database of the Natural Analogues study, including 1,800 references of analogues and safety assessment. A description of this database is presented in Annex D of the document.

Geological disposal of high level wastes

1. THE MULTIPLE BARRIER CONCEPT: COMPONENTS AND FUNCTIONS OF THE DISPOSAL SYSTEM

Deep geological disposal facilities are based on passive systems made up of *multiple barriers*, designed to *isolate* radioactive wastes and *retain* the activity for a time period long enough to ensure that the risks to human health associated to any potential release of radionuclides to the environment are lower than the acceptable limits currently established by the regulatory authorities in accordance with the principles and criteria of International Organisations.

The multiple barriers concept consists of placing a series of engineered and natural barriers between the wastes and the biosphere, providing isolation and retention of the radionuclides contained in the wastes and the overall safety required for the system. As is shown in Figure 1, these barriers comprise the chemical form of the waste, the canister or container in which they are disposed of, the backfill, buffer and sealing materials used for the repository and the surrounding geological formation or "geosphere" in which the repository is constructed. The main functions of each barrier are described below:

- The *waste form* limits the rate of release of the radionuclides present in it, given the high physical and chemical stability of both the uranium dioxide (UO2) matrix, in the case of spent fuel, and the glass matrix, in the case of high level vitrified wastes.
- The *metallic canister or capsule,* constructed using materials of suitable mechanical strength and resistance to corrosion, isolates the wastes from groundwater for an initial period that will vary depending on the type of material used. It may also contribute to maintaining reducing chemical conditions favouring radionuclide retention.
- The *bentonite*, which is the main constituent of the *backfill and sealing materials* used in disposal concepts in granites and clays because of its suitable properties (low permeability, high plasticity, swelling, filtering and retention capacity, etc.), limits the flow of groundwater towards the waste packages, provides an adequate chemical environment, accommodates the possible mechanical stresses that may act on the waste packages, dissipates the heat given off by the wastes, filters fine particles or colloids that may have been generated and contains or delays the migrating radionuclides.
- The *geosphere* protects the engineered barriers, providing the necessary long-term mechanical and chemical stability, ensures low groundwater flows in the repository and offers retention and retardation of migrating radionuclides. Furthermore, the geosphere constitutes a physical barrier against potential human intrusion in the repository. The main geological media that are being considered for waste disposal are salt formations, clays and granite media.

In the performance assessment of the disposal system it is also used the term *near field*, to refer to the assembly comprising waste, canister, backfill and sealing materials and the area of rock significantly altered by the excavation of the repository, and *far field*, to refer to the rest of the geological formation. The *biosphere* is defined as the living medium that finally receives whatever radionuclides might be released from the repository, the impact of which is analysed.

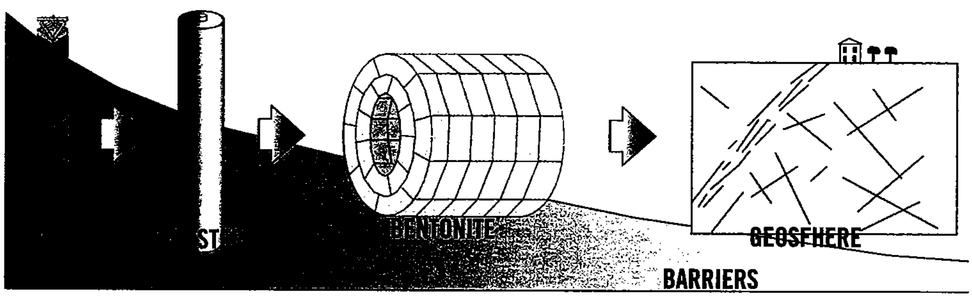


Figure 1. Schematic diagram of the multiple barrier concept of a geological disposal facility (Provided by Enresa)

Therefore, generic disposal concepts established in the majority of the national waste programmes contemplate isolation of the wastes by their encapsulation in hermetically sealed metallic canisters surrounded by clay backfill and sealing material in drifts excavated at depths of several hundred metres in stable rock formations.

The differences between disposal concepts are due mainly to the variety of sites of different specific characteristics available in each country, to the different types of waste to be disposed of and to the cost and availability of the materials considered for the engineered barriers. Table 1 compares the main characteristics of the repository concepts considered in those countries with most advanced disposal programmes (Germany, Belgium, Canada, USA, Finland, France, Japan, Sweden and Switzerland).

As regards the long-term high level waste management strategy adopted in Spain, the programme developed by Enresa in accordance with the requirements of the different General Radioactive Waste Plans (GRWP) approved by the Government has included the study of different types of geological formations (granites, clays and salts) for the potential siting of a disposal system, although the research and development activities are significantly more advanced in the case of the disposal concepts in clay and granite.

In the conceptual design of the repository in granite (Figure 2), the fuel assemblies are housed in carbon steel canisters and are emplaced, surrounded by a buffer of compacted bentonite blocks, in horizontal drifts excavated at a depth of 500 metres. Between the canisters and the bentonite blocks there is a perforated steel liner that allows the canister to be inserted into the receptacle. The galleries, tunnels and other cavities in the facility are backfilled with a compacted mix of bentonite and sand, and compacted bentonite seals are placed at the access shafts and other strategic points.

The disposal concept of the repository in clay (Figure 3) is similar to the disposal concept in granite, with the difference that the horizontal drifts, in which the canisters are emplaced, surrounded by the buffer of compacted bentonite, are excavated at a depth of 260 metres in a sedimentary clay formation. In addition, the backfilling of the facility cavities is achieved by compacting the clay arising from the excavation works, with the exception of the disposal drifts, which are sealed with blocks of compacted bentonite.

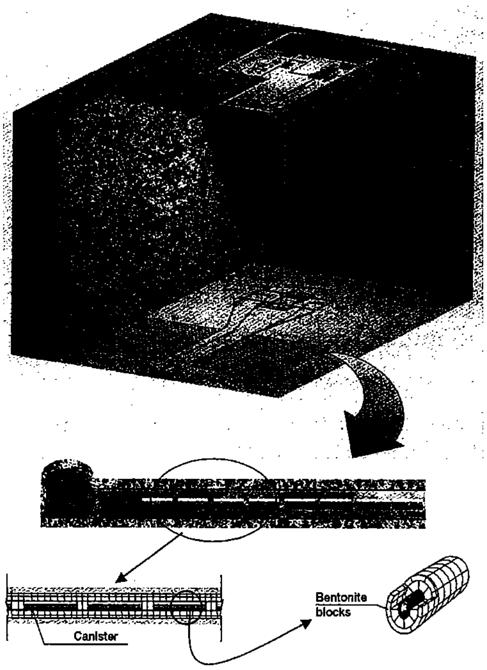


Figure 2. Schematic view of the underground facilities of the repository concept in granite, and details of the configuration of the canister and other near field barriers in this repository system. (Provided by Enresa)

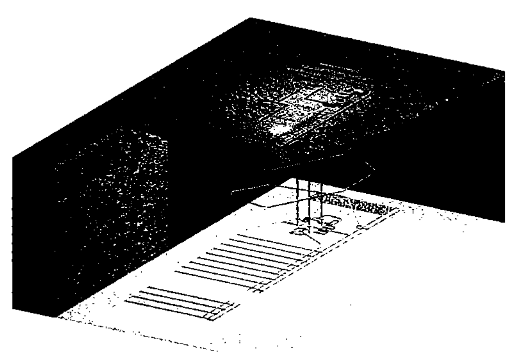


Figure 3. Schematic view of the underground facilities of the repository concept in clay. The details of the configuration of the near field would be like those of the granite disposal concept showed in Figure 2 (Provided by Enresa)

Table 1. disposal concepts considered in countries with the most advanced HLW management programmes

Country	Waste	Canister	Repository	Configuration	Backfill and sealing	Host rock	Site
Germany	Vitrified wastes spent fuel (LWR, THTR)	Stainless steel	Depth: 870 m	Individual vertical emplacement holes Vaults for the rest of the watses	crushed salt buffer backfill in salt rock Bentonite and mix of bentonite and sand in other types of rocks	Salt rock in the initial reference concept Extended to other types of rocks (sedimentary oolitic, granite)	Gorleben for the salt concept and Konrad for the oolitic formation
Belgium	Vitrified wastes, spent fuel and TRU			Horizontal tunnels	Tunnels lined with cement. Buffer: blocks of compacted bentonite. Backfill: crushed host rock	Clay	Mol
Canada	CANDU spent fuel (without uranium enrichment and burnup of 8 GWd/tU)	Titanium and pearls of glass as the filling material 1.36 tU/cask	Depth: 500 -1000m Disposal area: 4 km² Exclusion zone*: 46m	Individual vertical shafts excavated in the galleries	Buffer and backfill: mix of clay and silica sand or crushed granite in different proportions	Granite	Unspecified. Data from the 'Lac du Bonnet', Whiteshell (Pinawa, Manitoba)underground laboratory are used
USA	TRU wastes	Drums, steel canisters	Depth: 657 m	Vaults	Crushed salt and MgO	Salt	WIPP (Carlsbad, New Mexico)
USA	Vitrified wastes, LWR spent fuel	Multiple layer canister	Depth: 300m	Horizontal tunnels	No buffer	Compact, fractured, non-saturated tuffs	Yucca Mountain
France	Vitrified wastes, spent fuel and MOX	Iron alloy	Depth: 500 m Disposal area: 3.7 km²	Horizontal tunnels	Buffer and Backfill of clay	Clay	Meuse (Haute Marne)
Finland	BWR spent fue Burnup: 36 GWd/tU	Copper with internal steel container and filling of	Depth: 500 m Disposal area: 0.2 km² No exclusion zone	Individual vertical shafts in the galleries	Buffer: bentonite Backfill: sand and bentonite	Granite	Olkiluoto, Eurajoki. Selected on the basis of five sites investigated

* Distance to most significative fracture zones

Table 1. (Continued). Disposal concepts considered in countries with the most advanced HLW management programmes

Country	Waste	Canister	Repository	Configuration	Backfill and sealing	Host rock	Site
Japan	Vitrified wastes from the reprocessing of LWR spent fuel (PWR&BWR)	Carbon steel, thick overpack 1.4 tU/canister	Depth: 500 –1000m Disposal area: 5 km^2 Exclusion zone not specified	Horizontal tunnels	Buffer and Backfill of bentonite	Wide range of crystalline and sedimentary rocks	Unspecified
Sweden	LWR spent fuel (BWR&PWR) Burnup: 38 GWd/tU	Copper with iron backfilling 1.5 tU/canister	Depth: 600 m Disposal area: 0.9 km^2 Exclusion zone 100 m	Individual vertical shafts in the galleries	Buffer: bentonite Backfill: mix of clay and silica sand or crushed granite in different proportions	Granite	Detailed characterisation of the Forsmark, Simpevarp and Tierp sites approved Simpevarp Government
Switzer-land	Vitrified wastes from the reprocessing of LWR spent fuel	Stainless steel, thick overpack 1.4 tU/canister	Depth: 1000 m Disposal area: 0,5 km^2 Exclusion zone: 100 m	Horizontal tunnels	Compacted bentonite block cask support Buffer and Backfill of granular bentonite	Granite in the initial concept. Extended to other types of rocks (opaline clay).	Two potential areas: crystalline bedrock at Aargau (northern Switzerland) and opaline clays in Zurcher Weinland

2. LONG-TERM PERFORMANCE AND SAFETY ASSESSMENT

The development of a deep geological disposal system takes place through a series of successive steps (selection of conceptual design, characterisation and site selection, construction, operation, sealing and closure of the facility) accompanied by a long and flexible decision-making process that allows the incorporation of scientific and technological developments and the refinement of the repository design. The step-wise decision-making process implies long-term safety assessment of the disposal system at each relevant step, in order to have a reasonable assurance that the waste repository will not encompass an unacceptable risk for human health and the environment now and in the future.

The performance of safety assessments is also an iterative process that takes place throughout the different steps of repository development and implies activities for the integration and merging of the scientific and technical bases acquired in system development up to a given moment. This assessment requires forecasting of the performance of the different components of the repository and of the processes that determine its evolution, as well as of the overall system, along with evaluation of its long-term radiological consequences, for comparison with the acceptable limits.

It is generally recognised that the long-term safety assessment of a deep geological disposal system includes the following inter-related activities (NEA, 1991, 1997b and 1999a):

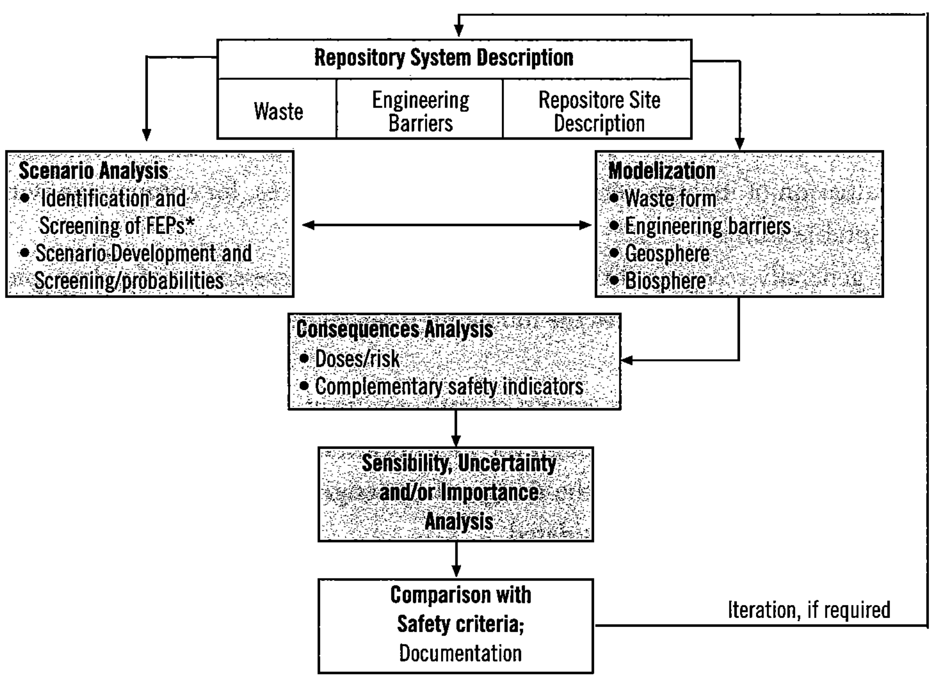
- *Definition of the context or premises of safety assessment*
- *Description of the system* through the identification and characterisation of the waste or types of waste, the design of the engineered barriers and the site (hypothetical or real) considered.
- Identification and study of the potential future situations that might affect the long-term performance of the system ("*scenario development*"). For this purpose it is first necessary to identify the features, events and processes of significance for the safety of the repository and that might affect its evolution (known as FEP's[11]), which are grouped into FEP's internal to the disposal system (reference system) and FEP's external to it. The combination of the reference system FEP's gives rise to the "reference scenario" or normal evolution situation and the action of the external FEP's on the reference system gives rise to other possible future situations or "alternative scenarios".
- *Development of the models* required to represent the performance of the system in each of the scenarios considered. The first step is the establishment of the conceptual model, the corresponding hypotheses subsequently being embodied in mathematical models and calculation codes.

11 FEP's: Features, Events and Processes

- *Integrated performance assessment of the disposal system and its possible radiological consequences,* through the application of the models drawn up for the different processes and components of the disposal system, following establishment of the input parameters and corresponding boundary conditions.
- *Sensitivity analysis* of the results of consequences calculation with respect to variations in the input data or hypotheses *and analysis of the uncertainties of the* assessment, associated mainly with the long timescales to be considered.
- Comparison of the results with the design objectives and applicable safety and radiological protection criteria.

Figure 4 shows a simplified illustration of the components of a safety assessment and its iterative nature.

Although there is international consensus regarding this general approach to safety assessment, the assessments performed within the different national pro-



* FEPs: Features, Events, and Processes

Figure 4. Schematic representation of the components of radioactive waste disposal system safety assessment.

grammes may vary depending on the phase of development of the disposal programme, the objectives of the study, the applicable safety criteria, the type of waste, the disposal concept and the site considered (NEA, 1997b). The activities of international organisations such as the NEA, the IAEA and the EC are key references for definition of the status of knowledge in this field (NEA, 1991, 1997b, 1999a, 2000 and 2002; IAEA, 1996 and 1997; Baudoin et al., 2000).

In recent years there have been numerous significant milestones and breakthroughs in the national disposal programmes of countries such as the USA, Finland and Sweden, the following being particularly outstanding:

- The entry into operation in March 1999 of the WIPP geological repository (USA) for military wastes containing long-lived transuranic elements, following certification by the EPA in May 1998.
- Ratification by the Finnish Parliament in May 2001 of the decision taken by the Government in 2000 to initiate detailed characterisation studies at the Olkiluoto site, selected for the disposal of irradiated fuel.
- The decision taken by the Swedish Government in November 2001, confirming the deep geological disposal design known as KBS-3 and allowing research work to begin for the characterisation of three potential sites.
- Approval by the US Congress in July 2002 of the Yucca Mountain site for the disposal of US HLW.

All these decisions have been supported to a large extent by a safety assessment of the disposal concepts proposed in each case (USDOE, 1996; Vieno and Nordman, 1999; SKB, 1999a; and USDOE, 1998) and the independent assessment performed by the regulatory organisations of each country and, in certain cases, by groups of experts within the framework of international organisations. *Annex A* includes a list of the most significant performance/ safety assessment exercises carried out by waste agencies and regulatory bodies of different countries in recent decades.

In Spain, Enresa has carried out generic safety assessments of disposal concepts in granite and clay formations:

- In 1997 an initial preliminary exercise was published (Enresa, 1997), with the objective of evaluating the conceptual design of a repository excavated at a site in a granitic formation, with priority given to achieving understanding of the overall operation of the disposal system serving as a guide for future activities in relation to design, site selection, R&D studies and performance assessment. The aim was also to implement safety assessment techniques and methodologies making it possible to guarantee transparency in the repository development phases, on the one hand, and flexibility for future assessment activities and exercises, on the other.
- As regards the geological repository concept in clay, a preliminary post-closure safety assessment has also been completed, the main objectives being to obtain an overall understanding of the operation of the system and of its main barriers, use the methodological techniques arising from the previous exercise in granite and acquire scientific and technological bases of the processes governing the performance of a repository in a clay host rock formation (Enresa, 1999).
- Following development of the general methodology for long-term safety assessment, implementation of the working system and setting up of the

team in these initial exercises, the strategy in this area, in accordance with the Spanish General Radioactive Waste Plan in force, consists of maintaining the safety assessment capacity developed, focusing on the performance of generic assessment exercises, resulting in the currently named "Basic-Geological Disposal in Granite" and "Basic Geological Disposal in Clay" integrating the results of the R&D and site characterisation studies carried out by Enresa.

The Spanish regulatory body, CSN, initiated in 1997 a project for the intercomparison of the most significant safety studies performed by waste management agencies and regulatory authorities of different countries. The general objective was to acquire own regulatory capacity, allowing the CSN to address the methodological developments, studies and assessments that might be requested in relation to the national radioactive waste disposal programme. The specific aims of this project were: a) to acquire a solid and far-ranging view of the state of the art of geological disposal safety assessments, and b) to assimilate the key aspects of this type of assessments, analysing aspects in common and differences in their treatment.

The results of the study, which analysed a total 14 safety assessment exercises in crystalline rocks performed between 1983 and 2000, reflect the evolution and the "state of the art" of the safety assessment key aspects, providing a basis for future activities by the CSN in this area. These results have also been published by the CSN (CSN, 2003).

3. ARGUMENTS FOR THE INCREASE OF CONFIDENCE

As it has been described in the previous section, the long-term safety assessment of a high level waste repository implies forecasting the performance and evolution of the different components and processes of the system and of the system overall, and assessing the long-term radiological consequences for their comparison with the acceptable limits.

It is recognised, however, that a perfect and detailed illustration of all the possible future situations and performance characteristics of a disposal system is not possible, and that quantitative assessment of system performance is subject to uncertainties, these being larger as longer periods of time are considered. For this reason, quantitative safety assessment should be completed with a series of complementary arguments (in what is known as the use of *"multiple lines of reasoning"*) in order to communicate and increase confidence in long-term disposal safety on the part of the decision-makers (NEA, 1999a, IAEA, 1997, 2002).

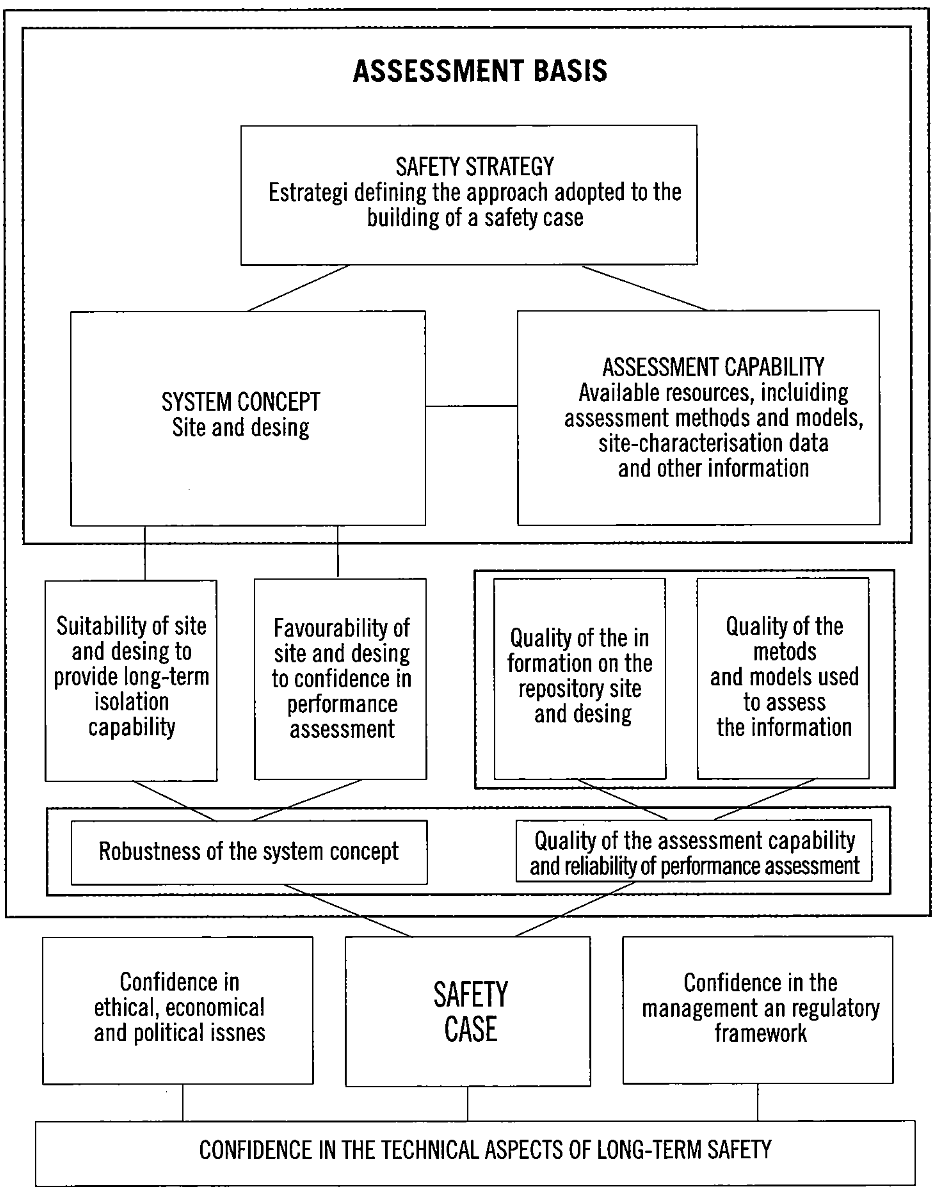
The need to obtain sufficient confidence in the safety of the geological repository by the regulatory body and the other agents involved in decision-making, in order to be able to advance in the process of repository development, has given rise to configuration of the "*Safety Case*", which goes further than "safety assessment", also including an evaluation of the confidence in the results obtained, recognising the current state of development, describing the progress made and presenting a work plan to address outstanding issues (NEA, 1999a).

Among the multiple lines of reasoning to increase confidence in the safety of the repository are the implementation of boundary or simplified calculations aimed at representing the "most unfavourable" case of disposal performance, the use of the qualitative and quantitative evidence that might be provided by natural analogues, the use of palaehydrogeological information on the sites under study and the use of different safety indicators complementary to dose and risk. The use of these multiple lines of reasoning may also help to answer the different questions and respond to the expectations of both technical and non-technical audiences involved in the decision-making process for development of the deep geological repository (IAEA, 1997 and NEA, 2002).

From the technical point of view, confidence in long-term safety is based mainly on (NEA, 1999a): 1) the robustness of the conceptual design, and 2) the quality of the assessment methodologies used and the reliability of its application. However, the confidence on the part of the social actors rests also, in addition to these indicators of technical assessment capacity, on general acceptance of the ethical, political and economic aspects of the option proposed, as well as on the reliability offered to these actors by the legal framework and the administrative and regulatory institutions involved in decision-making (see Figure 5).

The growing recognition of the importance of these ethical and social aspects in the step-wise decision-making process for the geological disposal development and the response of the waste management agencies and regulatory bodies involved, is facilitating progress and the acceptance of geological disposal programmes by the public in certain countries.

Figure 5. The elements to be considered in the evaluation of confidence in long-term safety of a geological repository.



The analogue concept. Historic evolution of analogue studies

The concept of analogue is widely used not only in the scientific field but also in many other walks of life. Its application has proven to be fruitful when attempting to understand ideas and even transfer solutions between areas of knowledge apparently distant or foreign one to the other. The great potential of the analogue concept is linked to the need to define the concept and pinpoint the details in each specific use, with a view to clarifying the objective mapped out and avoid misinterpretation.

In the field of the radioactive waste geological disposal, a natural analogue is understood as being *"the presence in the environment of materials or processes similar to, or that may be related to, those that are predicted to occur in some part of the disposal system"* (IAEA, 1989).

The emplacement of the high level radioactive wastes, especially the spent fuel, in a repository at a depth of several hundred metres in a rock formation, and the processes that might occur in this system (for instance, the potential dispersion of the contamination plume in the medium) constitute a situation intrinsically analogous to that of the uranium ore deposits in the form of UO2 that have existed in nature for periods of time much longer than those considered in waste disposal and to that of other natural geochemical anomalies.

In addition to these analogies applicable to the geological barrier and to the waste form itself, whose limited solubility under certain conditions confers upon it the function of an initial engineered barrier, there are numerous examples of analogous systems that cover a wide range of processes and environments, given the different disposal concepts currently contemplated at international level.

All analogues may be classified in a few groups representative of certain components of a disposal system and of their expected evolution (Miller et al., 2000):

- *Natural geochemical and geological systems*, applicable mainly to the waste itself and the geosphere;
- *Archaeological systems*, applicable to the canister, backfill and sealing materials and other engineered barriers; and
- Sites with anthropic contamination or *anthropogenic analogues*, applicable to knowledge of transport processes in the biosphere (not included in the scope of this project).

Until the work of Chapman *et al.* (1984), who established the basis for a quantitative treatment of natural analogues applicable to performance assessment, the majority of early analogue studies were simply extensions of geological and geochemical studies of chemical or isotopic anomalies oriented towards the interpretation of specific cases such as the *overall analogues* of a complete disposal system. There was as yet no detailed connection with the needs arising from the design of a repository or from safety assessment.

The information obtained from these early studies was fundamentally qualitative, and the overall analogue approach was in a sense not fully satisfactory. In fact, although in almost all analogues it is possible to find groups of analogies applicable to various components and processes of the disposal system, even in the most complete cases represented by uranium deposits, such as Cigar Lake or by the fossil nuclear reactors of Oklo, there are, however, no natural analogues totally representative of or suitable for a specific disposal system overall. (Miller *et al.*, 2000).

For this reason later analogue studies were focused on the study of *individual materials and processes* for which more adequate analogies could be found and which were more likely to provide quantitative and qualitative data on the long-term performance of disposal systems. Among the requirements established for a more rigorous treatment of the analogy is the definition of the following essential requirements to be considered in selecting an analogue:

1. The process involved should be clearly defined. Any other processes that might occur in the geochemical system under study should be identifiable and open to quantitative evaluation, such that their effects may be separated from those caused by the process analysed.
2. The chemical analogy should be a good one, since it is not always possible to study the behaviour of a mineral, element or isotope identical to the one to be evaluated.
3. It should be possible to estimate the magnitude of the different physical-chemical parameters, preferably using independent means, and these should not differ overmuch from those to be expected in the disposal system.
4. The system boundaries should be identifiable (understanding of whether it is an open or closed system and how much material has been involved in the process studied).
5. Given the importance of the process timescale for the analogue, it should be possible to measure this factor.

The possibility of applying the information of an analogue to a greater or lesser extent, and qualitatively or quantitatively, depends largely on how many of these conditions are satisfied and with what degree of confidence or reliability. However, it seems to be clear that in all cases there will be limitations, because it is not possible to define precisely all the conditions.

The most clear limitations of analogues are related to the following aspects: incomplete knowledge of the history of the analogue as regards details of the relevant processes and events overlapping of many complex processes that make their individual study difficult or even impossible, lack of knowledge on the initial and boundary conditions of the analogue materials and processes and those partial analogies between the analogues and the geological disposal systems leading to uncertainties in information transfer and as regards the relevance of the analogue data. It should be recognised that the materials studied are simply an approximation to those that appear in a disposal system. For example, uraninite is a good analogue of the overall composition and crystalline form of irradiated fuel, but does not contain either the internal segregations or the same quantity of transuranic elements as the latter.

Nevertheless, the aforementioned requirements, which have become "classical" in this field, allow interest in observing nature in general to be confined to the study of a series of individualised natural systems that are qualified as analogues and define clear objectives for this type of study.

The developments that have occurred in the last two decades in the study of analogues reflect the evolution of the concept of the analogue and of the types of analogues studied and underline the new approaches to such studies.

The initial definition of a natural analogue as "... the occurrence of materials or processes similar to those expected in a geological waste disposal system" (Côme and Chapman, 1986) was modified in subsequent definitions (IAEA, 1989; McKinley, 1993) to reflect the increasing use of analogues in the selection of materials for repository design, the identification and understanding of relevant processes for performance assessment of materials, components and of the overall safety of the system and in the development and verification of conceptual and mathematical models at the relevant spatial and time scales.

The interest of the different national disposal programmes has centred on different types of analogues depending on the evolution of these programmes and the associated design and safety assessment. Many of the early natural analogue studies were oriented towards determination of the stability and longevity of waste chemical forms and canister material. During the last decade the studies have been focused mainly on radionuclide transport and retardation processes in the geosphere, as a result of the important role played by dispersion and retardation in this barrier in the long-term safety of the repository system. Increasing attention has recently been observed in certain high level waste disposal programmes on the achievement of robust designs, with greater emphasis laid on the role of the engineered barriers, which will probably have an effect on future analogue studies.

More recently, growing interest in the use of safety demonstrations to non-technical audiences has been added to the aforementioned uses made of analogues, and any definition of natural analogues should reflect this particular application (Miller et al., 2000).

Consequently, as it is recognised internationally, natural and archaeological analogue studies constitute a source for understanding and explanation of the long-term behaviour of materials similar to those that will be part of high level radioactive waste disposal facilities, allowing the identification of the events and processes that might affect them and the development of more adequate models for their representation, which ultimately contributes to increase confidence in the safety of the system.

Furthermore, audiences not familiar with the technical aspects of deep geological disposal system might find the results of its performance and safety assessments more credible if they are accompanied by parallel findings from the observation of natural systems. For this reason, natural analogues might play a relevant role in communication to audiences with or without scientific and technical understanding, through the use of easily understood language.

Analogues of disposal system

Although, as has already pointed out, there are no natural analogues that are completely representative of specific disposal system as a whole, it is possible to find natural and archaeological systems in which there are groups of analogies applicable to various individual components and processes of a high level radioactive waste repository. Therefore, this study of possible disposal system *analogies* has centred on the two following major areas:

- the *materials and components* that will form part of the disposal system (waste, canister, backfill/sealing materials and geosphere) and the performance expected of these materials in the long term under the disposal conditions, and
- the relevant *processes* that might take place in the different components of the disposal system.

Identification of the possible analogues of the materials, components and important processes that might occur in the repository requires the study of the failure or degradation sequences of the engineering barrier and of the subsequent *mobilisation* and *transport* or *retention* of radionuclides, breaking down of their complex evolution into a number of interrelated mechanisms or processes that may be evaluated in a relatively independent manner.

The first section of this chapter describes the natural and archaeological systems that may be considered analogues of the different components of a geological repository. The next section deals with analogies between the main processes operating in the disposal system and the specific processes that occur in analogues and that provide the information required for the better understanding of this management option.

1. ANALOGIES OF DISPOSAL MATERIALS AND COMPONENTS

As has been explained in chapter 3, the main components or barriers of a geological disposal system for radioactive wastes are: the *chemical waste form* (nuclear spent fuel or vitrified wastes from reprocessing), the canister or capsule containing the wastes, the *backfill and sealing materials* which surround the canister and isolate it from the bed-rock, and the geological medium or *geosphere* in which the repository is located. Each of these disposal system components is briefly described below, indicating the natural and archaeological systems with which they show analogies.

1.1 Radioactive waste: Chemical forms

High level radioactive wastes are basically nuclear spent fuel (SF), when the option considered for the back end of the fuel cycle is the "open cycle" (in countries such as Canada, Finland, the United States, Sweden and Spain), and vitrified wastes from the reprocessing of SF when the "closed cycle" approach has been adopted (as in France and Japan).

1.1.1. Spent nuclear fuel

Nuclear fuel is constituted mainly by crystalline UO_2 (normally with more than 95% UO_2) manufactured in the form of pellets that are inserted into Zircaloy cladding. Once irradiated, this material is of great chemical complexity. The inventory of radionuclides present in spent fuel consists of fission products, actinides and their daughters, and of activation products formed from impurities existing in the UO_2. This inventory is distributed between the fuel pellets and the pellet-cladding gap.

The choice of this physico-chemical form of UO_2 pellets for burning in nuclear reactors is due to its great stability at high temperatures and its low solubility. This high degree of stability confers upon the irradiated fuel matrix the role of first barrier in the disposal system, since it significantly hinders the process of radionuclide dissolution in the event of failure or degradation of the other barriers.

The *natural uranium* oxides that appear in deposits of this element (uraninite and the less pure variety of pitchblende) are considered analogous to the spent fuel and may complement the information obtained on its behaviour in short duration experiments. These minerals have the same UO_2 composition as spent fuel, although there are certain differences between them. The matrix of the natural mineral usually includes as impurities traces of rare earths (REE) (La, Ce, Gd), Th and Y and a large quantity of Ca, Fe and Si; although the greatest discrepancies are in the content of radiogenic Pb (sometimes up to 25% by weight or more) these being a reflection of the age of the uraninite and pitchblende phases.

From the crystallographic point of view, spent fuel and *uraninite* are identical (they are both cubic), although they present important morphological differences that occasionally make it difficult to extrapolate the information on the natural system for use in a performance assessment exercise. In the case of spent fuel the crystalline structure may be somewhat disorganised by the intense radiation field to which it is subjected in the nuclear reactor, as a result of which it may be more soluble.

Despite these differences, the study of uraninite and its surroundings provides important clues for prediction of the long-term behaviour of the spent fuel matrix, and specifically on the process of dissolution and release of radionuclides from the fuel matrix (see section 2.1) and their retardation in secondary alteration products.

The radioactive inventory of the spent fuel includes natural series radionuclides (especially ^{238}U), fission products, actinides and their daughters and nuclear activation products. Most of these radionuclides are not found in measurable quantities in natural systems (uranium ores). For this reason, *chemical analogues* have to be used to assess the applicability of the laboratory data to the natural medium over geological timescales. In order to be considered as such, these chemical analogues must display physico-chemical properties similar to those of the radionuclides of interest (similar valence state, redox conditions and complex formation, similar type of species and stoichiometry, comparable ionic radii, sufficiently long lifetime to ensure comparable kinetic and chemical behaviour...). Uranium (U) and thorium (Th), for example, may (depending on their valence state) be used as chemical analogues of Neptunium (Np), Plutonium (Pu) and Paladium (Pa). Most of the lantinides are very closely correlated to Am and Cm. Given its high degree of mobility under very different conditions, Iodine-129 is a radionuclide that requires special attention. For high temperature conditions in the near field, its behaviour may be studied in hydrothermal deposits of copper or silver where iodide enrichment has been detected.

1.1.2. Vitrified high-level waste

The matrix foreseen in the Spanish concept for the solidification and inmobilitation of the high level wastes produced during spent fuel reprocessing[12] is *borosilicate glass*, although there are disposal concepts in which other types of chemical waste forms are considered, such as *mineral and ceramic matrices (SYNROC)*.

The advantages of a borosilicate glass as a stabilisation matrix are its physical and chemical longevity under repository conditions and its easy production. Borosilicate glasses are usually the most widely used as a matrix because of their stability a relatively low formation temperature (1100 °C), minimising waste losses due to volatisation during the vitrification process.

The natural analogue materials corresponding to these glass matrices are the *volcanic glasses*, which are fundamentally of aluminosilicate composition. Among these, the basaltic glasses are the natural materials most similar to borosilicate glasses in their composition. However, there are a number of dif-

12 Among the high level wastes to be managed in Spain are the vitrified wastes arising from reprocessing in France of the spent fuel from Vandellós I Nuclear Power Plant, which will be returned to Spain in the year 2010.

ferences between them. In particular, natural glasses do not contain any of the components of the waste, as a result of which it is not possible to observe in them the radiation-induced effects that might occur in the repository. Furthermore, boron appears only in trace amounts in natural glasses, for which reason it is not possible to study in natural analogues the impact of this element on glass durability. In addition, the ordering of certain atoms (such as Na) may be different in natural and artificial glasses, this possibly being a factor having some impact on long-term durability (Angeli *et al.*, 1998).

As well as volcanic glass, the study of *archaeological glasses* may help to understand the shorter-term behaviour of the vitrified waste matrix from reprocessing. Many of these ancient glasses have undergone conditions more unfavourable than those to be expected in a disposal system (as regards temperature and humidity) without major alteration rates.

There are abundant studies on natural glass analogues, although these have actually been carried out from a geological or mineralogical point of view more than from that of their use for safety assessment. Nevertheless, this has allowed the development of a corpus of doctrine regarding the behaviour and characterisation of this type of materials.

Most natural glasses have ages of more than two million years, and it has been observed that glass durability is greater in those cases in which these materials have been embedded in rock preventing them from coming into contact with water. It has been observed that low temperatures and very low flow rates considerably limit the hydration of the glass, making its dissolution a very slow process. For their part, the archaeological glasses appear to degrade as a result of the same mechanisms as observed in laboratory experiments on borosilicate glasses. Their study may help to gain insight into the type of alteration that is to be expected in the vitrified wastes present in a repository.

Furthermore, and although it is not applicable to the Spanish radioactive waste programme, it is interesting to note that there are many highly stable natural minerals that contain high concentrations of radioactive elements (for example zirconolite), although none of these are as abundant as uraninite. This has led to the development of mineral and ceramic matrices as an alternative to borosilicate glasses for the immobilisation of reprocessing high-level wastes. The composition of these ceramic matrices is based on stable, low solubility natural minerals belonging fundamentally to the group of titanium and zirconium oxides and to the spinels. In them, the radionuclides in the waste are strongly bound in the mineral network by isomorphic substitution. The best known ceramic matrix is SYNROC, although others are currently being researched. SYNROC is the generic name for a number of multi- phase titania based waste matrices formed by synthetic mineral assemblages (Savage, 1995), the analogues of which are the natural minerals in an environment similar to that one which will exist in a disposal system.

Finally, although the Study refers exclusively to high level waste repositories, mention should be made of the fact that there are a significant number of analogues in which studies have focused on the behaviour of the organic matter that might appear forming part fundamentally of low and intermediate level wastes[13].

1.2. Metal canister: iron (Fe) and cupper (Cu) materials

The metals present in the disposal system appear in the form of the canisters housing the spent fuel or vitrified wastes and as reinforcements for the cement structures supporting the excavations. In the case of the Spanish concept, most of these components will be made of steel. However, other metals such as copper (in the Swedish and Finnish programmes) and titanium (in the Canadian programme) may be used for the canisters. Furthermore, metals may be present as components of the wastes themselves (in the case of spent fuel assemblies: steel; Zircaloy, alloy with 98% Zr; and Inconel, alloy containing 73% Ni).

In the geological disposal concepts considered in the Spanish programme, the steel canister is designed to isolate the wastes, providing mechanical resistance against the lithostatic pressure and the stresses arising from swelling of the bentonite, as well as the increase in volume associated with the corresponding corrosion products for a period of several thousand years. It also reduces the importance of radiolysis process and brings iron to the system, contributing to the establishment and maintenance of a reducing environment around the wastes which delays radionuclide release.

Although many laboratory experiments have been performed providing abundant information on canister degradation mechanisms mainly by corrosion processes (section 2.4), their results are not easily extrapolated to periods as long as that applicable to deep geological disposal system. Archaeological analogue studies provide information on these effects at scales of hundreds to thousands of years.

The main difficulty in identifying analogues of the metals in a disposal system is that most of these metals are alloys that do not exist as such in nature.

13 The nature of the organic matter present in low and intermediate level wastes is variable and includes exchange resins, filters, plastics, paper and cleaning materials. It may also be found as part of the engineered barriers (in the form of the bitumen used as an immobilisation matrix for low level wastes) or as organic additives in the structural concrete and in the cement refill used to prevent cement-water separation and control hardening rate. The analogue studies in which the behaviour of organic matter is analysed refer mainly to the degradation of cellulose.

Consequently, the only natural analogues studied have been of Cu, Fe and steel. In the case of Cu, there are both archaeological and natural analogues (*massive deposits of native copper*), while for steel these are limited mainly to *archaeological artefacts. However,* some studies of the process of corrosion in this material have focused on natural systems of *deposits of native iron* which, although not exactly the same as the iron (steel) produced technologically (the natural mineral associations often include sulphur, oxides of Fe-Ti, magnetite and silicates), are the only natural analogues known to date.

Furthermore, studies of the reinforcing rods included in old cements in chemically reducing environments may provide useful information on steel corrosion products in environments similar to that of a disposal facility.

The information that may be obtained from the study of metal analogues refers to the following aspects of use in safety assessment: (1) the durability and longevity of iron, steel and copper (corrosion modes and rates); and (2) the properties of secondary alteration products.

1.3. Backfill, buffer and sealing materials

1.3.1. Bentonite

Clays may be used as backfill and buffer material and may even constitute the host bed-rock for the disposal system. The clay selected as buffer material in all geological disposal concepts in granite and clay formations, including those considered in the Spanish programme, is bentonite, the main mineralogical component of which is *smectite.*

Smectites are expansive clays, in other words they have the capacity to absorb water or organic liquids between their structural layers, causing an increase in volume. Smectites also have a high ion exchange and adsorption capacity and may act as pH and Eh buffers. Other important physical properties are their low permeability (limiting groundwater flows) and high degree of plasticity (that allows the bentonite to flow and seal empty spaces with a reasonable load supporting capacity, such that there be assurance that the canister will not collapse) and moderate thermal conductivity (that allows dissipation of the radiogenic heat generated inside the canister). These properties of swelling, ion exchange and hydraulic, microbiological and colloidal isolation make it one of the most important elements in the multiple barrier systems of radioactive waste repositories.

In a high level waste disposal system, the bentonite will undergo chemical processes that may cause changes in the capacity of the material to perform its functions as a barrier, especially during the thermal pulse. The main processes that may lead to degradation of the bentonite are chemical precipitation of the mineral phases in the pores or voids in this material (cementation) due to water-bentonite interaction and the progressive conversion of smectite into another mineralogical component known as illite (illitisation) when the smectite exchange cations are replaced with the potassium ion existing in the groundwater. Illite has a lower swelling capacity than smectite and higher permeabilities, therefore, illitisation implies a degradation of the initial properties of bentonites.

The study of *natural systems*, fundamentally *clays*, which have undergone alterations over time as a result of different geological processes, may be of great help in understanding the long-term behaviour of bentonite. The relevant information that may be obtained from such natural analogues is aimed at the following issues:

- Characterisation of their long-term stability, including the analysis of changes in their properties induced by natural thermal processes (for example igneous intrusions).
- Identification of the relative importance of diffusion and advection and small-scale physical heterogeneities in transport mechanisms, along with estimation of molecular diffusion coefficients.
- Study of redox front movements, including the effects produced by the presence of fractures.
- Study of speciation processes affecting radionuclides and other trace elements in the interstitial waters of these materials, including the formation of organic complexes as a result of the frequent presence of organic matter in clay materials.

The analogies of these and other processes that may occur in the bentonite barrier of a deep geological disposal system are dealt with in section 2.

1.3.2. Cement and concrete

Cement will be used to a greater or lesser extent in the construction of all radioactive waste disposal systems currently being considered. In the case of high level wastes, the main use will be limited to stabilisation of the access tunnels during the construction phase and as permanent plugs for the final sealing of tunnels and galleries. In low and intermediate level waste disposal facilities, the volumes of cement and concrete will be much larger (components of the wastes, immobilisation matrix, backfill material and engineered structures).

The favourable properties of cement for use as a barrier are its mechanical strength, its low permeability and the maintenance of an alkaline environment in the long term. The interstitial waters will have a highly specific chemical composition, characterised by a very high pH, for which reason they are known as hyperalkalines. This last property is particularly relevant as regards the radioactive waste confinement since the solubility of most radionuclides and, therefore, their potential transport, are substantially lower in strongly alkaline environments. Another beneficial aspect of these environments is the almost complete inhibition of bacterial activity due to the extreme pH conditions.

The cements and concretes that are foreseen for use in disposal systems are Portland cements, the main hydration products of which are the calcium silicate hydrates (briefly CSH compounds). These compounds form an amorphous gel that provides the binding force between the cement particles. CSH gels are thermodynamically unstable and spontaneously transform into stable crystalline forms (Steadman, 1986). The rate of this process is too slow to be measured experimentally and cannot be easily calculated, this being the reason underlying the interest of analogue studies. The process of cement degradation and the generation and evolution of the associated hyperalkaline plume, as well as their study in analogue systems, are described in section 2.9.

There are two approaches to the study of concrete and cement analogues. The first is the study of *the cements in archaeological buildings* or *ancient industrial constructions*. The greatest problem of this type of analogues is that the Portland cement used in current concretes has different physico-chemical properties from those of ancient cements, which were based fundamentally on limestone. However, certain older cements also contained CSH compounds, which have served to preserve them. The second approach consists of studying the natural occurrence of minerals analogous to the compounds encountered during the hydration of Portland cement. The presence of certain of the minerals in the cement or in hyperalkaline waters such as the interstitial waters found in these materials is not usual in nature. There are, however, certain *natural systems* in which hyperalkaline waters are found, these originating as a result of complex water/rock interactions in the alteration of ultramafic rocks or in the alteration of thermally metamorphosed limestones and marls.

The aspects of greatest interest that may be studied in these analogues are as follows:

- the cement longevity and its binding properties,
- the cement permeability to water and gas,
- the speciation and solubility of radionuclides and other elements under high pH conditions,

- the interaction of high pH fluids with surrounding rocks, as an analogy of interstitial waters migrating from the disposal system to the host rock,
- the nature and viability of microbially-induced geochemical processes, and their influence on the process of waste degradation and subsequent mobilisation of radionuclides in the near field, and
- the nature and stability of colloidal species formed in waters of high pH and in the interface between these and neutral waters.

1.4. Geosphere

In disposal concepts, the host bed-rock has the function of providing physical isolation from the potential impacts arising from the biosphere and of serving as a barrier against human intrusion, providing a stable geochemical and geomechanical environment and limiting the quantity of water coming into contact with the bentonite. Furthermore, in the event of engineered barriers failure or degradation, the very structure of the rock contributes to retarding the migration of the radionuclides released from the near field.

In order to be able to fulfil their function as a barrier for the disposal system, geological formations have to present adequate hydraulic characteristics (low permeability and hydraulic gradient) and mechanical, geochemical, seismic stability and structural characteristics. The main geological media being considered for the disposal of high level radioactive wastes are salt formations and granite and clay media. In this explanation of analogues of the geosphere and of the processes that take place in it. We shall focus on granites and clays, since the Spanish research and development activities for disposal concepts are more advanced in these media.

Granites suitable for hosting a disposal system are characterised by their low permeability and solubility, high resistance to mechanical and chemical alteration, tectonic stability and moderate thermal conductivity. The main solute transport mechanism is advection in the fractures and fissures in the medium, and radionuclide retardation is due fundamentally to their interaction with fracture filling and surfaces and to diffusion in the rock matrix.

Clay formations include diverse lithologies that range from non-consolidated clays and sludges to low-grade metamorphic rocks. Although their hydraulic, geochemical and compaction properties are variable, depending on their origin, their main characteristics are: low permeability, high retention capacity, variable plasticity and self-sealing capacity, low thermal conductivity and mineralogical/chemical homogeneity. Solute transport is by diffusion and radionuclide retardation is due to diffusion and sorption in the clay.

Most of the information on the geological medium in which the disposal system is expected to be constructed is obtained from field and laboratory observations, and from experiments in underground laboratories. The data and information obtained from characterisation studies and assessment of the sites required for the development of the disposal system are site-specific and are not necessarily of use for other potential sites. Nevertheless, studies of the geological medium in zones in which the information is transferable to other sites may be considered as analogues of the geosphere for disposal concepts in lithologies of the same type.

However, according to Chapman et al. (1984), the trend has been not to consider these sites as natural analogues since, although the number of processes of interest for a deep geological repository to be studied in these zones is very high, few of them fulfil the requirement of chemical analogy, due to such media not generally containing appreciable natural concentrations of radionuclides. For this reason, the trend has been to confine the concept of the analogue to individual processes and to systems fulfilling the requirement of the chemical analogue (and consequently with radionuclides of interest present in the analogue system) such as uranium deposits. The analogies of the main processes that may occur in the geosphere of a geological disposal system are described in section 2.

2. ANALOGIES ILLUSTRATING DISPOSAL SYSTEM EVOLUTION PROCESSES

The behaviour and evolution expected of a deep geological disposal facility for radioactive wastes once closed and sealed implies the progressive ingress of water into the disposal system, which would cause the bentonite to swell and saturate and eventually the water will reach the canister. The canister guarantees the confinement of the wastes for a period of time depending on material characteristics of at least one thousand years, after which the canister would fail due mainly to its degradation by *corrosion*. Subsequently, the groundwaters would come into contact with the wastes, causing their *alteration and dissolution* and the gradual release of the radionuclides contained therein.

The radionuclides released from the waste would be dissolved and transported in the groundwaters, firstly through the buffer materials in the near field. Saturated bentonite is practically impermeable to water, as a result of which solute transport occur fundamentally by *diffusion*. Chemical processes will take place in these materials, possibly leading to radionuclide retardation and retention (*dissolution-precipitation of impurities in the bentonite, sorption*) and to changes in the properties of the barriers, associated especially with the thermal pulse period (*cementation, transformation of smectite into illite* of poorer swelling and sorption properties).

In the case of fractured media, where the fractures are the main paths for water *flow*, the radionuclides that reach the geosphere are transported fundamentally by *advection-dispersion*. In clay formations, the main transport mechanism is *diffusion*. Likewise, a series of reaction processes takes place conditioning the chemistry of the groundwaters and possibly leading to radionuclide *retardation and retention* (sorption, dissolution-precipitation between groundwaters and host rock, diffusion in the matrix).

In addition to these main processes of radionuclide release, migration and retention, other processes take place in the disposal system that condition and have a major influence on them. These may be grouped into processes associated with the radiation emitted by the radionuclides present in the wastes (*radiolysis*, possibility of reaching *criticality* conditions), *thermal processes* associated with temperature variations of natural origin or induced by the heat generated by the wastes, processes relating to the properties and *mechanical behaviour* of the engineered and geological barriers, *processes* associated with *fluid flow* (water and gases) and *geochemical processes* that determine the geochemical evolution of the interstitial waters of the engineered and the geological barriers.

Table 2 shows the main processes that take place in each of the components of the disposal system.

A brief description of the previous processes, their occurrence and relevance in the performance of the disposal system and their study in natural and archaeological systems are presented below. A more detailed description of each of the processes and their analogies is included in Annex B.

The processes are grouped by each of the components of the disposal system affected, leaving to the end those processes that occur in various components (for example, speciation/solubility, sorption, colloid generation, microbial activity), in which the description of the process refers to occurrence in the affected components.

Componet	Process
Spent fuel/vitrified wastes	Alteration/dissolution
	Criticality
	Radiation effects (Radiolysis)
	Speciation – solubility
	Colloid generation
Canister	Corrosion
	Colloid generation
	Gas generation
Backfill and sealing materials	Dissolution – precipitation of impurities
Bentonite buffer	Dissolution-precipitation processes in variable temperature field: cementation
	Smectite-illite transformation (illitization)
	Speciation – solubility
	Molecular diffusion in the bentonite barrier
	Sorption (adsorption and ion exchange)
	Colloid generation and transport
	Gas generation and transport
Cement	Cement degradation and generation and evolution of hyperalkaline plume
Geosphere	Advection and dispersion
	Fluid flow
	Groundwater - rock matrix reaction
	Speciation – solubility
	Redox state, Redox front
	Diffusion in rock matrix
	Molecular diffusion in clay formations
	Sorption (adsorption and ion exchange)
	Precipitation- coprecipitation/ dissolution
	Colloid generation and transport
	Gas generation and transport
	Microbial processes
	Coupled processes

Table 2. Main processes that may occur in a waste disposal system, grouped by the system components that would be affected

2.1. Dissolution of the waste matrix (spent fuel or vitrified waste from reprocessing)

The chemical alteration of the UO_2 *spent fuel* matrix may arise as a result of simple dissolution or of transformation processes (oxidation) of the UO_2 into oxides having different stoichiometries. These processes are related and their coupled actuation depends fundamentally on the redox state of the system. The study of the sequence of alteration of uraninite (analogue of the UO_2 matrix) in uranium deposits under both reducing conditions (*Oklo, Cigar Lake and Palmottu*) and oxidising conditions (*Shinkolobwe, Koongarra and Krunkelbach*) has provided qualitative information on the evolution of spent fuel under different conditions and quantitative data on dissolution rates, contributing to the verification of radionuclide release models and codes.

The main processes of alteration that may affect *borosilicate matrices* are devitrification (solid state crystallisation), in which phases of greater solubility than the original glasses may be produced, and its dissolution on entering into contact with groundwater following failure of the canister. The observations performed on natural volcanic glasses and archaeological glasses have provided qualitative information on the processes and quantitative data on the ranges of dissolution rate, and have underlined the long-term stability of these materials under a wide range of conditions.

2.2. Criticality

The accumulation of fissionable isotopes (Uranium-235 and Plutonium-239) in the spent fuel, in the presence of neutrons and of a neutron moderator such as groundwater, might give rise to conditions of criticality in a high level waste repository, generating fission products and heat. In the most extreme case of the initiation of an uncontrolled chain reaction, considerable damage might be caused to the disposal system. The natural analogue of *Oklo*, where conditions of criticality were reached, provides a unique opportunity to study the transport of transuranic radionuclides and the stability of uranium minerals under such conditions. Information on this analogue has been used to analyse the possibility of criticality in a high level waste disposal system, concluding that it is highly improbable.

2.3. Radiolysis

This is the process of chemical degradation of a material as a result of the impact of high energy ionising radiations. The most important phenomenon of radiolysis in the near field of a repository is groundwater radiolysis, with its ionisation and electronic excitation and the generation of oxidising and reducing agents that might have an effect on canister corrosion, UO_2 dissolution and radionuclide solubility. The process has been studied in uranium deposits with high radiation fields, such as *Cluff Lake* and *Rabit Lake* (Canada) and *Shinkolobwe* and *Menzenschwand* (Germany), contributing to increased knowledge of its effects on the oxidisation and degradation of uraninite and to the development and improvement of models simulating the process.

2.4. Corrosion

This consists of the degradation of a material, mainly metals, as a result of reaction with the surrounding medium. It constitutes the main cause of canister failure in a disposal system. The phenomenon may occur in a generalised and/or localised manner, depending on the characteristics of the geological medium, the groundwater, the disposal system and the canister material itself. It may also give rise to the generation of secondary alteration products and gases. Its study in metallic material archaeological analogues (*Kronan Cannon*, the *Tournai Sarcophagus*, *Spanish archaeological analogues*, etc.) provides a way of quantifying the behaviour of materials with respect to corrosion on a scale of hundreds to thousands of years, and has underlined the capacity of certain canister corrosion products to delay radionuclide migration in the near field.

2.5. Dissolution-precipitation of impurities in the bentonite

The *dissolution-precipitation of the accessory products present in the bentonite* (quartz, feldspars, carbonates, sulphides, etc.) constitutes one of the main processes determining the geochemical evolution of waters in the barrier, and certain of these processes (precipitation of pyrite or siderite) favour the maintenance of a reducing environment in the bentonite. The observations carried out fundamentally in the *Cigar Lake* analogue in relation to the role played by sulphides and ferrous carbonates dispersed in the clay halo (analogue of bentonite) for the maintenance of a reducing and stabilising environment in the uranium deposit have served to corroborate the extrapolations of this effect based on laboratory studies and theoretical simulations.

2.6. Dissolution-precipitation processes in a variable temperature field: cementation in the bentonite barrier

The processes of chemical precipitation of the mineral phases in the pores or voids of the bentonite in its interaction with water due to thermohydrochemical effects are known as *cementation*. They may affect the rheological properties of the bentonite, increasing its brittleness, reducing its swelling capacity and consequently diminishing its qualities as a barrier. However, the study of the cementation of siliceous phases in the natural analogues of *Busachi* (Sardinia) and *Kinnekulle* (Sweden) has shown that the intensity of these processes would not appear to be sufficiently important to constitute a relevant phenomenon as regards the integrity of the barrier. This would confirm laboratory results, extrapolating them to the timescales of disposal safety assessment.

2.7. Transformation of smectite into illite: illitisation

This is a process of mineral replacement in which there is an exchange of the cations of the smectite for the potassium cation of the interstitial solution. It occurs naturally in geological systems at relatively high temperatures and might take place during the thermal transient period of the repository. It results in the generation of illite, with poorer properties as a barrier than smectite. Diagenetic processes and those associated with igneous intrusions are considered to be analogous to the process. Their study (*Kinnekulle, Hamra, Busachi, Burgsvik, Forsmark, Ignaberga, Murakami, Orciatico*, etc.) has provided data on the intensity or timescales required for the process to occur (confirming that these timescales should be longer than those considered in safety assessment) and has also allowed verification and calibration of the kinetic models used to describe the illisitation. At *Cigar Lake*, the clay halo surrounding the uranium deposit is made up fundamentally of illite, so, its study makes it possible to analyse the performance of this material as a barrier.

2.8. Speciation-solubility: solubility limits calculation

The speciation-solubility properties of radionuclides are specific to each element and depend on the chemical characteristics of the groundwater following its interaction with the near field barriers. The calculation of the solubility limits of the radionuclides in repositories, through the use of geochemical models and thermodynamic databases, makes it possible to determine the specific concentration of each radionuclide during its transport from the waste to the biosphere, although in most of disposal safety assessment exercises it is applied only in the near field, as a measure of conservatism. Natural analogue studies (*Oman, Poços de Caldas, Cigar Lake, Maqarin, El Berrocal, Oklo and Palmottu*) in which exercise of "blind prediction"[14] have been performed have facilitated the test and building confidence in the thermodynamic databases, conceptual geochemical models and associated numerical codes.

2.9. Cement degradation: generation and evolution of the hyperalkaline plume

The degradation of cement includes various stages that depend on the characteristics of the medium and that, as a result of interaction between the concrete and the groundwater, will condition the development and evolution of a halo of high pH fluids (hyperalkaline plume) and its effects on the near field materials (solubility of radionuclides, corrosion of metal, alteration of the host rock). The study of archaeological and industrial analogues (*Acquarosa Canal, Hadrian's Wall, Tank at Uppsala Castle*) or of natural analogues (*Oman, Maqarin*) may provide information on these processes and their effect on radionuclide transport, help in conceptual modelling of hyperalkaline plume development and, contribute to checking of the modelling approaches and codes through "blind prediction" exercises.

14 BPM: Blind Predictive Modelling exercises

2.10. Fluid flow

The flow of groundwaters in the geosphere of disposal concepts, in fractured media (granite) concentrates in the system of fractures, which act as the preferential flow path and whose geometry and configuration condition flow characteristics. This affects the transport and dispersion of the radionuclides, their retardation (by determining the quantity of surface available for diffusion in the matrix and for sorption), lithological stresses, temperature and the flow and transport of gas in the rock. Natural analogue studies (*Oklo, Cigar Lake, Palmottu, El Berrocal, Poços de Caldas*) may provide information and help to corroborate the results of field and laboratory studies on aspects such as transit times for water from the disposal repository to the biosphere, and have contributed to the development and checking of conceptual models and numerical flow codes, as well as the development and testing of innovative field techniques for hydrogeological characterisation.

2.11. Advection and dispersion

Advection (the movement of substances by the motion of the fluid in which is present) and hydrodynamic dispersion (natural mass transport from zones of higher concentration to zones of lower concentration of solute due to molecular diffusion and mechanical dispersion) are phenomena that control the transport of radionuclides dissolved in groundwaters in the geosphere of a repository, in the case of fractured formations. Although they are classical processes that have been widely studied in hydrogeology, their study in natural analogues (*Oklo, Palmottu, EL Berrocal, Maqarin*, etc.) has provided abundant information on the parameters involved in the advective-dispersive transport of solutes, and has contributed to the development of conceptual models of such transport.

2.12. Molecular diffusion

Molecular diffusion (net transport due to movement of substances from zones of higher concentration to others of lower concentration) is the main transport mechanism in the sealing materials (bentonite) and in the geosphere in the case of clay formations. In fractured media (granite), the molecular diffusion of solutes is important in the molecular diffusion of solutes in the rock zone around the fractures, known as "matrix diffusion".

Molecular diffusion in clay media has been studied fundamentally by means of laboratory and "*in situ*" experiments. Therefore, the contribution made by analogues to *diffusion in bentonite* is more limited, although it has been studied in some detail at *Cigar Lake. Diffusion in clay formations* has been studied at various analogues (*Loch Lomond, Koongarra, Dunarobba*) providing quantitative data for the diffusion of certain species of interest and underlining the conservatism of the coefficients of diffusion obtained in laboratory tests.

In fractured formations, *matrix diffusion* acts as a radionuclide sink, causing both a reduction in maximum activities and a delay in radionuclide movement towards the biosphere. It is one of the most widely considered and contrasted processes in natural analogue studies (*Palmottu, Poços de Caldas, El Berrocal, East Bull Lake*), allowing progress to be made as regards knowledge of the regulating factors (volume of interconnected porosity, importance of grain boundaries, microfractures and the mineralogy of the matrix), ranges of values of apparent diffusivity and their uncertainties to be obtained, insight to be gained into the relative importance of diffusion in the matrix compared to other retardation mechanisms and checking of the process models.

2.13. Water-rock interaction processes

These include the set of processes that condition the characteristics and evolution of the waters that will come into contact with the disposal system barriers and the evolution of certain mineralogical characteristics of the solid medium due to interaction with groundwaters, influencing radionuclide retention. They are dealt with jointly in safety assessment under the title "geochemical processes" and determine the performance of the materials in the engineered barriers and radionuclide transport in the geosphere. Their study in natural analogues has contributed to the development and checking of conceptual models for the dissolution-precipitation of secondary mineral phases, underlining their importance in the maintenance of hydrogeochemical stability in fractured media (*Cigar Lake, Poços de Caldas*), and in radionuclide retention (*Palmottu, El Berrocal*). The application in these analogue studies of geochemical or reactive transport and flow modelling methodologies and codes and the use of "blind prediction" exercises allows to increase confidence in these models.

2.14. Retardation processes: sorption and precipitation / coprecipitation

The main processes of radionuclide retardation in their interaction with the materials of the disposal system, slowing down their movement and reducing their concentration in solution are sorption, precipitation-coprecipitation, matrix diffusion, molecular filtration and ion exclusion. It is very difficult to distinguish one from another and, therefore, to assign a given effect to a single process.

Sorption is the fixation of dissolved chemical species to the electrically charged surfaces of minerals and generically encompasses adsorption and ion exchange. Its study in *bentonite* has not been considered in depth in natural analogues, although it has been studied *in clay formations (Loch Lomond)* and in the *materials filling granitic fractures (Palmottu, El Berrocal)*, which are responsible for retardation and sorption processes in fractured media.

Precipitation and coprecipitation are processes of fixation or structural immobilisation of solutes in a new mineral. Their study in analogues (*Oklo, El Berrocal, Palmottu, Koongarra, Poços de Caldas, Tono Mine, Needle's Eye, Broubster and Maqarin*) has provided evidence supporting the conceptual model of the process and has made it possible to test different calculation codes and models.

2.15. Redox state and redox fronts

The *redox state* of an aqueous system is defined by the concentrations of all the oxidised and reduced species present. Its characterisation in a natural hydrogeochemical system is an intensive methodological task due to the multiple processes by which it is conditioned and the difficulty of determining certain fundamental data, such as Eh. Work performed on the characterisation of redox processes in natural analogues (*Palmottu, Oklo*) have contributed to the refinement and improvement of the methodologies available for the in situ determination of the redox potential in groundwaters, and has provided information for conceptual models of the redox state evolution.

Redox fronts are created at the boundary between two interacting systems with different oxidisation environments. The main causes for their development in waste repositories are the oxidising and reducing agents generated during the radiolysis of water close to the canister, the introduction of air and oxidising waters during the phases prior to closure of the disposal facility and the ingress of oxidising recharge waters. As the solubility and speciation of many radionuclides are highly conditioned by the redox state of the system, their potential migration or retardation is affected when a redox front is crossed. Their study in natural systems (*Poços de Caldas, Oklo, Cigar Lake*) may help to understand their dynamics in different scenarios, providing information on the processes associated with redox front development, the parameters influencing their evolution and migration and the values of their propagation rates, contributing to the development and improvement of conceptual models of redox front evolution, etc.

2.16. Colloid generation and transport

Colloids are small solid particles (1 micron - 1 nm) dispersed in water, with a very high associated specific surface. In the waste disposal system they may exist in the natural medium or be generated as a result of degradation of the engineered barriers. They may adsorb radionuclides and constitute an additional transport mechanism, the radionuclides sorbed onto the colloid phase being excluded from possible retardation or retention effects. Their study in natural analogues (*Oklo, Cigar Lake, Palmottu, Poços de Caldas, Koongarra, El Berrocal, Maqarin*) has contributed to improving understanding of their generation processes in engineered barriers, the types, concentrations and stability of colloids in the geosphere, their reactions with radionuclides and the degree of reversibility of these reactions, their mobility and transport, etc. They also have provided quantitative data on certain processes relating to colloidal radionuclide transport and have contributed to verification and building confidence in the codes.

2.17. Microbial processes

In a radioactive waste disposal system, microbes may exist in the natural medium or be introduced during construction of the facility. The viability and possibility of their subsistence will depend on the radiation fields, competition for water with the bentonite and the availability of energy sources and nutrients. Microbial activity may contribute to the degradation of the engineered barriers, increase the solubility of radionuclides in the near field, reduce their sorption capacity and favour transport to the biosphere, because radionuclides may be sorbed selectively and irreversibly onto the microorganisms. Their study in analogues (*Palmottu, Poços de Caldas, El Berrocal, Oman, Maqarin*) has focused on the types of microorganisms and their effect on hydrogeochemical and radionuclide migration conditions, the availability of nutrients and energy sources for the microorganisms, and their tolerance to extreme radiation, temperature and alkalinity conditions in the near field of the disposal system.

2.18. Coupled processes

The thermal, mechanical, hydrodynamic, solute transport, geochemical and radiological processes that may take place in a geological disposal system are interrelated and should be analysed in a coupled manner, this being known by the term "*coupled processes*". Their study in analogue projects has contributed to their considerable development in recent years, especially as regards the so-called reactive transport models (relating to the coupling of hydrodynamic, solute transport and geochemical processes). Specifically, their study in certain analogues (*Oklo, Maqarin, Poços de Caldas*) has made it possible to develop and verify new reactive transport codes, study the genesis and behaviour of redox fronts and contrast different conceptual flow and geochemical models.

Table 3 includes a classification of some of the most significant analogues considered in the study, based on their contribution to the different processes in the components of a disposal system. The rows in the Table list the different processes that may occur in the repository, while the columns show the system components. The analogues are arranged in the Table depending on their analogies with the different processes and properties possible in the different components or sub-systems of the disposal system. Thus, for example, the analogues in which radiolysis process has been studied, due to its analogy with the radiolytic process expected in a waste repository as a result of the effect of waste radiation on the surrounding water, are listed in the box at the intersection between the column for this process (radiation effects) and the row corresponding to the spent fuel component.

The possibility of occurrence of a given process in a given component of the system is highlighted by shading the corresponding box in grey, regardless of whether or not it has been identified the study of the process in any analogue.

	Components and sub-systems				
Processes and properties	**Spent fuel and vitrified waste**	**Canister**	**Bentonite**	**Cement/ Concrete**	**Geosphere and geosphere/biosphere interface**
Disolution waste weathering / inventory/ distribution	Poços de Caldas Koongarra Cigar Lake Oklo El Berrocal Palmottu Shinkolobwe Peña Blança				
Criticality	Oklo				
Radiation effects (radiolysis, structural changes...)	Oklo Cigar Lake Cluff Lake Rabbit Lake Shinkolobwe Menzenschwand	Oklo			Rich U nodules
Corrosion		Kronan Trespaderne Mijangos Las Matillas Cerro Muriano Tournai			
Dissolution / precipitation of impurities in the bentonite			Cigar Lake		

Table 3. Summary of the potential contribution of the natural and archaeological analogues studied on the basis of their analogy with the processes that can occur in the different components of a deep geological disposal facility for radioactive wastes.

Table 3. (Continuation). Summary of the potential contribution of the natural and archaeological analogues studied on the basis of their analogy with the processes that can occur in the different components of a deep geological disposal facility for radioactive wastes.

	Components and sub-systems				
Processes and properties	**Spent fuel and vitrified waste**	**Canister**	**Bentonite**	**Cement/ Concrete**	**Geosphere and geosphere/biosphere interface**
Cementation			Kinekulle Busachi		
Smectite-illite transformation			Kinekulle Busachi Cigar Lake		
Speciation / solubility	Koongarra		Cigar Lake Oklo Oman	Oman Maqarin	Poços de Caldas Koongarra Cigar Lake Oklo El Berrocal Palmottu Oman Maqarin Needle's Eye Broubster
Diffusion in clay media					Koongarra Loch Lomond Dunarobba
Sorption (ion exchange, adsorption)					Loch Lomond El Berrocal Palmottu
Advection/ dispersion					Poços de Caldas Koongarra Oklo Cigar Lake Palmottu El Berrocal Maqarin
Fluid flow (water / gas two-phase)			Geothermal systems Cigar Lake Kinnekulle Dunarobb		Oklo Cigar Lake Palmottu El Berrocal
Precipitation / Coprecipitation			Oklo Poços de Caldas	Maqarin	Poços de Caldas Koongarra Oklo Cigar Lake Palmottu El Berrocal Maqarin Tono Mine Needle's Eye Broubster
Matrix diffusion					Cigar Lake Koongarra El Berrocal Palmottu Poços de Caldas Maqarin Oklo Tono Mine Marysvale Rich U nodules

Table 3. (Continuation). Summary of the potential contribution of the natural and archaeological analogues studied on the basis of their analogy with the processes that can occur in the different components of a deep geological disposal facility for radioactive wastes.

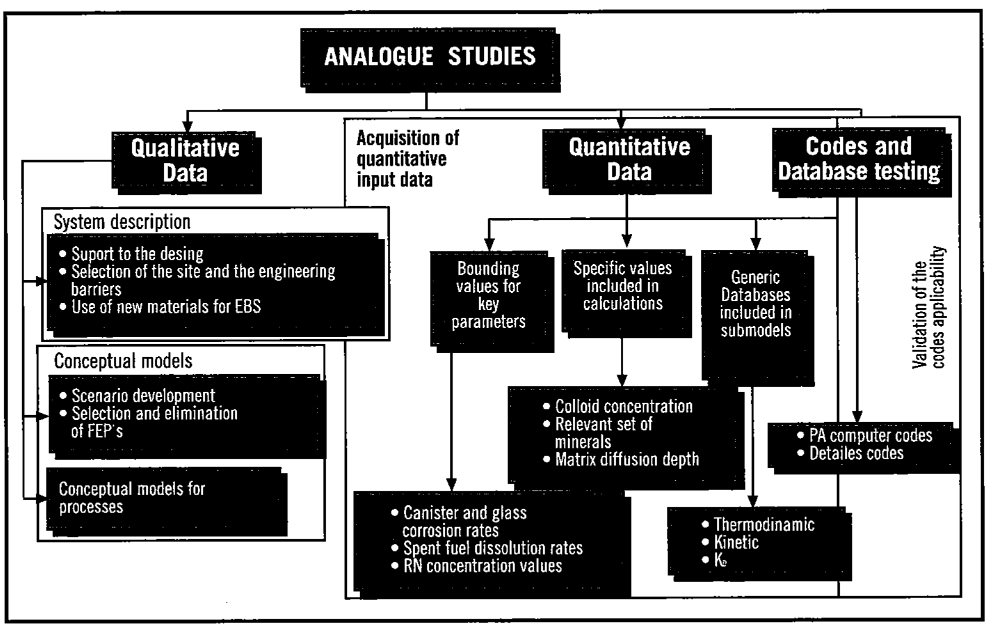
	Components and sub-systems				
Processes and properties	**Spent fuel and vitrified waste**	**Canister**	**Bentonite**	**Cement/ Concrete**	**Geosphere and geosphere/biosphere interface**
Water-rock matrix interaction				Maqarin Oman	Poços de Caldas Koongarra Cigar Lake Oklo El Berrocal Palmottu Maqarin Oman Kinnekulle Tono Mine Needle's Eye Broubster Steenkampskraal
Redox state / redox fronts	Poços de Caldas Oklo Palmottu Shinkolobwe			Oman	Poços de Caldas Cigar Lake Oklo Palmottu
Colloid generation and transport	Oklo		Cigar Lake	Maqarin Oman	Poços de Caldas Koongarra Cigar Lake Oklo El Berrocal Palmottu Krunkelbach Steenkampskraal Tono Mine
Microbiological processes			Dunarobba	Maqarin Oman	Poços de Caldas El Berrocal Palmottu
Thermal effects (heat generation, bentonite drying, thermal expansion, water chemistry changes...)	Oklo				
Mechanical processes: Swelling / creep			Cigar Lake Oklo Kinekulle		
Gas generation and transport	Oklo				
Coupled processes Termo-Hydro-Chemical (THC), reactive transport				Maqarin	Oklo Poços de Caldas Maqarin

Contribution of analogues to the areas of safety assessment

The study of the contribution made by natural and archaeological analogues to the safety assessment of radioactive waste disposal systems, described in this document, began with the identification of the main areas of such assessment that might receive support from analogue studies and a description of the potential contribution of analogue information to safety assessment. Subsequently, a review of numerous assessment exercises of geological disposal, and occasionally of references of reports supporting them, was performed, in search of the actual contributions that have been made by analogues to safety assessment (see Table 4).

This section sketches the potential and real contribution made by analogue studies to each of the areas of geological disposal safety assessment identified as being open to receiving support from them as follows: 1) system definition, conceptual design of the disposal system; 2) definition and selection of scenarios; 3) provision of quantitative data; and 4) testing of codes and databases. This contribution made by analogues to safety assessment is illustrated schematically in Figure 6.

Figure 6. Schematic illustration of the contribution made by natural analogue studies to the areas of geological disposal safety assessment which main gain support from such studies.



1. SYSTEM DEFINITION: CONCEPTUAL DESIGN OF THE DISPOSAL SYSTEM

Definition and description of the disposal system is one of the initial activities in the performance assessment. It implies definition of the system boundaries and identification of its components and of the relationships between them and with the environment. This activity receives feedback from the findings and experience acquired in the previous iterative steps of the safety assessments, providing a more detailed definition of the disposal system as this step-wise process moves forward.

The study of natural analogues present in the earth's crust for millions of years, especially of uranium deposits as geochemical anomalies that have been subjected to a natural evolution on spatial and timescales similar to those considered in a geological repository, is closely linked not so much to the genesis but rather to the evolution of the disposal concept itself. Indeed, the "independent" proof of deep geological disposal being a reasonable concept is that to a large extent it has been "copied", albeit unintentionally, from the nature. This is perhaps the clearest conclusion that may be drawn from the study of uranium deposits as natural analogues of a geological disposal system.

Consequently, one of the potential contributions of analogues in this area is their capacity to help in identifying both materials that have demonstrated properties favourable for the multi-barrier system safety and processes or situations reproducing the functions of these barriers, this possibly contributing to the definition or modification of the repository conceptual design.

Nevertheless, the study made of the actual contribution of analogues to the conceptual design of the disposal system underlines the fact that, up to date, natural analogue studies have not been widely used in this area. However, long-term reliability of the performance of the engineered barrier materials may be validated by means of natural analogue studies.

Furthermore, many of the sampling and analysis procedures, methodologies and techniques used to address the study of natural or archaeological analogues may be of use in the characterisation of the engineering barrier materials and the disposal site at different scales.

2. ANALYSIS OF SCENARIOS AND MODEL DEVELOPMENT

Analogues may help in identifying the main factors, Features of the system and its environment, Events and Processes (FEPs) affecting the long-term evolution of the disposal system, by providing evidence of them in analogue systems and, in certain cases, even the conditions under which they might occur. In this respect, they contribute to defining the most plausible future scenarios or states for which the performance of the disposal system should be studied.

They may also help in determining the relationships and influences existing between these factors or FEP's and their relative importance over time under specific conditions, since they integrate the results of processes on large spatial and time scales. They also supply information on processes that cannot be studied or even identified in the laboratory because of the spatial and time scales involved. In this way, they contribute to increasing confidence in the results of scenario development, since they make it possible to guarantee that no events or processes of relevance that might go unnoticed in laboratory studies without the complexities of a natural system have been forgotten.

In addition to helping to identify the most relevant factors from which the scenarios to be considered in safety assessment will be developed, analogue studies may contribute to the gaining of greater and more detailed insight into the individual processes that might affect the integrity of the different components of a disposal system and to the development of the corresponding conceptual models, given the analogy that exists between the processes of the disposal system and those that occur in natural systems, as has been pointed out in section 2 of previous chapter.

Different models are used to predict long-term performance of the barriers of the disposal system and the transport of radionuclides through it, these being based on extrapolations in time of the initial properties of the barriers, with consideration given to potential degradation mechanisms due to future changes in the geological, hydrogeological and hydrogeochemical conditions of the site. The study of analogues provides an opportunity to contrast these models and their hypotheses from a long-term perspective. However, in order for the analogue to be of use, it should be limited to a given process or a combination of processes of relevance in the overall model of the evolution of a disposal system. It should also confirm that the process or processes may occur and under what conditions, and that their effects and the spatial and timescales involved are similar to those simulated by the model of the process for an equivalent set of conditions.

3. SUPPLY OF QUANTITATIVE DATA

The acquisition of the data required for the application of the disposal system performance assessment models is carried out on the basis of laboratory experiments, field studies and expert judgement. The study of natural and archaeological analogues may support the numerical values of certain parameters obtained in the laboratory or, were this not appropriate, reduce uncertainties regarding the reasonable ranges of such parametric values.

4. TESTING OF MODEL AND DATABASE

One of the steps necessary to build confidence in safety assessment is validation of or increased confidence in the models simulating the performance of the disposal system, this making it possible to guarantee that these models adequately reproduce the processes that are to be modelled. Furthermore, it is necessary to know the ranges of applicability of the models, in order to ensure that these are used within an adequate range.

Strictly speaking, it is not possible to "validate" the predictive models used in safety assessment, given the impossibility of completely describing the system and of controlling the initial and boundary conditions and the spatial and time-related variation of the importance of the processes. Likewise, it is not possible to check the predictions made by the models through the direct comparison with observation of the system, due to the long timescales considered.

Natural analogues offer the possibility of: 1) calibrating the models of certain processes that have taken place under natural conditions and over timescales similar to those that will occur during the evolution of a disposal system; 2) studying the applicability of the associated databases (mainly to geochemical models) under these conditions; and 3) reducing the uncertainties and conservatism that these models usually incorporate.

Furthermore, analogues provide a "neutral" field for the intercomparison of models, which also contributes to increasing confidence in the results of the models and, therefore, in the capacity to predict the long-term performance of the DGD system (NEA, 1999b).

Table 5 summarises the actual use made of the information from natural analogues in the safety assessment exercises carried out by waste management agencies and regulatory bodies of different countries.

Table 4. Assessment exercises considered in the study. The information consulted in each case is indicated (MR: Main Report; SR: Support Report; BR: bibliographic references; PR: Peer Review), as are the analogues quoted.

Exercise	Country	Organisation / Year	Information consulted	Analogues quoted
Project-90	Sweden	SKI / 1991	IP, IA, RB	Archaeological A., Poços
SITE-94	Sweden	SKI / 1996	IP, IA, RB, PR	El Berrocal, Cigar, Poços, general references to analogues
SKB-91	Sweden	SKB / 1992	IP	Cigar, Hamra, Poços, Broubster
SR-97	Sweden	SKB / 1999	IP, IA, RB, PR	Cigar, Kinnekulle, Poços, El Berrocal, Omán, Maqarin, Krunkelbach, Nevada Test Site, Broubster, A. Archaeological (Kronan), Koongarra, Oklo, Palmottu, Dunarobba
TVO-92	Finland	YJT / 1992	RB	Cigar, Palmottu, Poços, Kronan Cannon
TILA-99	Finland	POSIVA / 1994	IP, IA, RB	Cigar, Palmottu, Hyrkkölä, Poços, El Berrocal, Omán, Maqarin
KRISTALLIN-I	Sweden	NAGRA / 1994	IP, IA, RB	Basaltic glasses , A. Archaeological, Hamra, Busachi, Poços
GEWÄHR	Sweden		RB	A. Archaeological, Oklo
AGP-Granitos	Spain	ENRESA / 1997	IP, IA	Indirect references to analogues
AGP-Arcillas	Spain	ENRESA / 1999	IP	Indirect references to analogues
Mol, Marivoet et al., (996)	Belgium	ONDRAF/ NIRAS / 1996	IP, RB	No references to analogues
NIREX-95	Great Britain	NIREX / 1995	IP	No references to analogues
NIREX-97	Great Britain	NIREX / 1997	IP, IA, RB, PR	No references to analogues
H-12	Japan	JNC / 1999	IP	Basaltic Glasses, Archaeological A, Oklo, Koongarra, Hamra, Busachi, Murakami, Tono Mine, Maqarin, Poços, Nevada Test Site
AECL-94	Canada	AECL / 1994	IP, IA, RB	Cigar, Oklo, Kinnekulle, Avonlea
NRC-IPA Phase II	USA	CNWRA	IP, RB	Peña Blanca, Valles Caldera, Santorini
TSPA-VA	USA	US. DOE / 1998	IP, IA, RB, PR	Climate A. y Termo-Hidro-Chemical, Peña Blanca, Santo-rini, Cigar, Poços, Nevada Test Site, Oklo
TSPA-SR	USA	US. DOE / 1998	IP, IA, PR	Multiple references (revision of information on analogues)

Table 5. Use of information from analogue studies in radioactive waste disposal safety and/or performance assessment

	AECL-94 (Canada)	Gewähr (Switzerland)	Kristallin-1 (Switzerland)	SKB-91 (Sweden)	SITE-94 (Sweden)	SR-97 (Sweden)	TVO-92 (Finland)	TILA-99 (Finland)	TSPA-VA (USA)
Poços de Caldas	Support for conservatism of the spent fuel dissolution model		Establishment of boundary conditions on the redox front Support for nuclide retention capacity by canister corrosion products Support for the difficulty of determining matrix diffusion processes Checking of thermodynamic codes and databases and solubility data	Data for contrasting of solubility limits Support for redox front propagation model Corroboration of the hypothesis of limited importance of colloids in retention and transport processes	Support for redox front propagation model	Data for contrasting of solubility limits Support for redox front propagation model Support for hypotheses regarding coprecipitation and colloidal transport processes	Provision of data on colloids and microbe population for model development	Data for contrasting of solubility limits	Checking of thermodynamic codes and databases
Koongarra						Support for the transport approach in the canister failure scenario Provision of data on matrix diffusion depth			
Cigar Lake	Demonstration of long-term stability of the system analogous to the DGD Support for conservatism of spent fuel dissolution model		Demonstration of long-term stability of the system analogous to the DGD Checking of thermodynamic databases	Proof of conservatism of spent fuel radiolytic oxidation rate. Contrasting of solubility values	Proof of conservatism of spent fuel radiolysis and oxidant dissolution model	Verification of solubility limits Development and testing of radiolysis model conservatism Checking of thermodynamic databases	Corroboration of spent fuel dissolution model	Development of SF dissolution model Verification of solubility limits. Checking of thermodynamic databases	Support for spent fuel stability under reducing conditions
Oklo	Support for SF performance model	Support for radionuclide release conceptual model	Checking of thermodynamic databases	Support for spent fuel radiolytic oxidisation model		Support for criticality scenario approach Development and testing of groundwater mixing code and hydrogeochemical codes			Support for conceptual models of nuclide mobilisation and transport processes Support for igneous intrusion and criticality scenario approach Checking of hydrogeochemical codes

Tabla 5. (Continuation). *Table 5. Use of information from analogue studies in radioactive waste disposal safety and/or performance assessment*

	AECL-94 (Canada)	Gewähr (Switzerland)	Kristallin-1 (Switzerland)	SKB-91 (Sweden)	SITE-94 (Sweden)	SR-97 (Sweden)	TVO-92 (Finland)	TILA-99 (Finland)	TSPA-VA (USA)
El Berrocal					Data on matrix diffusion depth	Reference data on solubility limits		Climatic scenarios study qualitative data and methodologies Palmottu	
Palmottu						Climatic scenarios study qualitative data and methodologies Data on matrix diffusion depth Checking of groundwaters mixing code		Climatic scenarios study qualitative data and methodologies	
Kinnekulle	Support for bentonite performance conceptual model Checking of illitisation process code				Support for siliceous cementation conceptual model	Support for siliceous cementation conceptual model Checking of illitisation process code			
Dunarobba						Evidence for the role of clay as a barrier to microbial activity			
Maqarin			Contrasting of solubility models and values			Support for development of hyperalkaline plume evolution conceptual models Corroboration of the long-term durability of concrete barriers/systems		Support for development of hyperalkaline plume evolution conceptual models	Checking of reactive transport codes
Oman			Contrasting of solubility models and values			Checking of thermodynamic databases		Checking of thermodynamic databases	
Kronan Cannon	Testing of copper corrosion models						Corroboration of copper corrosion rate		

Application of analogues in communication

Natural and archaeological analogues have the potential to provide some of the best evidence and arguments to increase confidence in the safety of radioactive waste deep geological disposal, since in addition to providing data or serving as cases for checking of the assessment models, they may provide an illustration or non-technical information for a wide range of audiences. Indeed, in environments not relating to safety assessment, this use of analogues is considered to be one of their main reasons for being.

Analogues constitute one of the main arguments that may be used to explain the long-term performance of the disposal system to non-technical audiences. In general there is wide agreement that many groups of people not familiar with the technical aspects of geological disposal will give greater credence to the conclusions of safety assessments if they are accompanied by natural parallel examples allowing for comparison and understanding (Miller et al., 2000).

1. METHODS AND SUPPORTS FOR THE USE OF ANALOGUES IN COMMUNICATION

There are currently good examples of the use of analogue information to illustrate relevant safety aspects of geological disposal in public communication, both by waste management agencies and regulatory bodies of different countries and by other institutions, organisations or private individuals. This illustrative use of information on analogues has been carried out using various types of material: videos, brochures, articles, Internet resources (websites), itinerant exhibitions and other types of public activities.

The following are particularly significant as regards materials on natural analogues published or developed directly by regulatory bodies or waste management agencies:

- The different types of illustrative brochures on natural analogues published by European waste management agencies such as NIREX (Great Britain), POSIVA (Finland) or SKB (Sweden), as well as two brochures incorporated in the website of the United States Yucca Mountain Project and entitled: "Oklo: natural nuclear reactors", which is dedicated to this particular analogue, and "Scientists look to nature for insight into how a repository would perform", which explains in simple terms the way in which analogues are used to assess the performance of the Yucca Mountain disposal concept. Among the analogues considered, some are particularly "graphic" for the general public, such as the Altamira caves in Spain. These brochures are one of the most widely used supports for public communication. They report on specific aspects clearly and directly.

- The widely distributed video "Traces of the Future: Lessons from Nature for Waste Disposal", which is the result of international collaboration within the framework of the European Community (coordinated by NAGRA and including the participation of SKB, NIREX, CEC, US-DOE, ONDRAF, ENRESA and AECL). This video refers to different natural and archaeological analogues (Cigar Lake, Oklo, Poços de Caldas, Dunarobba, Hadrian's wall, etc.) to help the public to understand the processes that might affect a disposal system in the long term and contribute to increasing confidence in the safety of the DGD concept. With similar objectives in mind, SKB has published a collection of three videos entitled: "Nature´s own repository" (dedicated to the Cigar Lake analogue), "Nature's own technology" (on archaeological analogues) and "Nature's own nuclear waste" (focused on the Oklo analogue). Likewise, Enresa has published a video on the work carried out at the analogue of El Berrocal (Spain) and its relationship to disposal system safety assessment.
- The more general illustrative publications on deep geological disposal concept, such as the one entitled "This is how we manage Sweden's radioactive waste" (SKB Activities, 1995), which dedicates a few pages to analogues and their use in the area of safety assessment, referring to a large number of natural and archaeological analogues.
- The websites of waste management agencies and regulatory bodies, in which there are numerous references to analogues, although some are lists of projects and links to documents of a technical nature. Outstanding for their educational interest are the websites of NAGRA, POSIVA and SKB, and the one of the US Yucca Mountain project, which contains references to certain natural and archaeological analogues in relation to the long-term effectiveness of the disposal system barriers and the study of the disposal system evolution.
- The itinerant exhibition performed by SKB during the summer aboard the ship Sigyn along the Swedish and Finnish coasts, which presents different analogues (Cigar Lake, Oklo, Dunarobba, Kronan Canton, etc.). This exhibition allows for greater interaction with the public, who are able to submit their doubts and questions for replies by technical personnel at hand, and constitutes a highly positive experience that is receiving a warm welcome (Smellie et al, 2000).

Among the material published by other organisations, independently from waste management agencies and regulatory bodies, special mention might be made of the websites of organisations such as the IAEA and NEA, as well as of the website of the Natural Analogue Working Group (NAWG; http://www.natural-analogues.com/), which is a basic reference, if not the most important, as regards the study of natural analogues, their communication and their application in deep geological disposal studies.

Table 6 illustrates the potential uses of analogues analysed in public communication, and their actual use, when this exists, indicating the medium or support used for this communication.

2. ASPECTS OF DEEP GEOLOGICAL DISPOSAL ILLUSTRATED THROUGH THE USE OF ANALOGUES

The material reviewed in relation to the use of analogues for illustrative purposes includes both natural and archaeological analogues and fundamentally illustrates the deep geological disposal concept and aspects of the performance of the materials in the barriers and of the processes that might control their evolution over time, as indicated below:

- The suitability of the multiple barrier design of a disposal facility and its performance over time is an aspect that is frequently highlighted in the use of analogues for public communication through simple diagrams illustrating the similarities between the Cigar Lake analogue and the geological disposal system. This analogue clearly illustrates the parallelism that exists between the geological context of the deposit and the majority of the disposal concepts considered internationally. This similarity, along with the stability of the deposit on geological timescales (without traces of its presence on the surface), constitute a classical illustration for communication of the geological disposal safety.
- The performance of the materials in the barriers and the maintenance with time of their isolating properties is another frequently illustrated aspect in educational material on analogues. The observations carried out in these systems confirm the stability and longevity of borosilicated matrix, of the different types of casks (fundamentally steel and copper), and of cement and concrete and the isolating capacity of bentonite barriers in the long term.
- Relevant processes for the evolution of the disposal system. Analogues underline the effectiveness of long-term radionuclide retention processes in engineered barriers, and especially in the geosphere.

3. IDENTIFICATION OF AUDIENCES AND COMMUNICATION STRATEGIES

The overall corpus known as "non-technical audiences" includes large groups of different origins, such as the social and political actors involved in decision-making, various non-governmental organisations, some environmentalists and the public. All of them have in common the fact that they are not familiar with the conceptual and technical aspects of deep geological disposal. This characteristic is common also to a large part of the "technical" audiences or those having a scientific background, although in these cases the communications should be oriented differently from those aimed at other groups. In general, the communication and demonstration of safety should be adapted to the background and expertise of the different groups, the message being modified to suit their characteristics (NEA, 2002).

The use of information from analogue studies in communication to non-technical audiences has been aimed mostly at the general public, for which reason its educational use emphasises simplification and focuses on the parallelism existing between analogues and the disposal system. In such cases it has become clear that difficulties are encountered in finding a suitable way to present the evidence provided by analogues of the long-term safety of disposal systems, balancing the simplicity required to facilitate understanding by non-technical and non-specialist audiences with the scientific rigour necessary to prevent the value of the information from being minimised or made trivial.

The use of audiovisual methods allows for a more complete and detailed presentation of the results obtained from analogue studies and of their role for explaining geological disposal. When this type of support is used, both natural and archaeological analogues are normally included and very varied messages are used regarding the repository concept, the barrier materials and radionuclide migration processes.

It is quite clear that the public has difficulties in understanding geological processes, which occur on a scale of tens or hundreds of thousands of years, a fact that requires careful selection of the analogues considered and greater effort in the way they are treated. On the other hand, the greater familiarity of the public with archaeological materials and the timescales involved in the history of this type of findings, makes archaeological analogues a closer analogy suitable for communication to non-technical audiences.

Almost all the communication media used to date have been basically passive. The experience initiated by SKB in Sweden on board the ship Sigyn, which allows for greater interrelations with the public, who are able to submit their doubts and questions for replies by technical personnel at hand, constitutes a highly positive experience that is receiving a warm welcome (Smellie et al, 2000).

Table 6. Summary of the potential and actual use made of information on natural and archaeological analogues in communicating to the public certain aspects of relevance for the deep geological disposal (DGD) concept and its safety

Analogue	Potential contribution to communication	Actual use in public communication
Poços de Caldas (Brazil)	Morro do Ferro: Evidence of the mobility of thorium from the deposit being very low Osamu Utsumi: Shows the capacity of the redox fronts that may occur in the near field of a repository system to retain and delay radionuclides	Video "Traces of the Future: Lessons from Nature for Waste Disposal"
Koongarra (Australia)	Illustration of the capacity of the near field and the geosphere of a DGD system Ito retain radionuclides, even in the case of possible failure of the sealing system	
Cigar Lake (Canada)	Illustrative example of a disposal system due to the parallelism between this and the natural geological system Illustration of the isolating capacity of the multi-barrier system for deep geological disposal: uranium deposit of 1,300 million years, with no traces on the surface	Subject of the video "Nature's own repository" published by SKB and of the video "Traces of the Future: Lessons from Nature for Waste Disposal" Websites of Posiva (www.Posiva.fi/ englanti/ index.html), SKB (www.skb.se) and NAGRA (www.Nagra.ch/) SKB exhibition on board the Sygn
Oklo (Gabon)	Evidence that radioactivity is a natural phenomenon: even nuclear chain reactions have taken place in nature (natural reactors) Proof that the products of nuclear reactions may be retained in natural systems	Subject of the SKB video "Nature's own nuclear waste" and of the video "Traces of the Future: Lessons from Nature for Waste Disposal" Brochures: "Oklo: natural nuclear reactors" and "Scientists look to nature for insight into how a repository would perform" Websites of Posiva (www.Posiva.fi/ englanti/ index.html), SKB (www.skb.se), Yucca Mountain (www.ocrwm.doe.gov) and a university (www.curtin.edu.au/curtin/ centre/waisrc/OKLO/ index.shtm) SKB exhibition on board the Sygn
El Berrocal (Spain)	Overall and simple example of disposal safety, showing the limited mobility of uranium in the granitic pluton	Subject of a video published by ENRESA
Palmottu (Finland)	Shows the stability of spent fuel	
Kinnekulle (Sweden)	Evidence of the longevity and maintenance of the desired properties of the clay barrier	
Dunarobba (Italy)	Illustration of the isolating and preservation capacity of clays over long periods of time (more than one million years)	Wide broadcasting of its existence In the video "Traces of the Future: Lessons from Nature for Waste Disposal" In the websites of Nagra (www.nagra.ch) and SKB (www.skb.se) In the SKB exhibition on board the Sygn
Loch Lomond (Scotland)	Illustration of the excellent isolating capacity of clay	In the video "Traces of the Future: Lessons from Nature for Waste Disposal" In the SKB exhibition on board the Sygn
Oman	Illustration of the hyperalkaline conditions associated with the existence of cement/concrete seals in the disposal facility	

Table 6. (Continuation). *Summary of the potential and actual use made of information on natural and archaeological analogues in communicating to the public certain aspects of relevance for the deep geological disposal (DGD) concept and its safety*

Analogue	**Potential contribution to communication**	**Actual use in public communication**
Maqarin (Jordan)	Illustration of the effect on DGD safety of the hyperalkaline conditions associated with the existence of cement/concrete seals in the disposal facility	SKB website (www.skb.se)
Kronan Cannon (Sweden)	Illustration of the corrosion resistance and longevity of copper, increasing confidence in its use as a material for casks	In the video "Traces of the Future: Lessons from Nature for Waste Disposal" SKB website (www.skb.se) In the SKB exhibition on board the Sygn
Inchtuthil Nails (UK)	Evidence that where there are large volumes of steel, part of it may not be affected by corrosion	In the video "Traces of the Future: Lessons from Nature for Waste Disposal"
Tournai Sarcophagus (Belgium)	Illustration of the role of the protective layer of corrosion on metals preventing their deterioration	
Acquarossa (Italy)	Evidence of the good degree of conservation of a concrete with carbonates in its interior	
Hadrian's Wall (UK)	Understandable demonstration of the durability of cement	In the video "Traces of the Future: Lessons from Nature for Waste Disposal"
Changsha tomb (China)	Evidence of the isolating capacity of clay	

Dockets illustrating the analogues selected

1. SELECTION OF ANALOGUES FOR CONSIDERATION IN THE PROJECT

The large quantity of information on analogues in existence at international level and its wide dispersion, the different degree of detail and depth of the analogue studies and their different levels of interest for the Spanish geological disposal programme made it necessary to select the most significant analogues to be considered in the CSN study, taking into account the following criteria:

- Representativity in the analogues of the deep geological disposal concepts for high level wastes contemplated in the Spanish programme in both granites and clays.
- Relevance of the analogue studies, valued on the basis of the quantity and quality of the information provided by them and the number of organisations involved. Special attention has been paid to studies in which regulatory bodies have participated.
- Potential capacity to provide information for the different aspects of the assessment of the long-term performance of the repository materials and components, and recognised relevance of the contribution made to safety assessments consulted in a preliminary manner (SKI, 1991 and 1996; Goodwin et al., 1994; Nagra, 1994; Neal et al., 1994; USDOE, 1998; SKB, 1999a; Vieno and Nordman, 1999; and JNC, 2000).

The reference framework for this selection might be modified in the future as a result of the evolution of the analogue concept, of possible developments in studies of these systems and of changes associated with the progress of the Spanish disposal programme, to which the lessons learned in this Study are to be applied, since they are still in the initial phase of definition. If considered necessary, the selection might be updated in keeping with these modifications.

As a result of the selection process, a total 43 natural geological analogues have been considered in this Study, classified into 10 main analogues and 33 secondary analogues according with the importance to safety assessment of the information they might provide; 8 archaeological analogues have likewise been selected, grouped on the basis of the same criterion into 6 main analogues and 2 secondary analogues[15].

15 Among the main analogues, hydrothermal systems, analogues of materials such as glass or bitumen and anthropogenic analogues have not been studied due to being beyond the scope of the project or because they are very specific systems, even not strictly natural analogues in certain cases.

The natural analogues have in turn been classified depending on the geological system that they represent, as follows:

- Uranium deposits
- Clay media
- Hyperalkaline environments

The archaeological analogues have been grouped as follows:

- Analogues of metallic materials
- Analogues of cement and concrete
- Analogues of the clay barrier

Furthermore, given the archaeological wealth of the Iberian Peninsula and the performance of a previous project by the UCM group on national archaeological analogues (Enresa, 2000), the following Spanish archaeological findings have been analysed (selecting the most representative among all those studied in recent years): Trespaderne Bar (Burgos), Las Matillas nail (Madrid), Mijangos nails (Burgos), Cerro Muriano nails (Córdoba).

Table 7 lists the analogues described, and figure 7 their geographical location.

Table 7. Natural and archaeological analogues analysed (refer to their location in figure 7)

Level	Type of system	Analogue	
Main Analogues	Uranium Deposits	Poços de Caldas (Brazil) Koongarra (Australia) Palmottu (Finland)	Oklo (Gabon) Cigar Lake (Canada) El Berrocal (Spain)
	Clay Analogues	Kinnekulle (Sweden)	Dunarobba (Italy)
	Hyperalkaline Environments	Oman	Maqarin (Jordan)
	Archaeological metals	Kronan Cannon (Sweden) Roman nails (Inchtuthil, United Kingdom) Tournai Sarcophagus (Belgium)	Spanish deposits: • Trespaderne Bar • Las Matillas nail • Mijangos nails • Cerro Muriano nails
	Archaeological ceramics and vitreous materials (cements, glasses)	Acquarossa (Italy) Hadrian's Wall (Scotland)	
	Archaeological clays	Changsha tomb (China)	
Other Analogues/ Secuondary Analogues	Uranium Deposits	Tono Mine (Japan) Shinkolobwe (Zaire) Needle's Eye (United Kingdom) Broubster (United Kingdom) South Terras (United Kingdom)	Krunkelbach (Germany) Marysvale (United States) Lianshanguan (China) Steenkampskraal (South Africa) Kanyemba (Zimbabwe) Peña Blanca (Mexico)
	Studies in sediments	Loch Lomond (United Kingdom) Abyssal Plain of Madeira (Portugal)	U-rich nodules (Switzerland, United Kingdom, Holland) Gorleben (Germany)
	Zones of contact with igneous rocks	Stock Eldora (United States) Stock Alamosa River (United States)	Caldera Valleys (United States)
	Clay materials	Orciatic (Italy) Counties of Li An and An Ji (China)	Murakami (Japan)
	Alteration of rocks	Falmouth Breakwater (United Kingdom)	Eye Dashwa pluton (Canada)
	Hydrothermal systems	Auriat granite (France) Newberry (United States)	East Bull Lake pluton (Canada)
	Iron and Copper	Bühl (Germany) Disko (Greenland)	Keweenaw (United States) Hyrkköla (Finland)
	Glass and SYNROC	Basaltic glasses Rhyolitic glasses	Zirconolites
	Archaeological metals	Santorini (Greece)	
	Archaeological ceramics and vitreous materials (cements, glasses)	Tank of Uppsala Castle (Sweden)	

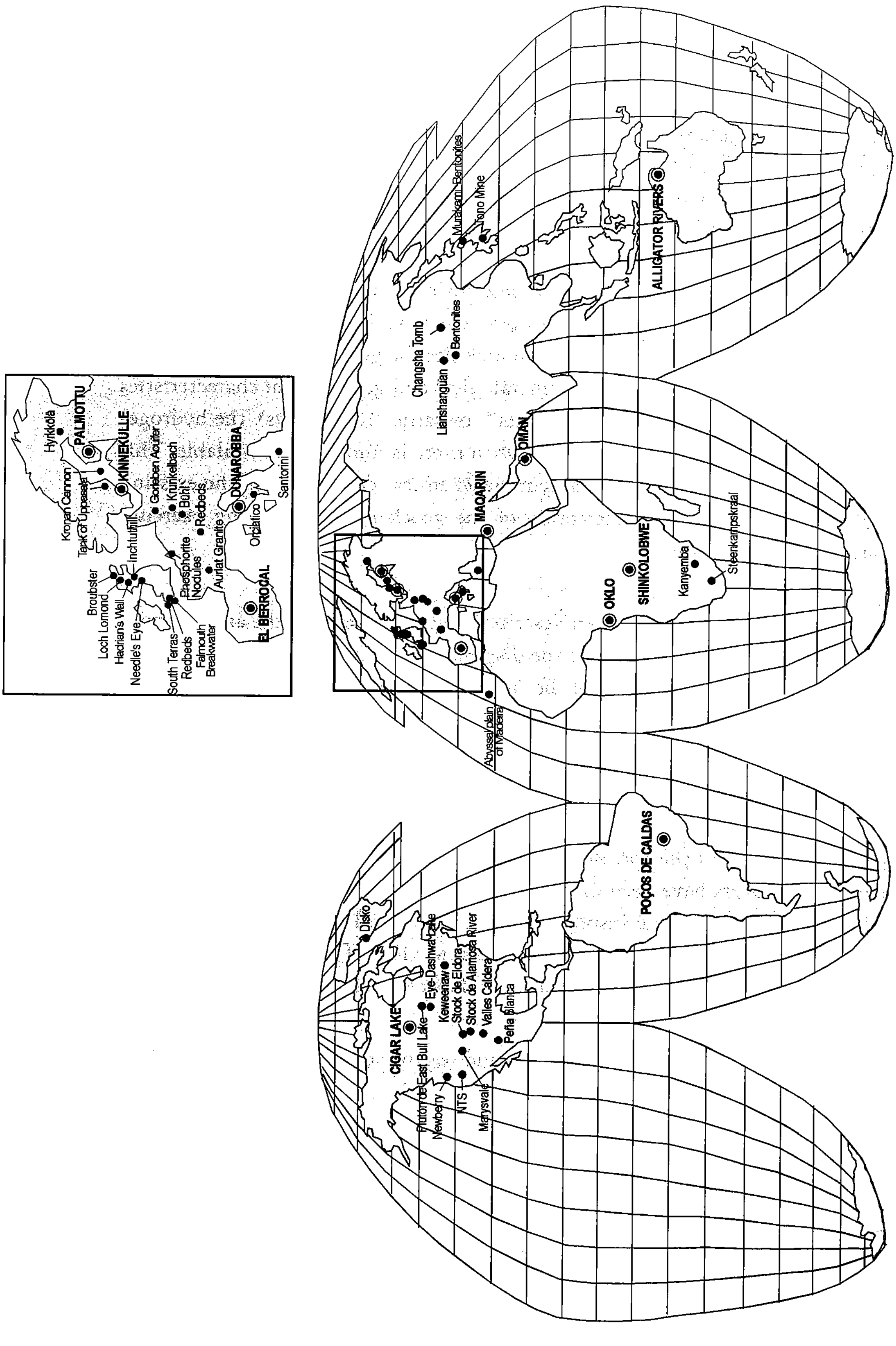


Figure 7. Geographical location of the natural and archaeological analogues studied

2. DESCRIPTION OF ANALOGUES: ILLUSTRATIVE DOCKETS

A detailed analysis has been performed on the most significant analogue studies carried out by different international organisations, waste management agencies and regulatory bodies. The results of this analysis are included in the previously mentioned project document "Analogue Catalogue", which also contains a preliminary analysis of their potential contribution to safety assessment and communication.

Specifically, for each analogue, a description of the natural system is presented, this obviously being much more extensive in the case of the natural analogues. In this case the description includes the geographical framework in which the system is located, the mineralogical and geochemical characteristics of the solid phases (in uranium deposits[16], metallic ores or clays), the hydrogeology and groundwater flow models, when there is information available. This information is completed with a hydrogeochemical description of the existing aqueous systems[17] and a description of the geochemical modelling exercises carried out in each case.

Once the analogues has been described and characterised, the analogy with the components and processes of the disposal system is analysed and explained and a brief indication is given of the contribution made to the different aspects of safety assessment (description of the system, scenarios and conceptual models, provision of quantitative data, checking of models and databases), as well as of their contribution to communication.

On the basis of the corresponding detailed descriptions above mentioned, summary dockets have been drawn up for each of the main analogues selected and analysed, these being inspired by those developed by De Putter & Charlet (1991) and attempting to provide a graphic view of the most outstanding aspects of these analogues. Each of these dockets includes the following:

- objectives of the analogue study and organisations involved,
- geographic location of the analogue,
- description of the natural system or archaeological finding and relevant information,

16 In the case of uranium deposits, an attempt has been made to describe the history of their genesis.

17 Noteworthy progress has been made in the study of these natural systems as regards sampling and analysis methodologies. These have not been addressed since they are considered to be beyond the scope of the present summary document.

- main analogies presented with respect to the materials and processes of a disposal system,
- contribution to safety assessment and communication, and
- some of the most significant bibliographic references where the information on the analogue included in the docket may be extended upon.

The descriptive dockets of the main analogues selected are included below, classified into the following groups: uranium deposits, clay materials and hyperalkaline environments (corresponding to the division made of the geological media type analogues) and archaeological analogues.

2.1 Uranium deposits

Of the three most relevant groups of natural systems analysed, uranium deposits are the best represented in the project, with 6 main and 11 secondary analogues classified (see Table 7).

Their interest as natural analogues springs from the mechanisms responsible for their formation and for any subsequent radionuclide remobilisation. The secondary deposits and remobilised regions adjacent to the deposits are even more interesting, due to the fact that they are normally formed at temperatures more representative of the conditions of the disposal system (<100 °C) than of the conditions in which the deposit is formed.

One of the limitations of many deposits, as regards their study as analogues, is that they are normally found at relatively shallow depths where important flows of oxygenated groundwaters dominate the transport processes, this differing from the conditions of low flow and a reducing environment to be expected in a repository system. However, this type of situations has been used to increase knowledge of the evolution of disposal systems subjected to much more aggressive conditions than initially expected in which the foreseen retention capacity is lost.

An important issue to be borne in mind when characterising these systems is that deposits that have been subjected to mining activities may be disturbed, this complicating the understanding of the bounding conditions of the geochemical and hydrogeological systems.

The main characteristics of uranium deposits and the processes that take place in their surroundings that are of potential interest for study as analogues are the following:

- the composition and long-term performance of the uraninite (stability, corrosion/dissolution) as an analogue of spent fuel,
- the role of redox processes in radionuclide mobilisation and retardation, including redox fronts and other geochemical discontinuities,
- the controls of radionuclide speciation and solubility in groundwaters, including the formation and behaviour of colloids,
- the retardation processes affecting remobilised radionuclides, including the phenomena of sorption on surfaces and diffusion in the matrix,
- the possibility of using radioactive unbalance series to estimate the longevity of various mobilisation and retention processes, and
- the influence of colloids and microbial populations in radionuclide mobility.

POÇOS DE CALDAS (Brazil)

The project was carried out between 1986 and 1989 under the supervision of SKB, NAGRA, UK-DoE and US-DOE. The main objectives of the study were the geochemical characterisation of the transport of radionuclides sensitive to changes in redox conditions and the checking of thermodynamic codes and databases.

Photograph 1. Redox front (courtesy of John Smellie)

Description of the analogue

Poços de Caldas is a volcanic caldera measuring 35 km in diameter and located in the region of Minas Gerais (southern Brazil), one of the areas of highest natural radioactivity levels in the world. This caldera was formed some 75 million of years ago and the subsequent hydrothermal activity in the zone gave rise to the formation of numerous metallic accumulations, among which are: *Osamu Utsumi* open cast mine, where predominan minerals of uranium (U) with subsidiary thorium (Th) and rare earth elements (REE); and the Th and REE-rich mineralisations of *Morro do Ferro.*

The most important characteristic at *Osamu Utsumi* (Fig. 1) is the formation of a redox front arising from the remobilisation of uranium and other elements from the mineralised zone, this being associated with the appearance of uranium enrichment on either side of the front: as pitchblende in the reduce zone and associated with oxyhydroxides of iron in the oxidated zone (Photo 1).

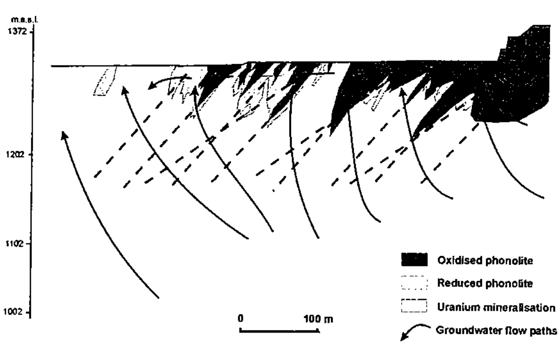


Figure 1. Schematic representation of the mineralisation environment of Osamu Utsumi and predominating flow directions (Modified from Chapman et al., 1991).

This front arose from the infiltration of oxidising stormwaters and its interaction with the terrain. The arrangement of the oxidated and reduced zones (Fig. 1) corresponds to this past situation, in which weathering penetrated preferentially via the fractures (Photo 2). At present, the hydrogeology of

Photograph 2. Redox front from a fracture (courtesy of Fred Karlsson, SKB)

Osamu Utsumi is highly influenced by the exploitation of the deposit, such that what is observed is an ascending flow of reducing waters in zones that originally were oxidising waters recharge areas. The flow rates are generally high ($2 \cdot 10^{-2}$ m/day), with advection predominating over diffusion. At *Morro de Ferro* (Fig. 2), unlike Osamu Utsami, the hydrogeological conditions have not been affected by mining activities, this having made it possible to better study water-rock interaction processes and confirm the immobility of transuranic elements under normal pH and Eh conditions.

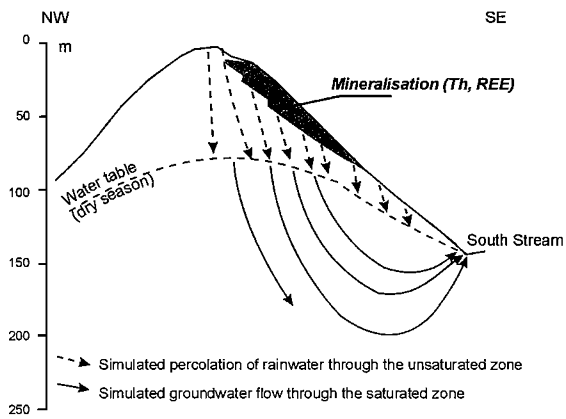


Figure 2. Schematic representation of water-rock interaction at Morro do Ferro (Modified from Chapman et al., 1991)

Analogies

The main analogies are as follows:

- The dissolution of the uraninite and pitchblende is analogous to the process of dissolution of spent fuel, including the study of lantanides (present in very high concentrations in this system) as chemical analogues of actinides.
- The retention processes of uranium and other trace elements in the iron oxyhydroxides in the oxidated area of the redox front are analogous to the possible retention processes in the corrosion products of the canister.
- The redox front developed at Osamu Utsumi is analogous to the one that might be developed in the near field of a disposal system as a result of radiolysis.
- Characterisation of the interactions between the radionuclides, colloidal phases and mineral surfaces helps to understand colloidal stability and the processes of migration of these elements in a natural system.

Contributions to safety assessment

The main contributions consist of the following:

- Support for the conceptual model of spent fuel stability under reducing conditions: the oxidising dissolution of the uraninite occurs under Eh conditions in excess of 300 mV (the values deduced in the laboratory and used in the assessment exercises are around 120 mV).
- Support for the conceptual model of canister corrosion: the radionuclides are immobilised by coprecipitation and sorption in the oxyhydroxides of iron.
- Quantitative data on the rate of advance of a redox front: 1-20 mm in 1000 years.
- Checking of different approaches to reactive transport modelling.
- Verification of thermodynamic databases under different redox conditions.

This analogue has been used in various safety assessment exercises, such as: Kristallin-1 (Switzerland), SKB-91, SITE-94 and SR 97 (Sweden), TSPA-VA (USA) and TVO-92 and TILA-99 (Finland) (see table 5 in chapter 6).

Contribution to communication

The numerous water samples collected at Morro do Ferro demonstrate that the mobility of thorium in the deposit is extremely low (two times lower than what is permitted in drinking water).

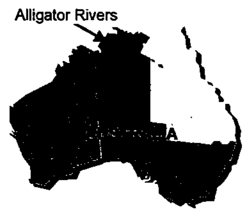
At Osamu Utsumi, there is evidence that the redox fronts that may occur in the near field of a repository system may delay radionuclide mobility and retain them.

This analogue is used in the video "Traces of the Future: Lessons from Nature for Waste Disposal", developed by different European waste management agencies (see table 6 in chapter 7).

References

Chapman, N.A., McKinley, I.G., Shea, M.E. and Smellie, J. (-1991). The Poços de Caldas project. Summary and implications for radioactive waste management. (Technical Report SKB TR 90-24), SKB, Stockholm, Sweden.

Gumtensperger, M.A., Lindquist, J. (1999). "Public Perception and natural analogues". Report EUR 17543 EN p. 543. Proceedings of the Fourth Conference of the EC on the Management and Disposal of Radioactive Waste, Luxembourg.



KOONGARRA (Australia)

The Alligator Rivers Analogue Project (ARAP) was initiated in 1987 by OECD/NEA, ANSTO, JAERI, PNC, SKI, UK-DoE and USNRC and concluded in 1992.
The fundamental objective of the Project was to contribute to the development of radionuclide transport models.

Description of the analogue

The study focused on the uranium deposit of Koongarra, located at the geosyncline of Pine Creek in northern Australia, in the region of Alligator Rivers. This deposit was formed around 1,800 million years ago as a result of important hydrothermal activity. Although this area includes various mineralised bodies rich in uranium (up to 1% richness), only the Koongarra deposit (Fig. 1) was considered in the ARAP project since this mineralisation is the one that has undergone the greatest alteration and weathering over the last 1-3 million years.

The greatest part of the groundwater flow takes place in this zone through the partially weathered schists of the Cahill formation. Recharge to this aquifer occurs from the Kombolgie formation via the Koongarra fault and by direct infiltration of rainwater. This last component of the recharge produces dilution and gives rise to vertical stratification of the hydrochemical characteristics in the Cahill formation. Given the intense weathering, the uranium and other elements initially contained in the mineralised veins (in the form of uraninite and pitchblende) have undergone processes of dissolution and subsequent precipitation, passing into the groundwaters and being retained by different processes. These processes have given rise to the appearance of different zones of secondary mineralisation and of a uranium (U) dispersion halo (of some 80 m from the mineralised body southwards), where various zones may be differentiated on the basis of their mineralogy (Fig. 1):

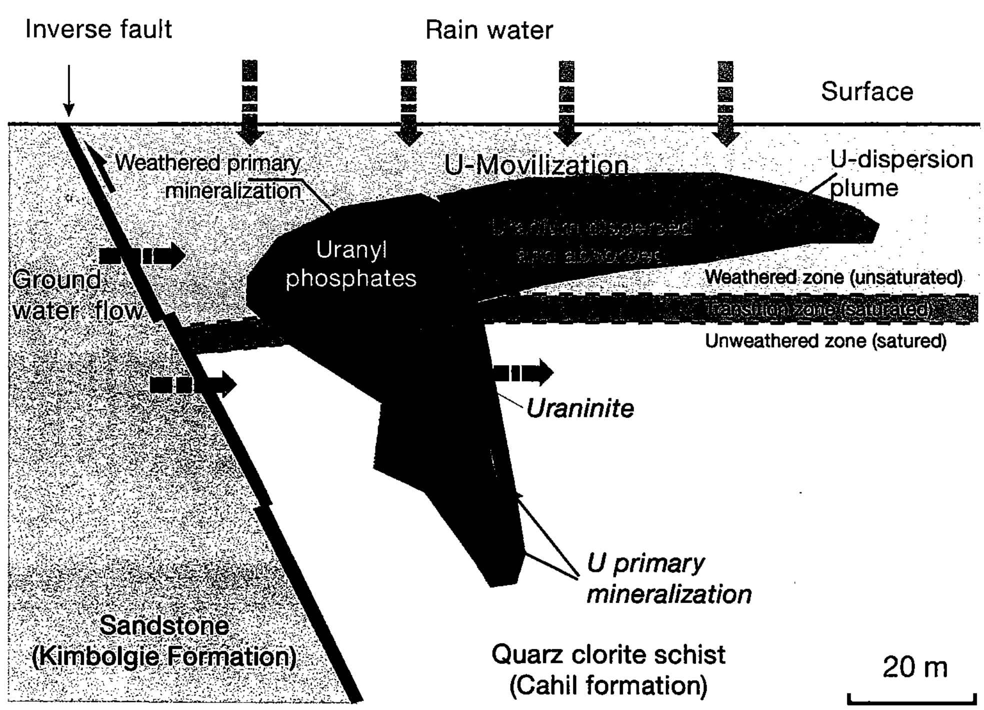


Figure 1. Morphology of the Koongarra mineralisation. The diagram shows the position of the different weathering zones in depth and the uranium minerals characterising each sector of the mineralisation. Also indicated are the groundwater flow paths.

- Secondary uranium silicates due to in situ alteration of the primary minerals.
- Secondary phosphates of uranyl currently washed by the groundwaters.
- Disperse uranium associated (by sorption and coprecipitation) with clays and oxyhydroxides of iron.

The characteristics of each of the zones, the distribution of uranium in them and the processes through which they have originated are the most widely studied issues in this system.

Analogies

The effect produced by processes of uraninite leaching under oxidising conditions, radionuclide migration and retention and the movement of weathering fronts are excellent analogues of the behaviour to be expected of a high level waste disposal facility in an advanced phase of degradation under normal conditions or in the event of loss of retention and sealing capacity.

Contributions to safety assessment

The main contributions are as follows:

- Support for the conceptual model of spent fuel stability under reducing conditions.
- Support for the conceptual model of processes of immobilisation by precipitation, coprecipitation and sorption of radionuclides on secondary minerals (phosphates, clay minerals and oxyhydroxides of iron).
- Development of a conceptual sorption model.
- Confirmation of the low degree of relevance of matrix diffusion.
- Confirmation of the importance of the alpha recoil process.
- Quantitative data on the migration rate of U: 25-100 mm in 1000 years.
- Verification of the methodology for scenario generation developed by SKI.
- Contrasting of flow, geochemical, reactive transport and sorption models and of thermodynamic databases.

It was used in the Swedish SR 97 exercise (SKB, 1999) to help define canister failure scenario (see table 5 in chapter 6).

Contribution to communication

This analogue might be used to illustrate the capacity of the near field and geosphere of a repository system to retain and delay radionuclides, even in the event of possible engineering barrier or system sealing failure.

References

Duerden, P., Lever, D.A., Sverjensky, D.A. and Townley, L.R. (1992). *Alligator Rivers Analogue Project Final Report, Vol. 1: Summary of findings.* (Final Report DOE HMIP RR 92/071), DOE-UK, London, UK, 138 p.

NEA (1994). ARAP. *The international Alligator rivers analogue project. Background and results.* OECD Nuclear Energy Agency, Paris, France, 77 p.

SKI (1996). *SKI SITE-94. Deep repository performance assessment project. 2 vols.* (Technical Report SKI R 96/36), SKI, Stockholm, Sweden, 305-660 p.

CIGAR LAKE (Canada)

The project was carried out between 1984 and 1993 as a result of cooperation between AECL, COGEMA and CLMC. SKB joined the study in 1989, followed later by the US-DOE through LANL. In 1995, SKB financed a review of the data obtained during the project with a view to taking advantage of this information in the light of the results of other analogues and of models not originally used (Cigar Lake is the only major analogue whose data have been re-assessed in a subsequent project).

The objectives were the evaluation of the stability of uraninite, the evaluation of thermodynamic codes and databases, the study of transport through bentonite and the role of colloids and organic compounds in radionuclide transport.

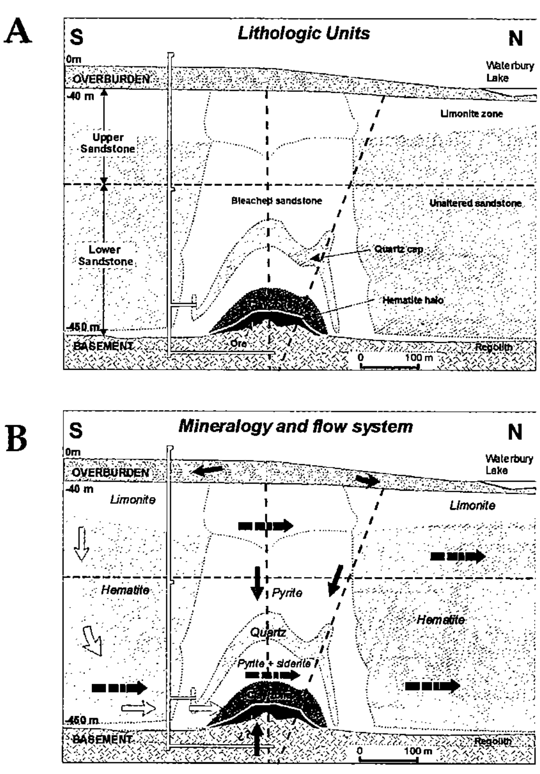


Figure 1. Geological section: A) Lithologies and halos of hydrothermal alteration associated with the origins of the deposit and B) model of hydrogeochemical evolution (Modified from Cramer and Smellie, 1994)

Description of the analogue

The Cigar Lake uranium deposit is located in the Athabasca basin in the province of Saskatchewan, Canada. It was formed some 1,300 million years ago by the activity of hydrothermal fluids rich in uranium at 150-200ºC, and there are no traces on the surface of its presence at a depth of 450 m. It is a deposit of uraninite and pitchblende (with an average uranium richness of 14%) surrounded by a clay halo and installed in an active hydrological system with highly diluted waters that lose their initial oxidising characteristic due to reaction with the host arenites, becoming of reducing nature in the surroundings of the deposit (Figure 1).

The results of hydrogeological characterisation and of the flow models indicate the existence of a shallow local flow system, another intermediate system and a semi-regional deep flow system (Figure 2). The spatial variations of the topography control the flow to a depth of 200 m. From this depth the flow is quasi-horizontal with a gradient of approximately 0.3%.

The studies performed in this system have focused on characterisation of the processes of genesis of the deposit and on the processes that have given rise to the current distribution of radionuclides, taking into account the interaction with groundwaters and all the processes that influence the transport and retention that might have affected them throughout their geological history (fundamentally radiolysis, microbial activity, colloidal transport and isolation of the clay halo).

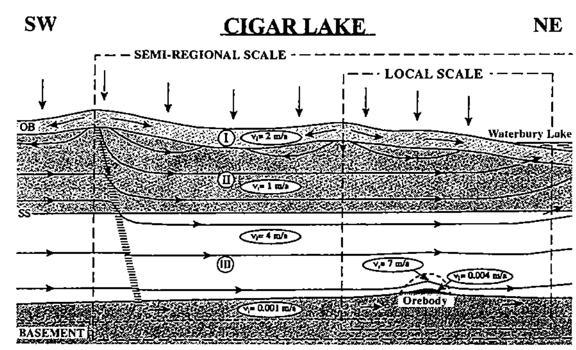


Figure 2. Schematic representation of flow at local, intermediate and semi-regional scale, with an indication of water flow rates (Modified from Winberg and Stevenson, 1994)

Analogies

Cigar Lake represents a large-scale analogy of the high level waste disposal concepts in crystalline formations (Fig. 3). It is located at a depth of 450 m (similar to the depths considered in repositories), surrounded by a clay halo (bentonite barrier) embedded in sandy materials with an active circulation of groundwaters (geosphere) and without any trace on the surface of its presence for the last 1,00 million years (long-term stability of the disposal concept).

Contributions to safety assessment

The main contributions are as follows:

- Support for the conceptual model of spent fuel stability under reducing conditions: the oxidising dissolution of uraninite occurs under Eh conditions in excess of 200 mV (the values deduced in the laboratory and used in disposal safety assessments are around 120 mV, a more conservative value).
- Support for the conceptual model of hydraulic isolation and filtering of colloids by the bentonite, represented by the clay halo.
- Development of more highly elaborated radiolytic models.
- Support for the conceptual model of irreversible nuclide sorption on colloids.
- Quantitative data on the solubility of trace elements.
- Quantitative data on uraninite dissolution rate: 10^{-8}-10^{-9} $year^{-1}$.

The information provided by this analogue has been used for the aforementioned purposes in numerous disposal safety assessments: AECL-94 (Canada); Kristallin-1 (Switzerland); Project-90, SKB-91, SITE-94 and SR 97 (Sweden); TSPA-VA (EE.UU); TVO-92 and TILA-99 (Finland) (see table 5, chapter 6).

Contribution to communication

Cigar Lake is a clear and illustrative example of a disposal system generated by nature itself and of the system's isolation capacity. Therefore, this is one of the analogues most widely used for communication to a wide range of audiences. It has been the subject of the video "Nature's own repository", published by SKB, and is mentioned in the video "Traces of the future", in brochures, websites and the SKB exhibition on board the ship Sygn (see Table 6, chapter 7).

References

Cramer, J.J. and Smellie, J.A.T. (Ed.) (1994). *Final Report of the AECL/SKB Cigar Lake Analog Study.* (Tech. Rep SKB TR 94-04), AECL, Pinawa, Manitoba, Canada.

Smellie. J.A.T. and Karlsson, F. (1996). *A reappraisal of some Cigar Lake issues of importance to performance assessment.* (Technical Report SKB 96-08), SKB, Stockholm, Sweden, 93 p.

Goodwin, B.W., McConnell, D.B., Andres, T.H., Hajas, W.C., LeNeveu, D.M., Melnyk, T.W., Sherman, G.R., Stephens, M.E., Szekely, J.G., Bera, P.C., Cosgrove, C.M., Dougan, K.D., Keeling, S.B., Kitson, C.I., Kummen, B.C., Oliver, S.E., Witzke, K., Wojciechow et al. (1994). *The disposal of Canada's nuclear fuel waste: postclosure assessment of a reference system.* (Technical Report AECL-10717), AECL, Pinaw7, Manitoba, Canada, 684 p.

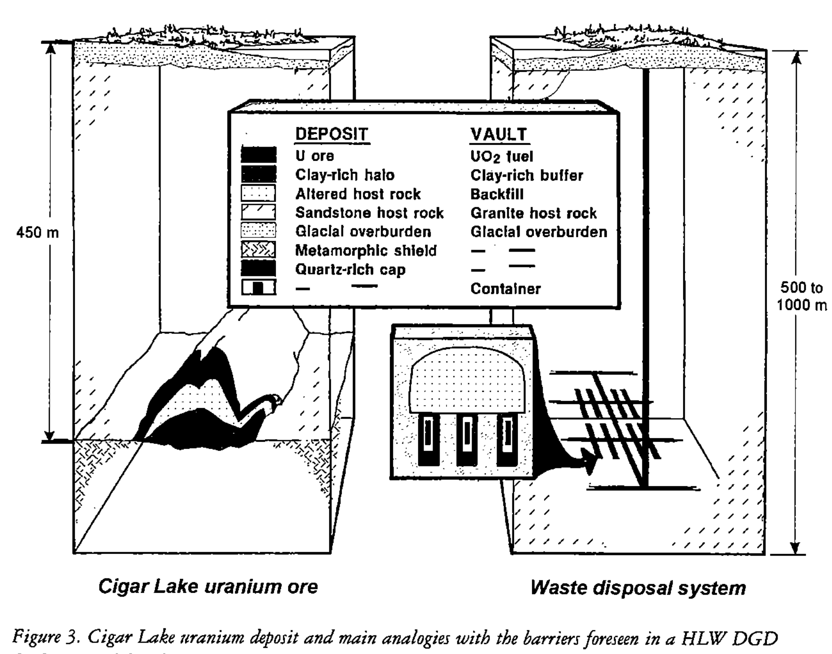


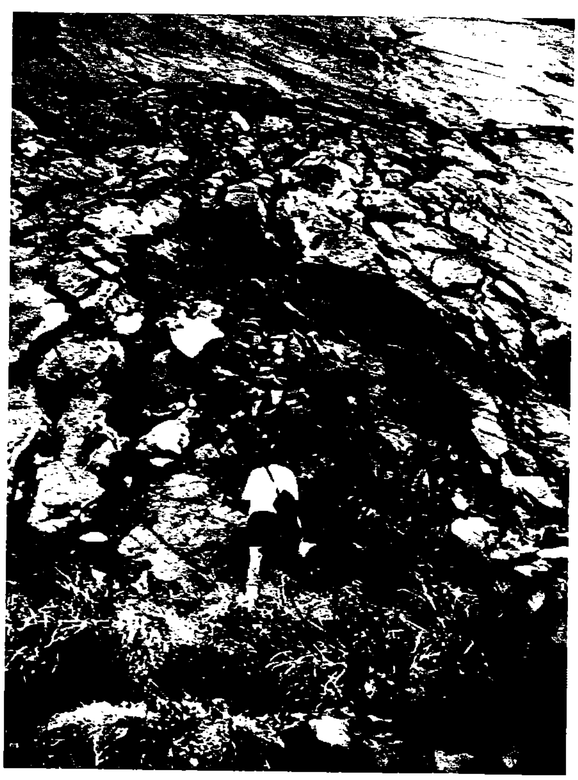
Figure 3. Cigar Lake uranium deposit and main analogies with the barriers foreseen in a HLW DGD facility. (Modified from Goodwin et al., 1989)

OKLO (Gabon)

The project known as "Oklo as a natural analogue" began in 1989, financed by CEC, CEA, SKB, ANDRA and Enresa, and ended in 1999.
The main objectives were the study of processes of uraninite alteration and the radionuclides transport and retardation in the very long term.

Description of tfihe analogue

The most distinctive characteristic of the Oklo uranium deposits (south east of Gabon) is the fact that they were natural fission reactors (photo 1) in which nuclear chain reactions took place spontaneously 2,000 million years ago. This makes Oklo the only place on Earth in which a ratio of $^{235}U/^{238}U$ uranium isotopes of 0.0038 has been encountered, this deviating significantly from the average value of 0.0072.



Photograph 1. Outcropping of a nuclear reactor in Gabon. (courtesy of L.P. del Villar)

The natural reactors of Oklo are located in the sedimentary basin of Franceville. 15 reactors have been discovered, all located in the Oklo/Okélobondo deposit to a depth of 400 m (Fig. 1), with the exception of one discovered at Bangombé at a depth of 11 metres (Fig. 2).
In all cases the reactors are in the zone of contact between the arenites formation (through which oxidising fluids enriched in uranium flowed) and the organic matter-rich lutites formation, where the uranium was accumulated in the form of uraninite as a result of the reducing conditions in the zone.

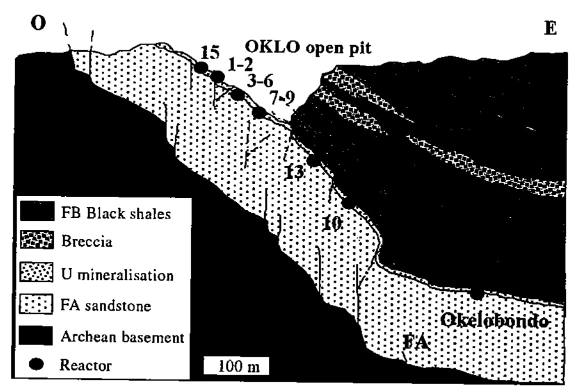


Figure 1. Idealised section of the Oklo/Okélobondo uranium deposit. Some of the reactors have been represented at the depths at which they are located. (Modified from Gauthier - Lafaye., 1996)

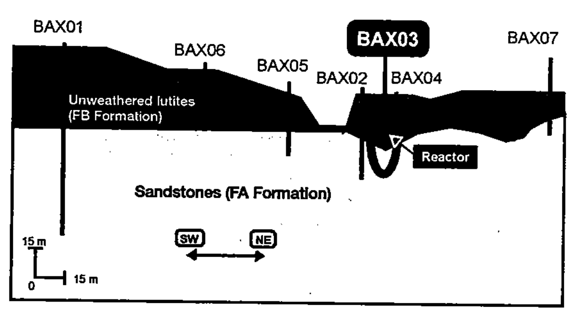


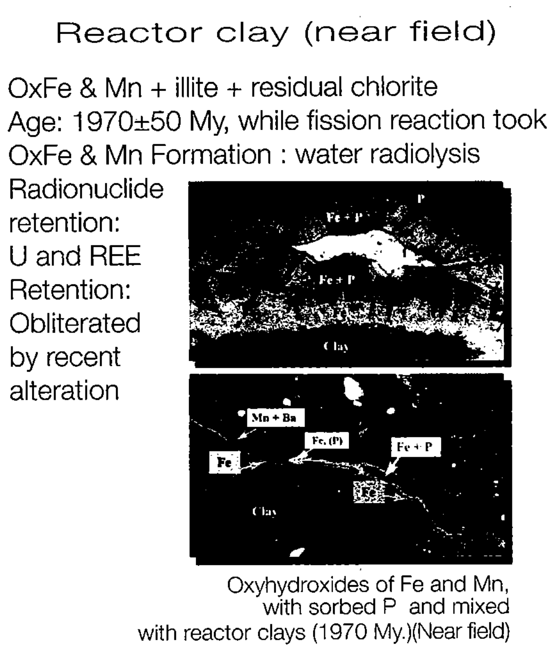
Figure 2. Idealised section of the Bangombé deposit showing the different boreholes drilled for its study (BAX01 to BAX07). (Modified from Duro et al., 2000)

The reactors are zoned bodies characterised by two main facies: (a) the reactor core, with a content of uranium of up to 87%, where the fission reaction took place (this is made up of uraninite grains embedded in a clay matrix); and (b) the reactor clays, which surround the core and are formed by hydrothermal clays, fundamentally chlorite and illite (Photo 2).
The different environments in which the reactors are located have made it possible to study and model their stability and the distribution of the radionuclides under different conditions and from different points of view.

Analogies and contributions to safety assessment

The processes of alteration that occurred in these deposits make Oklo a unique analogue of the long-term behaviour of fission and activation products in a HLW DGD system.
Its main contribution to safety assessment is as follows:

- Support for the conceptual model of spent fuel stability under reducing conditions: more than 90% of the uraninite in the reactors has remained stable for 2,000 million years.
- Support for the conceptual model of the isolation and retention capacity of the clay barrier.



Photograph 2. Zoning of the clays in the reactor (courtesy of L.P. del Villar)

- Checking of radionuclide retention capacity in oxyhydroxides of iron, phosphates and graphite and of the low degree of influence of radiolysis (due to buffering by organic matter).
- Increased knowledge of a scenario of criticality.
- Confirmation of the control of redox conditions in the reactors due to radiolysis of the water and the presence of organic matter.

It has been used in exercises such as AECL-94 (Canada), SR 97 (Sweden) and TSPA-VA (USA) (see table 5 in chapter 6).

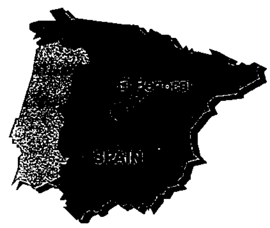
Contribution to communication

This analogue provides some of the best evidence that the products of nuclear reactions may be retained in natural systems, and is one of the analogues with the greatest potential for public information. It is the subject of the video "Nature's own nuclear waste" by SKB and is explained in the video "Traces of the future", in brochures and websites and in the SKB exhibition on board the Sygn (see table 6, chapter 7).

References

Gauthier-Lafaye, F., Ledoux, E., Smellie, J., Louvat, D., Michaud, V., Pérez del Villar, Oversby, V. and Bruno, J. (Ed.) (2000). *Final Report:* OKLO – *Natural Analogue Phase II. Behaviour of nuclear reaction products in a natural environment (The Oklo site, Gabon, Western Africa).* (Final Report EUR-X), EC-NST, Luxembourg.

The "El Berrocal project" began in 1988 under the supervision of ENRESA and CIEMAT and with the participation of CIMNE, CEA/IPSN, AEA Technology, BGS, INTERA and JRC-CEC. The project finished in 1995.



EL BERROCAL (Spain)

The main objective of the project was to gain understanding of natural radionuclide transport processes in a fractured granite environment.

Description of the analogue

El Berrocal is located at a distance of 92 Km to the south of Madrid, in the south-eastern part of the Sierra de Gredos hills (Central System). It is a granitic pluton of sub-rounded morphology with an outcropping surface area of some 22 km^2, enriched in uranium and thorium. The emplacement of the pluton is estimated to have occurred some 297 million years ago.

The geological environment of El Berrocal is characterised by intensive fracturing, which favoured hydrothermal activity in the areas close to the fractures (at least 1 million years ago). As a result of this activity there was remobilisation of uranium, thorium and other elements, initially contained in the pluton in the form of disperse uraninite, and their subsequent reprecipitation in the main mineralisation (*mineralised dyke of quartz and uranium*, Fig. 1) and in the form of secondary mineralised veins.

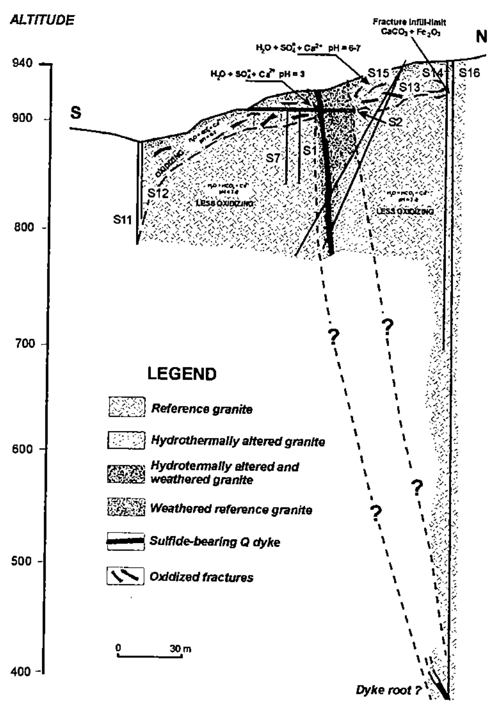


Figure 1. Schematic lithological and geochemical representation of the surroundings of El Berrocal. Also shown are the boreholes drilled for the study (S1 to S16). (Modified from Pérez del Villar et al., 1993)

The subsequent weathering processes remobilised the uranium (U), thorium (Th) and other elements, which were subsequently retained (by sorption or precipitation) in the minerals filling the fractures (silicates, phosphates, silicophosphates of U associated with oxyhydroxides of iron, carbonates and clay minerals).

The hydrogeological system is strongly conditioned by the discontinuities present (fractures and dykes of quartz and uranium), which constitute the preferential ways for groundwater flow.

The studies carried out address the characterisation and modelling of the water-rock interaction system, taking into account the different lithologies, the different physiochemical and water flow environments and the varied solid forms in which the uranium is found.

Analogies

The main analogies encountered in this system are the presence of uraninite and pitchblende (as analogues of the spent nuclear fuel) subjected to different alteration processes and the effects of these processes on radionuclide mobilisation and retention in the geosphere, both in the fracture filling materials and as a result of diffusion in the matrix.

Contributions to safety assessment

The main contributions to safety assessment made by El Berrocal are as follows:

- Support for the conceptual model of spent fuel stability.
- Support for the conceptual model of matrix diffusion and retention in the geosphere, fundamentally by means of processes of precipitation, coprecipitation and sorption on fracture filling minerals.
- Development of models of radionuclide transport favoured by fluorated and carbonated complexes and microbial activity.

- Checking of three-dimensional hydrogeological modelling assuming the mixed approach of fractures embedded in an equivalent porous medium.
- Quantitative data on penetration depth by matrix diffusion: 35-80 mm.
- Quantitative data on the solubility limits of elements of interest for safety assessment.

The data provided by this analogue have been used in safety assessment exercises such as SITE-94 and SR 97 (Sweden) (see table 5, chapter 6).

Contribution to communication

At El Berrocal the mobility of uranium has been restricted to the areas close to the fractures and no uranium is observed outside the granitic pluton. This fact might be used to increase public confidence in the repository concept in granite. This analogue has been the subject of a video published by Enresa (see table 6, chapter 7).

References

Enresa (1996). El Berrocal Project. Characterization and validation of natural radionuclide migration processes under real conditions on the fissured granitic environment. Volumes I-IV. (Topical Reports PTNP 01-04/96), Enresa, Madrid, Spain.

PALMOTTU (Finland)

The Palmottu analogue project began in 1988 as a result of cooperation between CEC, GTK, STUK, SKB, Enresa and BRGM and was completed in 1999.
The main objective of the project was the study of radionuclide transport in fractured crystalline massifs.

Description of the analogue

The Palmottu uranium and thorium mineralisation is located in the southwest of Finland and is associated with crystalline rocks: gneisses and granites. This mineralisation was formed as a result of hydrothermal activity 1,700-1,800 million years ago. It is calculated to contain a million tons of ore with an average richness of uranium of 0.1%, and forms a vertical structure parallel to the granitic dykes, which extend to depths of some 400 m (Fig. 1).
During recent geological history the area has been subjected to glaciations, the last finishing some 10,000 years ago. These stages alternated with interglacial periods, favouring the ingress of oxidising waters from ice melting towards deep zones, through the fracture systems.

Interaction between the rock and the groundwaters has given rise to a series of characteristic hydrochemical subsystems (Fig. 2) that have conditioned the mobilisation of uranium and thorium. These elements, initially contained in the mineralisation, have migrated towards the fractured zones, close to which they re-precipitate as secondary minerals. The analysis of this behaviour and of its effects has been the main objective in studying this system. A distinction may be made between three flow systems (Fig. 2):

1) Superficial system, located above the subhorizontal fracture. This is the most permeable zone, characterised by young waters containing calcium bicarbonates.
2) Deep system, located below the previous system. Waters containing sodium bicarbonates and residence times ranging from centuries to a few thousand years.
3) Deep quasi-stagnant system. Waters with very low flow rates and a sodium sulphated or sodium chlorided chemical composition. The values of ^{18}O suggest that these waters may have recharged during glacial periods 10^5 years ago.

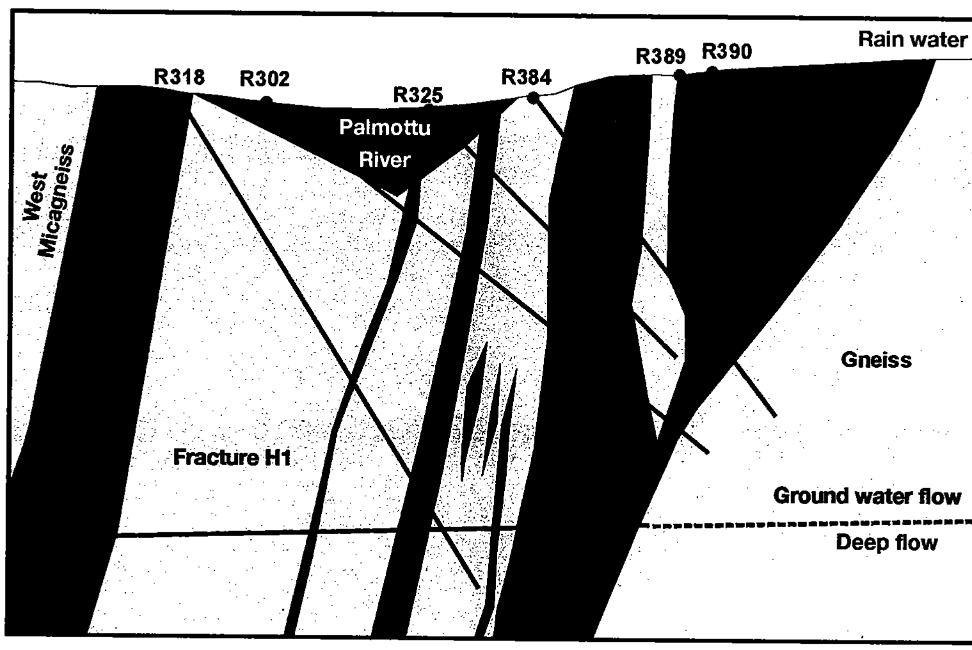


Figure 1. Schematic section of the different lithologies present in the Palmottu system. (Modified from Blomquist et al., 2000)

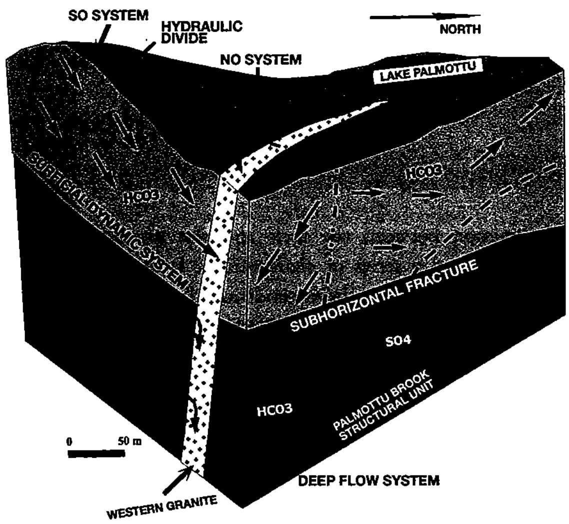


Figure 2. Schematic representation of the granite environment of E and predominating flow systems. (Modified from Blomquist and Ruskeeniemi, 1997)

Analogies

The main analogies found in this system are the presence of uraninite (as an analogue of spent fuel) subjected to different alteration processes and the effects of these processes on radionuclide mobilisation and retention in a crystalline rock similar to those considered probable in certain disposal system concepts.

Contributions to safety assessment

The main contributions made by this study to safety assessment are as follows:

- Support for the conceptual model of spent fuel stability thanks to the model suggested for the dissolution of uraninite and to the confirmation of Eh control by the uranium system itself.
- Confirmation of the capacity of the rock to maintain reducing conditions despite the ingress of oxidising flows.
- Support for the conceptual model of radionuclide retention in the geosphere, fundamentally through processes of precipitation, coprecipitation and sorption on the minerals filling the fractures.
- Quantitative data on penetration depth by matrix diffusion: 25 mm.
- Qualitative data and study methodologies for climate scenarios (palaeohydrogeological aspects relating to glaciations).

The information on this analogue has been used in the following safety assessment exercises: SR 97 (Sweden) and TVO-92 and TILA-99 (Finland) (see table 5, chapter 6).

Contribution to communication

This analogue provides evidence of the stability of spent fuel, for which reason it may be used to increase public confidence in the disposal concept.

References

Blomqvist, R et al. (Ed.) (2000). *The Palmottu natural analogue project. Phase II: transport of radionuclides in a natural flow system at Palmottu.* (Final Report EUR-X), EC-NST, Luxembourg, Luxembourg, 171 pp.

Vieno, T., Hautojärvi, A., Raiko, H., Ahonen, L. and Salo, J.P. (1992). TVO-92 *safety analysis of spent fuel disposal.* (Report YJT 94-21), YJT, Helsinki, Finland.

2.2. Clay materials

As regards the characterisation of the long-term stability of bentonite, it is considered that the analogues of these clay materials may provide relevant information on five aspects of interest in safety assessment (Miller et al. 2000):

- the longevity and alteration of bentonites,
- their functions as a hydraulic barrier and colloids filter,
- physiochemical changes in bentonites relating to the heating induced by the canister (smectite-illite transformation),
- collapsing of the canister, and
- interactions with other materials in the engineered barriers.

Of all the subjects of interest pinpointed, the longevity and degradation rate of the bentonite (transformation of smectite into illite) is the one that has been most profusely analysed both in analogues and on the basis of data from scientific studies on natural systems of these lithologies.

However, for some of the issues above mentioned the data provided up to now by natural analogues are not completely reliable, scarce or do not yet exist, as occurs, in the cases of collapsing of the canister or interaction with other engineered barrier materials.

As regards the physiochemical changes due to heating, analogue studies have focused on analysis of the processes of cementation and, in general, on the mineralogical changes due to the thermal effects of magmatic intrusions and their repercussions on the properties (degradation) of the barrier. However, the results obtained from the majority of these analogues (such as, for example, the Orciatic analogue in Italy; Benvegnú et al., 1988), do not appear to be particularly suitable since the analysis has centred on the materials closest to the intrusion, subjected to temperatures far higher than those that would be expected in the disposal system.

The growing emphasis on the study of coupled thermohydromechanical or thermohydrochemical processes and their long-term effects on the bentonite barrier has increased interest of the study of this type of analogues, in which the checking of codes capable of modelling this type of processes may be addressed (Gera et al., 1996; Pellegrini et al., 1999).

Outstanding among the clay material analogues studied (see table 7) is the one at Kinnekulle, one of the analogues studied most completely and for the longest time. The research currently covers the complex coupled thermohydromechanical processes that condition the long-term stability of the bentonite barrier. In addition, Dunarobba Forest Analogue has been described, as an unique example of tree trunk preservation for million years due to the isolation capacity of the clay where they were buried.

KINNEKULLE (Sweden)

Since the 1980's there have been a large number of works performed on the Kinnekulle bentonites, both from the scientific point of view and as an analogue of the longevity of the clay materials in the engineered barrier (the latter carried out fundamentally as part of the activities of SKB). This has favoured the use and re-evaluation of the results obtained at Kinnekulle, a task that continues at present.

The objective of the SKB study of this analogue has been to analyse the processes that give rise to cementation of smectite affected by heat.

Description of the analogue

450 million years ago (Ordovician, Fig.1) volcanic ash was deposited on a series of clay and calcareous sediments at Kinnekulle. The levels of ash were covered by materials similar to these and were left sandwiched between a series of marine sediments measuring several hundred metres in thickness. These ashes became transformed into bentonitic and smectitic layers and were consolidated under a pressure of 5-10 MPa, reaching a density of 2.0-2.2 g/cm^3 and a porosity of 30-40% with a water content of 15-25%.

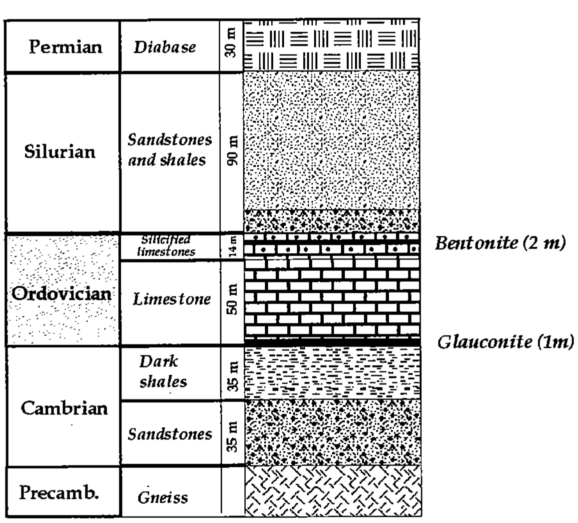


Figure 1. Geological column of Kinnekulle. The main bentonitic layer appears above the Ordovician limestones (Modified from Pusch and Madsen, 1995)

300 million years ago (Permian) the series was penetrated laterally by a magmatic intrusion that generated a diabasic dyke measuring several tens of metres in thickness, which was emplaced some 95 metres above the main bentonitic level (some 2 m in thickness). The thermal surge associated with this intrusion affected the surrounding sediments and collaborated in their transformation into consolidated sedimentary rocks.

The estimates performed using indirect methods of the temperatures to which the bentonitic layer was subjected indicate that it did not exceed 160 °C. Another aspect of interest studied was the degree of alteration suffered by the bentonites as a result of heating, both transformation of smectite into illite (greater at the edges than in the centre) and cementation by silica. These studies were completed with exercises modelling the process of degradation along with sensitivity analysis of the parameters influencing this degradation.

Analogies

The Kinnekulle bentonites are a good analogy of the clay barrier and its performance as regards the following aspects of interest in the safety assessment of this barrier: longevity and physiochemical changes due to heating.

Contributions to safety assessment

The main contributions made by this analogue to safety assessment are as follows:

- Support for the conceptual model of clay barrier performance: despite the thermal effect on these bentonites, the degradation suffered has not caused any alteration of its properties as a barrier to radionuclide release.
- Identification through sensitivity analysis of the parameters intervening in the process of transformation: temperature, concentration of potassium and activation energy.
- Development of codes for the modelling of the smectite-illite transformation process, and checking of the applicability of coupled codes.

The information from this analogue has been used explicitly in the exercises AECL-94 (Canada) and SR 97 (Sweden) (seetable 5, chapter 6).

Contribution to communication

The Kinnekulle analogue shows the longevity and the maintenance of the desired properties of the clay barrier. For this reason, it might be used to increase public confidence in the safety of geological disposal systems (see table 6, chapter 7).

References

Pusch, R., Takahase, H. and Benbow, S. (1998). *Chemical processes causing cementation in heat-affected smectite. The Kinnekulle bentonite.* (Technical Report SKB TR 98-25), SKB, Stockholm, Sweden.

SKB (1999b). SR 97. *Processes in the repository evolution. Background report to SR 97.* (Technical Report SKB TR 99-07), SKB, Stockholm, Sweden.

DUNAROBBA (Italy)

The general study of this analogue began in 1992, within the framework of the MIRAGE Project (CCE). The results were published in 1997.
The objectives of the project centred on analysis of the isolating capacity of clay materials.

Description of the analogue

The Fossil Forest of Dunarobba, located near Todi (Central Italy) consists of 50 trunks of fossilised trees (dating back some 2 million years) still in their original position. The most outstanding characteristic of these fossil trees, what made them different from others existing in the geological register, is that they are still constituted by wood.
The Dunarobba forest developed some 2 million years ago in a marshland system beside a rather shallow lake that filled with fundamentally lutitic materials: clays and silts laminated with frequent bands of sands and gravel and abundant organic matter (Photos 1 and 2). Given the high rate of sedimentation (3m/1,000 years), the forest was buried and rapidly isolated from the external oxidising medium.

Above the level at which the forest is located, the series becomes more homogeneous and made up almost completely by laminated silty clays typical of deep marshland environments, these being much less permeable than the underlying clays. In fact, the exceptional degree of preservation of the trunks is due to the isolation generated by these materials, which acted as a geological barrier to processes of organic degradation.
The area of Dunarobba has been affected by active neotectonic processes and the maps of gas distribution in the soil also indicate that it is located on one of the preferential routes for fluid migration in the basin. These processes would not initially favour the preservation of a fossil forest.
At basin scale, the lutitic refill does not behave as an effective barrier against the migration of fluids due to the short-circuit caused by the fracture systems. However, at a smaller scale, these systems are the boundary for blocks that behave coherently as a barrier due to their low hydraulic conductivity (Fig. 1). The forest is located in one of these blocks, which would explain its preservation in an area in which there is an important system of discontinuities in the materials.

Photograph 1.Panoramic view of the Dunarobba forest. (Miller et al., 1994)

Photograph 2. Detail of one of the tree trunks. (Miller et al., 1994)

Analogies and contribution to safety assessment

The Fossil Forest of Dunarobba is considered to be an analogue of the long-term isolating capacity of clay materials, since these have isolated and preserved organic matter over a timescale of millions of years.

Its main contribution to safety assessment is its support for the conceptual model of the performance of bentonite as a hydraulic barrier and as a barrier limiting microbial degradation: conservation of wood in an area not favourable to it. Reference is made to this analogue in the exercise SR 97 (Sweden) (see table 5, chapter 6).

Contribution to communication

The extraordinary degree of conservation of the forest has its greatest potential in non-technical audiences communication to illustrate the isolating capacity of bentonite. Information on its existence has been broadcast in widely varying media, both modern (journalistic) and historic (there are references to it since the 17th century).

It is one of the most widely used analogues in public communication: it is quoted in the video "Traces of the Future", in brochures and websites such as those of Nagra and SKB, and in the itinerant SKB exhibition on board the Sygn ship (see table 6, capter 7).

References

Valentini, G., Lombardi, S., Bozzano, F. and Scarascia Mugnozza, G. (Ed.) (1997). ***Analysis of the geoenvironmental conditions as morphological evolution factors of the sand-clay series of the Tiber valley and Dunarobba forest preservation.*** (Report EUR 17479), EC-NST, Luxembourg.

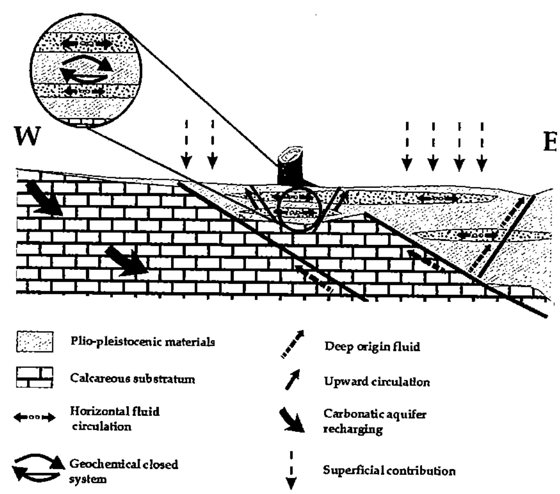


Figure 1. Conceptual model of hydrogeological performance in the Dunarobba basin.

2.3. Hyperalkaline environments

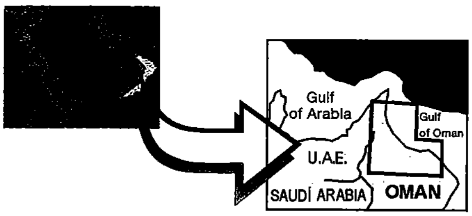
Hyperalkaline environments are natural occurrences of secondary minerals analogous to those formed during the hydration of Portland cement and give rise to interstitial waters characterised by a very high pH. The study of this type of natural systems may be of use in the following aspects of interest for the safety of a disposal facility:

- the longevity of cement and its binding properties,
- the permeability of cement to water and gas,
- the speciation and solubility of radionuclides under high pH conditions,
- the interaction of high pH fluids with the surrounding rock, as an analogy of interstitial waters migrating from the disposal system to the host rock,
- the nature and viability of microbiologically-induced geochemical processes and their influence on the process of waste degradation,
- the nature and stability of colloid species formed in high pH waters and at the interface between these and neutral waters.

One of the best natural examples of hyperalkaline environments is the one that may be observed at Maqarin. Here, low temperature reactions with the mineral phases of the natural environment (hydration, carbonatation and sulphatisation) have created a set of alteration minerals, many of which are common to those existing in cement paste (portlandite, ettringite and calcium silicate hydrates, briefly CSH).

Another example is constituted by the hyperalkaline upwellings of Oman, whose hydrochemical characteristics and certain associated mineral precipitates (portlandite) are very similar to the conditions that it is assumed will exist in a disposal facility containing cement.

Unlike Oman, the area of Maqarin presents rocks enriched in a series of elements analogous to those of relevance in a disposal system radionuclide inventory (isotopes of uranim, strontium, barium, selenium, molibdenium, rare earth elements, cobaltum, tin, copper, zinc and lead). Furthermore, in this area the process of alteration continues to be active at present, and the hyperalkaline sources and other discharges of groundwaters associated with this alteration offer a unique opportunity for the study of: (a) the mobility of analogous trace elements in alkaline water systems; (b) evaluation of the long-term effects of cement hydration and of the stability of cement phases; (c) investigation of the interaction between a hyperalkaline plume and the host rock and (d) analysis of the geomicrobiology of the system of hyperalkaline waters.



OMAN

The Oman analogue project was undertaken jointly by UK Nirex Ltd. and NAGRA and was completed in 1987.
The interest of this analogue focused on the study of hyperalkaline waters and the concentration and speciation of trace elements in solution.

Description of the analogue

The study centred on five of the hyperalkaline upwellings or surges found near Muscat, in north-eastern Oman. The examples of upwelling studied (identified by name on Figure 1) appear in a mountain chain whose structure runs parallel to the coast of Oman and which is formed by igneous materials belonging to the upper seam (mainly harzburgites) and the oceanic crust (ofiolytes).
During the lifting (obduction) of these materials, surface waters penetrated them and subsequently, on moving downwards and interacting with the ultramafic minerals of the rock, became transformed into hyperalkaline and frequently very reducing groundwaters (pH value of around 11), which could later well up and return to the surface (the location of the five examples studied is shown in Figure 1).

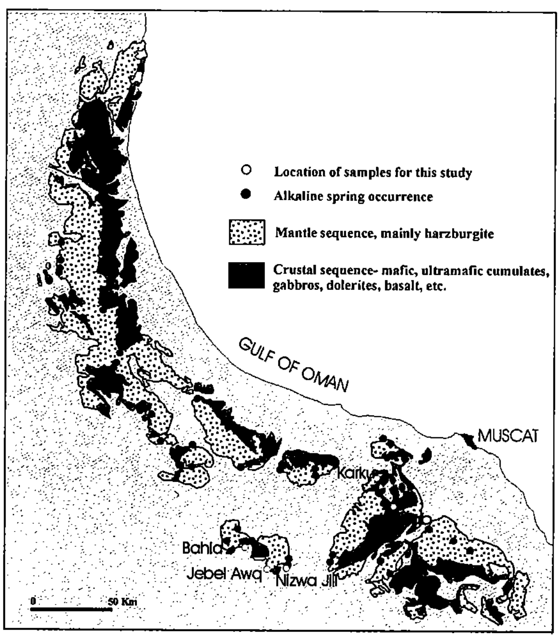


Figure 1. Schematic representation of the geology and location of the upwellings studied in Oman. (Modified from Bath et al., 1987)

At two of these upwellings studies were performed on the microbial populations present in the waters. The results obtained show a wide variety (albeit in low concentrations) of microorganisms adapted to these extreme conditions.
The geochemical modelling calculations carried out in the waters of Oman focused almost exclusively on study of the validity and applicability of the geochemical codes and thermodynamic databases used in safety assessment. These were valid for other types of conditions but had not been checked in systems having characteristics as extreme as these (hyperalkaline and reducing).

Analogies

Many of the disposal concepts currently considered include large volumes of cement that determine the presence of very alkaline waters in the near field (pH of 13 or higher). The Oman waters are very similar to these, since they are strongly alkaline and reducing. For this reason, Oman constitutes an analogy for the study of such hyperalkaline waters and of their effect on the process of radionuclide transport in the disposal system.

Contributions to safety assessment

The main contributions made by this analogue to safety assessment are as follows:

- Support for models of radionuclide release and migration in a geochemical environment difficult to simulate in the laboratory.
- Checking of the adaptation of microbial communities to extreme conditions: the limiting factor is the supply of nutrients, as normally considered in safety assessments.
- Confirmation of the low degree of influence of colloids, in spite of their abundance, due to their instability under these conditions.
- Verification of the inapplicability of the redox control model in extreme conditions.
- Support for methodological development in the definition of climate scenarios through the determination of climate variations based on isotopic study of hyperalkaline upswelling precipitates.

- Checking of the applicability of geochemical and reactive transport codes and thermodynamic databases under extreme conditions.

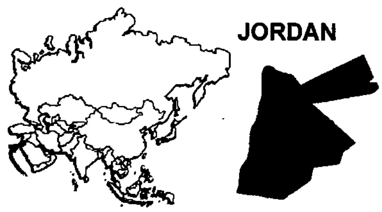
Information on this analogue has been used in the Kristallin-1 (Switzerland) TILA-99 (Finland) and SR 97 (Sweden) safety assessments (see table 5, chapter 6).

Contribution to communication

This analogue might be used to illustrate the performance of the components of barriers made of cement and concrete.

References

Bath, A.H., Christofi, N., Neal, C., Philp, J.C., Cave, M.R., McKinley, I.G. and Berner, U. (1987). *Trace element and microbiological studies of alkaline groundwaters in Oman, Arabian Gulf: a natural analogue for cement pore-waters.* (Technical Report NTB 87-16), NAGRA, Wettingen, Switzerland.



MAQARIN (Jordan)

The Maqarin analogue project was jointly initiated in 1990 by NAGRA, Ontario Hydro and UK Nirex Ltd. In 1991 SKB joined the project, followed in 1993 by the UK-HMIP.

Description of the analogue

The Maqarin site (NW Jordan) is characterised by the development of a layer of cement due to the alteration of metamorphic minerals generated during the spontaneous combustion of marls very rich in organic matter in which temperatures of up to 1,000 °C were reached. The subsequent interaction of seepage waters with the set of natural cement minerals gave rise to the development of groundwaters with very high pH values (up to 13), oxidising conditions and the set of secondary minerals characteristic in the evolution of a hyperalkaline aureole (Figure 1). The discharges of these groundwaters occur along the banks of the river Yarmouk, generally across the colluvions and hillside deposits on which the observations and sampling have focused.

In addition to the characterisation of the solid materials in the cement zone, an exhaustive study was carried out on the groundwaters and their interaction with the host formation. This study allowed the development of a conceptual model of the evolution of a hyperalkaline aureole. Also studied was the distribution of trace elements in the cement minerals and the secondary minerals generated by interaction with the hyperalkaline plume (See table 1).

Analogies and contributions to safety assessment

Maqarin is the best known analogue of the long-term behaviour of the hyperalkaline aureole expected in a disposal system containing cement. The most important analogies are shown in Figure 2.

In the case of hyperalkaline waters of Maqarin and of waters of a hypothetical disposal system including cement, there are three different stages of evolution (Fig. 2): 1) Percolation of rainwater (A); 2) Interaction between these waters and the cement (B); and 3) Flow of hyperalkaline waters charged

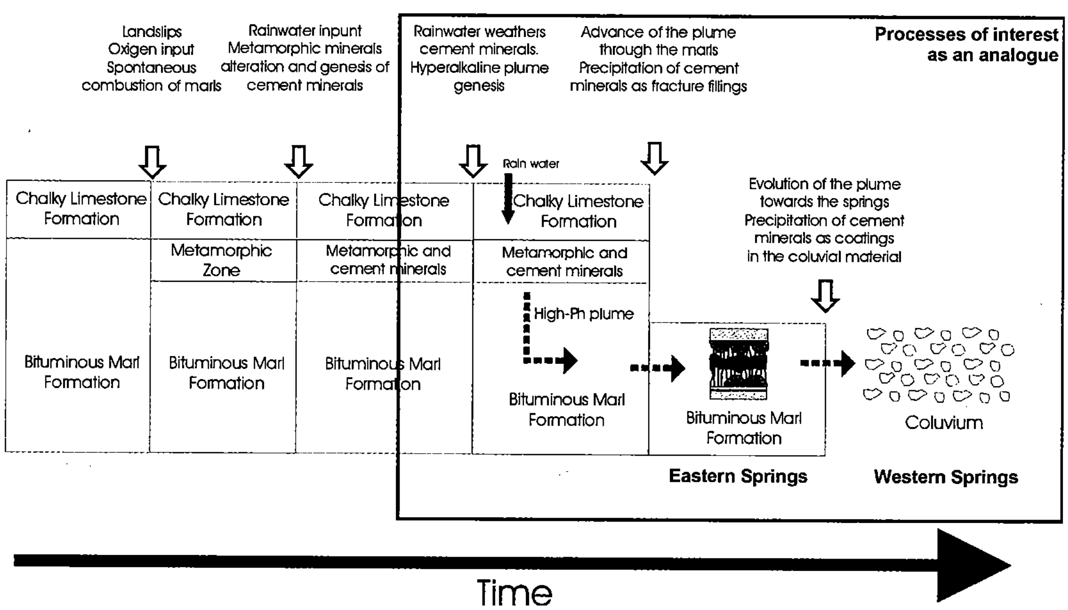


Figure 1. Schematic representation of the timescale of the processes that have given rise to development of a cement zone, to the hyperalkaline plume and to the secondary minerals produced by interaction between the plume and the host marls.

with elements captured during the processes of interaction with the fractured rock and the rock matrix (C).
The main information provided by this analogue relates to the behaviour of certain trace elements under extreme geochemical conditions, as well as to the very evolution of the characteristics of the water. Furthermore, a study has been made of the potential effect of fracture sealing on matrix diffusion.
Other contributions of interest as regards safety assessment are as follows:

- Support for conceptual models and development of new models of the evolution of hyperalkaline aureoles from a disposal system containing cement and their orientation with respect to underground flows.
- Although the porosity of the matrix close to the fractures is reduced by blocking of the pores by secondary minerals, the geochemical data indicate that the rock matrix continues to be accessible for water-rock interaction, which might delay the transport of non-sorbent radionuclides, transported by advection by the hyperalkaline waters via fractures that are still open.
- Proof of the applicability of geochemical and reactive transport codes and of thermodynamic databases under extreme conditions.
- Checking of microbial population prediction codes.

The information on this analogue has been used in the Kristallin-1 (Switzerland), SR 97 (Sweden), TILA-99 (Finland) and H12 (Japan) safety assessment exercises (see table 5, chapter 6).

Contribution to communication

This analogue might be used to illustrate the performance of the components of cement and concretpe barriers. It is quoted in the SKB website (see table 6, chapter 7).

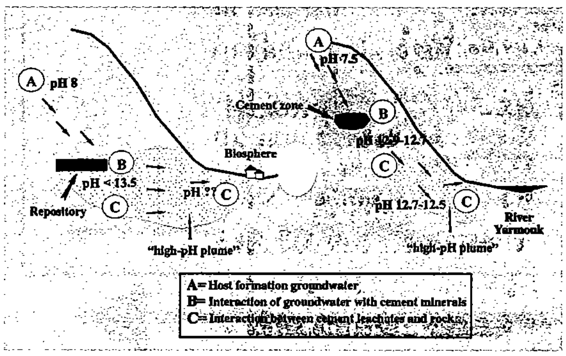


Figure 2. Schematic representation of analogies between the evolution of the hyperalkaline waters of Maqarin (right) and that expected in a disposal system containing cement (left). (Modified from Alexander and Smellie, 1996).

References

Smellie, J.A.T. (Ed.) (1998). *Maqarin natural analogue study: Phase III.* (Technical Report SKB TR 98-04), SKB, Stockholm, Sweden, 400 pp.

Mineral	Ba	Sr	Cr	Cu	Zn	Mn	Pb	U
Jennite								✪
Wairakite	✪	✪	✪	✪		✪	✪	✪
Ettringite			✪		✪			
Calcite		✪				✪		
Aragonite		✪						
CSH	✪	✪			✪	✪	✪	✪
Brucite							✪	✪
Portlandite	✪	✪		✪	✪	✪	✪	✪

Table 1. Fracture refill minerals acting as sinks for different trace elements. (CSH: Hydrated tricalcium silicate). (modified from Linklater, 1998)

2.4. Archaeological analogues

The areas of research that might be enriched by the study of archaeological materials are as follows:

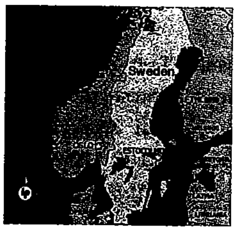
- the *corrosion of cement or metal objects* (iron, copper, bronze) analogous to waste containers or the waste matrices themselves,
- *the degradation of glasses and cementitious or bituminous material* as an analogue of the wastes,
- the *long-term evolution of physico-chemical properties of cements* and other building materials analogous to the structures of the disposal system,
- the *decay and breakdown products of organic material* and complexation with trace elements, analogous to waste degradation, and
- chemical interaction between buried objects and host rocks or soils that might be analogues of near field processes.

Given that archaeological materials and the environment in which they are found may differ to quite an extent from the situation of a disposal system, it is very important to be very careful when selecting a system of this type for research as an analogue. Not all well preserved ancient artefacts may provide information of use in safety assessment.

In order to help in the selection of adequate archaeological analogues from which to obtain qualitative and quantitative information, Miller and Chapman (1995) proposed a list of basic parameters that should be known in relation to the artefact: (1) its age, (2) the composition of the material, (3) the history of the deposit and its preservation, (4) the physiochemical environment in which it has been preserved, (5) any event or environmental change that might have affected the artefact once it was already buried and (6) the composition of the surrounding soil or sediment.

Normally, the greatest problem encountered in studying archaeological deposits as analogues is the characterisation of the burial environment. Most artefacts appear on the surface or at very shallow depths, under conditions that are normally more aggressive chemically than those that would be expected in a disposal system (in terms of chemical flows and the redox environment). The approach adopted for the study of archaeological materials is generally that of estimating degradation rates and mechanisms on the basis of the chemical characteristics of the preservation environment and of extrapolating or interpolating the data to the chemical conditions expected in a disposal system.

The results of archaeological analogue studies may be complemented in most cases with natural analogues of similar materials or processes. Therefore, among the analogues selected in this project, a certain degree of complementarity may be sought between the following archaeological and natural analogues: 1) Inchtuthil nails (Scotland; Fe) and deposits of native iron at Bühl (Germany) and Disko (Greenland); 2) Kronan Cannon (Baltic Sea, Cu) or Santorini (Greece, Cu) and deposits of native cupper at Keweenaw (USA) and Hyrkkölä (Finland); 3) Hadrian's Wall (United Kingdom, cement), Acquarossa (Italy, concrete) or the water tank at Uppsala castle (Sweden, cement) and the hyperalkaline environments of Oman and Maqarin (Jordan); and 4) the Changsha tomb (bentonite, China) and the fossil trees at Dunarobba (Italy).



KRONAN CANNON (Sweden)

The Kronan cannon is the most important archaeological analogue in the Swedish research programme. In the Swedish disposal concept, the canister that will house the radioactive wastes (spent fuel in this case) will be manufactured in copper.

Description of the finding

The Kronan was a Swedish warship, built in 1668. During the battle of Öland it was hit and sunk by the German-Danish fleet in the Baltic Sea, at a distance of 5 Km from the coast of Öland.

Between 1680 and 1686, some 60 cannon were recovered from the sea bed. More recently, between 1980 and 1987, a further 32 cannon were recovered.

Relevant information

The subject of the study has been one of the bronze cannons, which remained vertically positioned with the muzzle downwards with 1.6 metres of its length buried in the sea bed and the remaining 0.2 metres above the bottom in direct contact with the water (Fig.1). The cannon has a very high copper content (96.5%) and contains 3.3% of tin and 0.5% of iron.

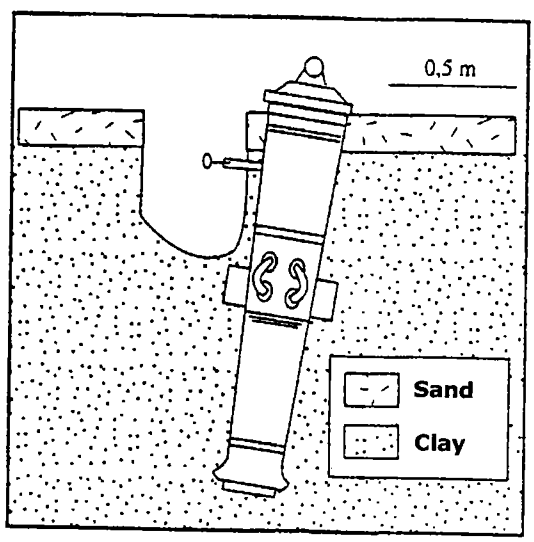


Figure 1. Situation of the cannon on the sea bed (From De Putter Th., Charlet JM., 1994, modified by the UCM)

Analogies

Marine clays may be considered an analogue of bentonite. The corrosion of the copper canister might be less in the reducing conditions expected to exist in the repository.

Contributions to safety assessment

The Kronan cannon supplies information on the type of corrosion that copper undergoes in a clay matrix with oxidising properties. The corrosion products identified included Cu_2O and Fe_3O_4. The average corrosion rate was determined to be 0.15μ/year.

Photograph 1. Moment of recovery from the sea bed of the monitored cannon
Photo courtesy of Fred Karlsson (SKB)

The information provided by this analogue has been used in the TVO-92 (Finland) and SR 97 (Sweden) disposal safety assessments (see table 5, chapter 6).

Contribution to communication

The Kronan cannon provides evidence of the corrosion resistance and longevity of copper, and might be used to increase confidence in the suitability of copper as a canister material.

This analogue is mentioned in the video "Traces of the Future: Lessons from Nature for Waste Disposal" developed by different European waste management agencies, in the SKB website and in the SKB exhibition on board the Sygn ship (see table 6, chapter 7).

References

Hallberg, R.O., Östlund, P., Wadsten, T. A 17th century bronze cannon as analogue for radioactive waste disposal in Côme, B., Chapman, N.A (eds). Natural analogues in radioactive waste disposal, CEC Report EUR 11037 EN: 135-139. 1987.

Smellie, J.A.T. Some Swedish analogue studies: a review in Natural Analogue Working Group, 3rd meeting, Snowbird, CEC Report EUR 11725 EN: 96-103. 1988.

Gumtensperger, M.A., Lindquist, J. (1999). "Public Perception and natural analogues". Report EUR 17543 EN p. 543. Proceedings of the Fourth Conference of the EC on the Management and Disposal of Radioactive Waste, Luxembourg.

Photograph 2. Cannon recovered for study. Photo courtesy of Fred Karlsson (SKB)

INCHTUTHIL NAILS (Great Britain)

Description of the finding

The Roman legionary fortress of Inchtuthil is located in Perthshire (Scotland) and was abandoned in 87 a.d. During their withdrawal, the Romans buried around 875,000 nails in a hole measuring 5 metres in depth, which they then filled in with 3 m of compacted earth.
These nails remained buried until 1950, when the fortress was excavated and the nails recovered.

Relevant information

The nails presented a great many heterogeneities in their composition, with many variations in carbon content. The degree of corrosion of the nails depended, among other factors, on their position in the pile.
The nails located in the interior of the pile showed minimum corrosion, while in the case of those on the outside, and in particular those at the top of the pile, the corrosion was so intense that a large layer of iron oxide had been formed.
In certain of the nails, in very limited areas, localised corrosion (pitting) was observed. No information is available on the position within the pile of the nails affected by this type of corrosion, and the pitting factor has not been quantified. It might be that the pitting was influenced by the iron composition. The nails with the highest carbon content were the largest.

Analogies

At Inchtuthil, the corrosion rate of the outer nails was higher because they were subjected to oxidising conditions. These nails acted as a "barrier" against the corrosion of those located in the interior of the pile.
In general terms this situation may be considered analogous to the one to which the steel and iron of the casks will be subjected in a DGD system.

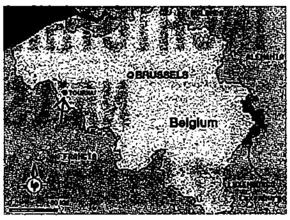
Contributions to safety assessment and communication

Qualitatively, it may be concluded that where there are large volumes of steel in a repository, a large part of it might not be affected by corrosion. In this case it has not been possible to identify the additional information (for example Eh, pH and groundwater chemistry) required for quantitative evaluation of all the variables involved.
This analogue has been used for public communication in the video "Traces of the Future" developed by different European waste management agencies.

References

Angus NS, Brown GT, and Cleere HF (1962). The iron nails from the Roman legionary fortress at Inchtuthil, Perthshire. Journal of Iron and Steel Institute, 200, 956-968.

TOURNAI SARCOPHAGUS (Belgium)



The casks for high level wastes will be constructed from metallic materials, whose long-term performance will need to be assessed. The most widely studied candidate materials for canister manufacturing are carbon steel and copper. Although the Tournai Sarcophagus is constructed of lead, the changes suffered by this material during its burial are being studied.

Description of the finding

The artefact in question is a French-Roman lead sarcophagus (280-300 a.d.) that was discovered in 1989 in the Belgian city of Tournai.

Relevant information

The sarcophagus is made up of two parts: a tub and a cover (Fig. 1). The tub is formed by a sheet of lead measuring from 6 to 9 mm. in thickness, shredded and folded in the shape of a trunk. The wall joints are by lead-zinc welding. The cover was manufactured in the same way.

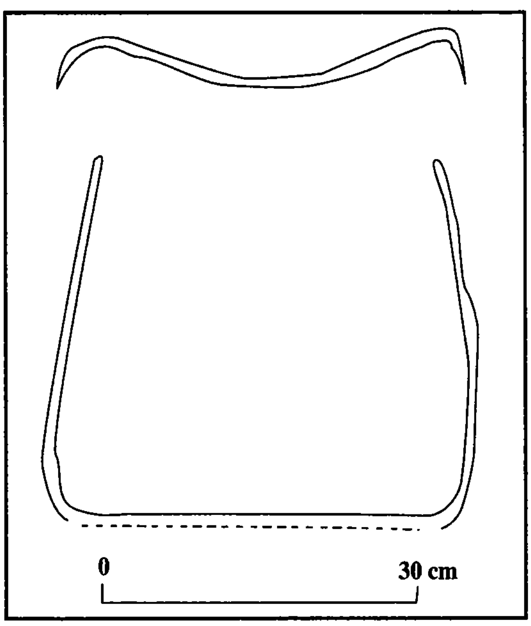


Figure 1: Transverse section of the sarcophagus (From De Putter Th., Charlet JM., 1994).

Figure 1: Transverse section of the sarcophagus (From Putter Th. de, Charlet JM., 1994).

Buried at the end of the third century of our era (280-300 a.d.), the lead sarcophagus contains the body of a man, a glass, a cup and some fifty pieces of bronze.

Within the sandy-clay environment in which it was found, the sarcophagus has been subjected to important hydrostatic pressures that have caused sediments to penetrate the tub.

From the point of view of the mechanical performance of the container, the rapid filling of the sarcophagus gave it greater resistance to deformation.

Chemically, the sarcophagus is made up of metallic lead, protected by a layer of corrosion products: lead oxides and carbonates (PbO and $PbCO_3$) and lead phosphates.

In summary, the corrosion products form a fairly homogeneous protective shield that has preserved the metallic lead against subsequent attacks.

Analogies

The Tournai sarcophagus and the study carried out allow insight to be gained into the corrosion processes that affect a part of the metal buried in a relatively permeable, micro-aerated and periodically saturated geological environment.

Contributions to safety assessment and communication

Although lead is not used as a protective covering for high level radioactive wastes, this study demonstrates (the Tournai sarcophagus testifies this fact) that the corrosion products of a metallic container may contribute to maintenance of the integrity of the container over long periods of time.

References

Brulet, R., 1990. "El sarcófago franco-romano de Tournai"; Publications d'Histoire de l'Art et d'Archéologie de l'Université Catholique de Louvain. LXXIX. (Coll. d'archéologie J. Mertens, IV), Louvain-laNeuve, 76 pp.

ACQUAROSSA (Italy)

The durability and conservation status of concrete with the passing of time is studied at the Etruscan underground water conduit discovered at Acquarossa.

Description of the finding

This construction is an Etruscan underground water conduit dated at between the 7th and 6th centuries b.c. and located at Acquarossa, in the Viterbe region of Italy. The floor and walls of this conduit are coated with a concrete that was in a perfect state of conservation.

The conduit is a tunnel measuring 80 cm in width and approximately 1.80 m in height (Fig. 1). The base of the channel measured some 15 cm in thickness, while the average thickness of the walls was some 5 cm. The upper part of the channel was vaulted, the height of the vault being 35 cm.

Relevant information

The tiling of the floor consisted of bricks joined with a carbonated gypsum (1/1.5 parts on the surface and 1/4.5 parts in the interior). The surface of this area was rough, while the walls, of the same type of concrete but finer, had a smoother finish. The upper part of the walls was covered with a layer of carbonated plaster of a pale colour, the surface of which was highly polished.

The plaster had not altered significantly with time: the calcium silicates found probably came from a reaction between the carbonates and the sand or brick dust present.

Finally, the deposit shows low density (1.85 g/cm^3) and low resistance to compression (9 MPa).

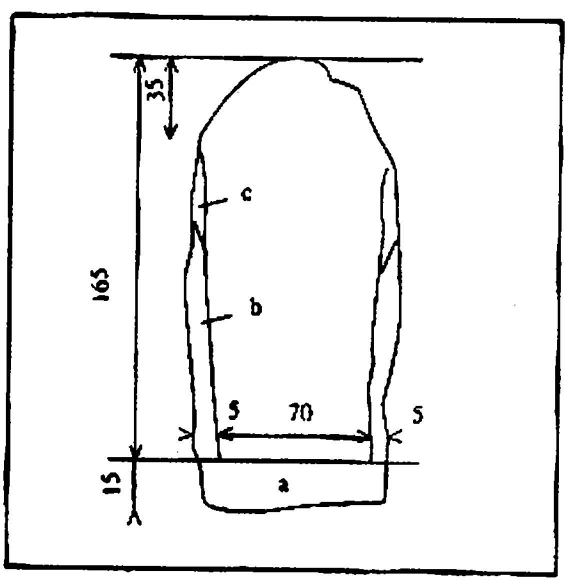


Figure 1: Schematic drawing of the conduit, transverse section (From De Putter Th., Charlet JM., 1994).

Analogies and contributions to safety assessment and communication

The most important analogue of Acquarossa is to be found in the effect of a good degree of conservation of concrete containing carbonates inside silicated rocks.

The analogy with the concrete matrix for intermediate and high level wastes is not particularly significant, since the Acquarossa conduit does not contain important quantities of radionuclides.

References

Malinovske, R., 1979; *"Concretes in ancient aqueducts and conduits"*; Concr.Intern. , 1 (1): 66-76.

Mallinson, L.G., Davies, I.L1., 1987; *"A historic examination of concrete"*; CEC Report EUR10937EN, 309pp.

HADRIAN'S WALL (Great Britain)

Description of the finding

In the year 122 a.d., the Emperor Hadrian ordered the construction of a wall crossing the country, from Solway Firth in Scotland to Wallsend in England. The wall measured 117 km in length and an average 5 m in height.

The wall was of blocks of stone and a cement that acted as a binder. Small fortifications, watchtowers and fortresses were built along the wall (Photo 1). After the jacobite rising in 1745, a part of the wall was destroyed, and today the best conserved parts measure only one metre in height.

Photograph 1. Hadrian's wall view

Relevant information

Hadrian's Wall contained important quantities of CSH compounds (hydrated calcium silicates), the basis of modern Portland cements. The mortar was compact in appearance and had a low porosity. Practically all the calcium oxide in the mortar was uncarbonated, which along with the evidence of reaction in certain aggregates, suggests the presence of CSH compounds.

Two fragments of calcium silicate hydrates (CSH) were identified, one completely impregnated in resin. This material shows a CaO/SiO_2 ratio of ~ 1. The second fragment was very similar to the first in its composition. The presence of wollanstonite and tridimite was identified by X-ray diffraction.

Analogies

Hadrian's Wall is of interest as an archaeological analogue due to the excellent state of conservation of the cement used to join the stone blocks.

The surface environment in the north of England is very different from the conditions that will be found in a disposal system. However, the chemical and mineralogical similarities between the cement used by the Romans and modern Portland cement allow certain qualitative conclusions to be drawn as regards its stability and longevity, which might be extrapolated to the modern cements used in a repository.

Contributions to safety assessment and communication

The main contribution made by Hadrian's Wall is its illustration of the durability of the cement that might be used in the engineered barriers of the DGD system. It is an understandable example that is easily communicated to the public, and has been used in the video "Traces of the future", developed by different European waste management agencies.

References

Rayment DL and Pettifer K (1987) Examination of durable mortar from Hadrian's Wall. Materials Science and Technology 3, 997-1004.

CHANGSHA TOMB (China)

This analogue has been used to study the sealing capacity of clay, preventing the ingress of air and water.

Description of the finding

The burial site consists of two barrows located in rice paddies in Changsha, Hunan province, (southern China). The body was buried in the barrow located furthest to the east, at a depth of 16 m. Burial sites as elaborate as the Changsha tomb were not common in the Ch´u and Han Dynasties, between 770 b.c. and 220 a.d.

Relevant information

The tomb measured 16 m. in depth and was located in a soil whose exact characteristics are not available in the literature. A layer of natural coal measuring 30 or 40 cm. in thickness was placed in contact with sarcophagi to absorb the humidity of the soil. Around this coal layer was another of kaolin, which was what gave the system its waterproofing characteristics (Fig. 1).
The outer sarcophagi contained a large number of funeral offerings: silk garments, bamboo objects, musical instruments, bronze artefacts and even food. The sarcophagus in the innermost part of the shaft contained the body, carefully wrapped in different cloths (silk, linen). Study of the body revealed that it was in an exceptional state of conservation. The fundamental reason for the state of conservation of the body and of the materials found in the sarcophagus is the layer of kaolin, which prevents water and air from seeping into the sarcophagus. There is a direct relationship between the thickness of the layer of kaolin and its effectiveness in conservation.

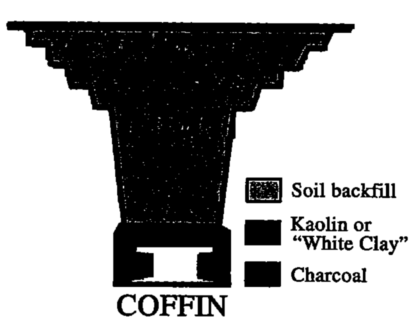


Figure 1: Schematic transverse section of the shaft housing the tomb. (Lee, C.F, 1986. Modified by the UCM).

Analogies

The Changsha tomb illustrates the isolating capacity of clay. This type of burial constitutes an analogy of the clay barrier of the multiple barrier system contemplated in most radioactive waste deep geological disposal concepts.
The limitations of this analogue are the absence of radioactivity and of the thermal effect in the layer of clay. The difference in mineralogy between the type of clay found in the area of Changsha and bentonitic clays should also be underlined.

Contributions to safety assessment

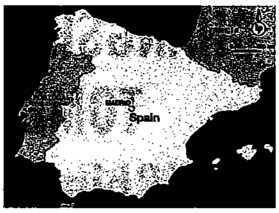
The main contribution to safety assessment made by this analogue is the evidence that it provides of the isolating and sealing capacity of the clay layer present in a radioactive waste disposal system.

Contribution to communication

This analogue might be used to increase public confidence in the safety of DGD, since the timescale involved in this burial site is more easily understood by the general public.

References

Lee, C.F. (1986). "Un caso histórico sobre la capacidad selladora de la arcilla con el paso del tiempo"; NAWG 2nd Meeting, Interlaken, CEC Report EUR 10671, 173-190.



SPANISH DEPOSITS UNDER STUDY

The Spanish programme for the disposal of high level radioactive wastes contemplates carbon steel as the material to be used to construct the capsule. Since 1998 the Mechanical Technology and Archeometallurgy research group of the Complutense University of Madrid has been working with ENRESA in the field of archaeological analogues. Four such analogues are described here.

Analogies

Within the framework of the Spanish analogues a study has been made of the steel evolution over periods of hundreds of years of burial, with a view to predicting the long-term performance of a material placed in service today. The determining factors influencing the phenomenon of corrosion of a buried metal are the type of steel in question and the treatment to which it is subjected during the manufacturing process and subsequent mechanical forming, and the burial conditions.

Contributions to safety assessment and public communication

The study of these deposits has allowed progress to be made as regards the understanding of corrosion processes and estimates of the rate of these processes under different conditions. The results obtained allow corrosion of the metal and the physiochemical variables of the surrounding terrain to be related.

References

ENRESA Technical Publication 7/2000. "Análogos Arqueológicos e industriales para almacenamientos profundos: Estudio de piezas arqueológicas metálicas." (Archaeological and industrial analogues for deep disposal. Study of archaeological metallic artefacts).

(Archaeological steel analogues for the high level nuclear waste container capsule). 5th National Congress on Corrosion and Protection, University College of Mine Engineering, June 2000.

(1) TRESPADERNE BAR (BURGOS)

Description of the finding

This artefact was discovered in a Visigoth necropolis in Trespaderne (Burgos) at a depth of 1.75 m, and dates back to approximately the end of the 4th century a.d. It is a V-shaped steel bar (Fig. 1) of large dimensions. It is in a good state of conservation, with a large part of the original metal still present.

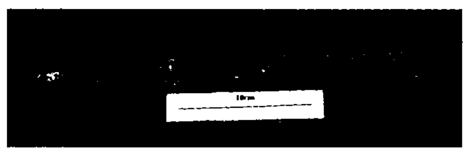


Figure 1. Trespaderne Bar (Enresa Technical Publication 7/2000)

Relevant information

The distribution of carbon in the bar is fairly heterogeneous, ranging from 0.1% to 0.6% by weight depending on the zone in question. There are impurities of elongated morphology (mixed silicates of calcium, magnesium and manganese) resulting from the hot forging process.

The artefact also shows a thin layer of oxides, porous and with the presence of cracks. The pores are filled with substances of a crystalline appearance, with a mainly calcium oxide composition, from the surrounding soil. The corrosion rate of this bar is estimated at around 1.1 µm/year.

As a result of the passing of time, processes of structural ageing and of the diffusion of metallic cations have been detected. Also observed is the presence of bacterial corrosion due to the characteristics of the surrounding terrain.

(2) LAS MATILLAS NAIL (MADRID)

Description of the finding

The nail that constitutes the subject of this study was discovered at an urgently performed excavation in the deposit of Las Matillas (Roman necropolis, 2nd-3rd centuries a.d.), located in Alcalá de Henares, Madrid), alongside several tombs.

Relevant information

The nail is quite mineralised, with wood remains adhering to the oxide. Elements of varying morphology are found on the external part of the transverse section. Energy dispersive spectroscopy analysis (EDS) has served to detect variations in the iron-oxygen proportion in the oxides analysed, suggesting that they are of different stoichiometry.

The analyses performed also indicate the presence of calcium, which would appear to suggest that certain iron atoms have been replaced by atoms of calcium in the crystallographic lattice.

The high degree of mineralisation that this piece has undergone might be explained by the high concentration of chloride and sulphate ions present in the surrounding environment.

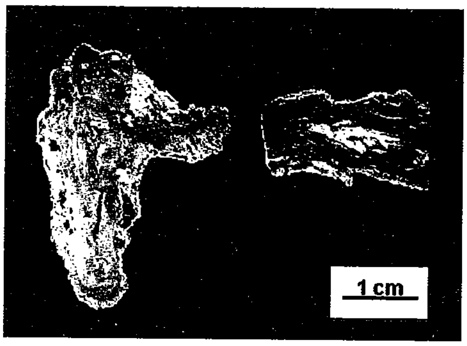


Figure 2. Longitudinal and transverse sections of the piece (Enresa Technical Publication 7/2000).

(3) MIJANGOS NAIL (BURGOS)

Description of the finding

This piece is a steel nail that was found in a grave at Hermitage of Santa María de Mijangos, Burgos (Visigoth era, end of the 5th century a.d.). It weighs 7.52 g and measures 4.4 cm in length by 1.0 cm in diameter.

Figure 3: Transverse section of the Mijangos nail, showing mineralisation. (Enresa Technical Publication 7/2000).

Relevant information

The nail, discovered in a clay soil at a depth of between 2 and 3 metres, is fairly abundantly mineralised, conserving two zones of the original metal arranged on either side of the centreline. The composition and structure of these two zones are different.

The left-hand side zone is ferritic in its structure, while the right-hand zone has a perlitic structure.

The outer part of the nail is completely oxidised. The oxide in this layer has been identified as Fe_3O_4, the inner layer being of Fe_2O_3. Silicates have also been found, these arising as a result of interaction with the soil. This piece has undergone microstructural ageing.

The corrosion rate has been estimated at 0.995 µm/year.

(4) CERRO MURIANO NAILS (CÓRDOBA)

Description of the finding

Various pieces have been studied at the Archaeological Complex of Cerro Muriano (Córdoba), the ones chosen as being the most representative being two nails, one discovered in the deposit of Las Mochas and the other in the deposit of Cerro de la Coja. The Las Mochas deposit is a Roman necropolis dating back to the 1st century b.c. The deposit of Cerro de la Coja is catalogued as a settlement and dates from the 3rd millennium b.c.

The artefact found at Cerro de la Coja showed an internal through-wall crack, and the oxides located inside this crack were completely different in their stoichiometry from those examined on the outside of the piece.

The two nails show structures and impurities typical of a process of mechanical forming such as forging.

Figure 4. Transverse and longitudinal sections of the two artefacts. (Enresa Technical Publication 7/2000)

Conclusions

The main conclusions drawn from the CSN's study on "Natural Analogues" are indicated below, grouped by the contribution made by the analogues to the high level waste disposal safety assessment and to the communication of the disposal concept and safety to a wide range of audiences.

These are general considerations, since the specific conclusions regarding the contribution of natural and archaeological analogues to understanding of the particular processes that may occur in the disposal system and of the areas of safety assessment that might receive support from them are included in the detailed document (in publication) .

In any case, some of the main specific conclusions regarding the potential and real use of analogue studies to the above indicated areas may be drawn from direct observation of tables 5 and 6 and the analogue dockets .

1. CONTRIBUTION MADE BY ANALOGUES TO GEOLOGICAL DISPOSAL SAFETY ASSESSMENT

As it is recognised internationally, the study of analogues constitutes an important source of knowledge for understanding and explanation of the long-term performance of disposal systems. It also represents a complementary approach to support and understand the fundamentals and results of safety assessment and to increase confidence in the capacity of these systems to isolate and contain radioactive wastes for as long as necessary.

This capacity of natural and archaeological analogues to increase confidence in the radioactive waste repository safety is based on the fact that they are the only existing "test beds" or "laboratories" against which to evaluate processes over time periods comparable to the duration expected of the disposal system and they have not been influenced or controlled by mankind during these periods.

To date there has been a series of outstanding contributions made by analogue studies to disposal system safety assessment. There is no doubt that analogues contribute to the identification and understanding of certain processes relevant for safety of the disposal system and provide a "neutral" ground for the checking of hypotheses and evaluation of the suitability of models and codes. They also help to train experts in the analysis of aspects of type and complexity similar to those dealt with in safety assessment.

Specifically, information on analogues has served to (a) provide semi-quantitative estimates of the evolution of certain processes (especially those involved in degradation of the engineered barriers) or ranges of values for certain parameters for testing the conservatism of those considered in calculations,

(b) develop conceptual models and scenarios helping to define the relevant processes, providing arguments for the incorporation or removal of certain processes in assessment calculations, and (c) provide an element to increase confidence in modelling codes and thermodynamic databases.

However, it is difficult to detect the real use made of the information on analogue studies in main disposal safety assessment reports analysed in this study (see Table 4). Only in a few cases there is explicit recognition of the contribution made by analogue studies, or detailed discussion of how this information has been used to support specific aspects of safety assessment.

Nevertheless, a more frequent mention and explicit references to analogues may be found in the detailed documents supporting the above mentioned safety assessment reports, in which explicit reference to analogue studies is much more frequent. This due among other things to the fact that the use of analogues is a substantive part of the scientific basis and knowledge background of the working teams in each organisation responsible for radioactive waste management, even though this is not expressly recognised.

The scarce use actually made of information on analogues is also due to the fact that most waste geological disposal programmes are still far from addressing a licensing process. In the most advanced disposal programmes, where a previously selected or candidate site is available, greater use has been made of analogues because of the possibility of choosing the most adequate as regards similarity with the selected disposal concept and easing direct transference of analogue information, as complementary argument demonstrating safety.

Although analogues allow to place the processes or models considered for the waste disposal assessment within the spatial and time-related scales of interest in safety assessment, the impossibility of establishing with absolute accuracy the initial and boundary conditions affecting the evolution of the analogue and the problems associated with the variability and intrinsic heterogeneity of natural systems, imply some limitations in the application of their results to the performance assessment of deep geological disposal.

Consequently, the use made to date of the information obtained from analogue studies has been more qualitative than quantitative in most cases. Furthermore, it has been seen that the application of natural analogues to safety assessment requires closer collaboration between the working teams involved in disposal performance assessment and those in charge of the study of analogues.

A future line of work that remains to be exploited is the use of analogues in relation to alternative scenarios in repository safety assessment since, up to now, the use of information from natural analogues has focused excessively on scenarios of normal evolution (in fact, many of the problems or criticisms that have emerged in evaluating the degree of analogy assume the conditions expected in these base case or reference scenarios).

It cannot be forgotten, however, that in disposal safety assessment exercises a series of alternative scenarios is analysed (glaciation, disruptive scenarios, etc.), being the evaluations of their evolution usually more qualitative than quantitative. Therefore, it is foreseeable that part of the information obtained from analogues whose conditions were not initially those expected in a normal scenario be perfectly usable in support of these scenarios (the same reasoning might be applied to the selection and use of possible future analogues).

The application of analogue information to knowledge of transport phenomena in the biosphere, which has not been analysed in this project, might also be of interest.

Furthermore, the possibility of reviewing and re-evaluating the information from previous analogue studies, in the light of the results of experiments developed in underground laboratories and using new models and tools (as has been carried out in the case of Cigar Lake) is an option that remains open for obtaining greater benefit from other analogue studies.

2. CONTRIBUTION OF ANALOGUE STUDIES TO COMMUNICATION

Natural analogues are one of the main arguments that may be used to explain the long-term performance of the disposal system and to communicate the results of safety assessments to technical and non-technical audiences using easily understood manner. In general, there is ample agreement as regards the fact that many groups not familiar with the conceptual and technical aspects of geological disposal will give greater credibility to those conclusions of performance assessment that are accompanied by parallel natural examples facilitating comparison and understanding (Miller *et al.*, 2000).

However, despite this recognition of the potential of natural and archaeological analogues to illustrate and communicate relevant aspects of radioactive waste disposal systems and their long-term safety, especially to non-technical audiences or those not familiar with the subject, there is still no generalised use made of analogues for these purposes by waste management agencies and regulatory bodies.

In almost all the cases in which analogues have been used for communication purposes, the information was oriented towards explaining the most generic aspects of geological disposal concept to a broad range of audiences.

Analogues have been used to transmit easily understood messages on: 1) the geological disposal concept and its potential safety; 2) the similarity existing between the materials and radionuclides in the disposal facility and those found in nature, 3) the fact that the relevant processes that may take place in a waste repository occur also in natural analogues and 4) that the timescales addressed in disposal safety assessment may be observed in natural analogues. Another series of important ideas that are not always explicitly transmitted underlie these messages, such as the degree of general knowledge acquired regarding processes that might affect the disposal system (for example, none of the analogues has pointed to the existence of unexpected processes of radionuclide transport or retardation).

On the other hand, the need to simplify the contents in order to facilitate their understanding by non-technical audiences should not imply an overestimation of the validity of the analogues or an underestimation of the audience. Increasing confidence in the safety of high level waste repositories must necessarily rest on the incorporation of arguments supporting perceptions on long term stability of disposal components and characteristics and on the intrinsical safety of the deep geological disposal concept (NEA, 2002).

Finally, and as recommended by the NEA (2002), the use of information from natural and archaeological analogue studies should constitute one of the multiple lines of reasoning used to demonstrate the safety of a disposal system. As has already been pointed out, the step-wise decision-making process for waste disposal development is not only a technical question, but one in which the political and social actors should also take part. For this reason, the challenge of answering the interests and concerns of the public in relation to disposal system safety will require the development of material and information aimed at a broad range of audiences which incorporate analogue information.

According with everything aforementioned, the application of natural analogues to the safety assessment of waste disposal systems and to public communication is gaining interest at international level. Proof of this is the European Commission NANet project[18], driven to promote the use of natural analogues for these purposes, which includes the participation of the CSN on the basis of the experience acquired in the Analogue Study summarized in this publication.

In addition, mention should be made of the growing and declared tendency among all the radioactive waste agencies to include analogue studies in their safety assessments, having been also requested by the regulatory bodies of some countries or even considered in some national regulations. Finally, it should be pointed out the consideration of natural analogues in the IAEA disposal safety requirement currently in development, as one of the multiple lines of reasoning aimed at increasing confidence in safety assessment (IAEA, 2002).

18 NANet: "Network to review natural analogue studies and their applications to repository safety assessment and public communication"

References

Angeli, F., Faucon, P., Charpentier, T., Petit, J.C. y Virlet, J. (1998). *Comparative structural study and dissolution of simplified glasses: a radioactive waste glass (R7T7) and a basaltic glass*. In: McKinley, I.G. and McCombie, C. (Eds.) Scientific Basis For Nuclear Waste Management XXI. MRS, Warrendale, PA, USA, pp. 71-78.

Alexander, W.R. and Smellie, J.A.T. (1996). The Maqarin natural analogue project: an overview. In: von Maravic, H. and Smellie, J.AT. (Eds.) Proceedings of the Sixth EC Natural Analogue Working Group (NAWG) Meeting. Sante Fe, NM, USA, September 12-16, 1994. EC NST, Luxembourg, Luxembourg, pp. 123-135.

Bath, A.H., Christofi, N., Neal, C., Philp, J.C., Cave, M.R., McKinley, I.G. and Berner, U. (1987). *Trace element and microbiological studies of alkaline groundwaters in Oman, Arabian Gulf: a natural analogue for cement pore-waters.* (Technical Report NTB 87-16), NAGRA, Wettingen, Switzerland.

Baudoin, P., Gay, D., Certes, C., Serres, C., Alonso, J., Lührmann, L., Martens, K.H., Dodd, D., Marivoet, J., y Vieno, T. (2000). *Spent fuel disposal Performance Assessment (SPA project)*. Report EUR 19132, 114 p.

Benvegnù, F., Brondi, A. y Polizzano, C. (1988). *Natural analogues and evidence of long-term isolation capacity of clays occurring in Italy: contribution to the demonstration of geological disposal reliability of long-lived wastes in clays.* Nuclear Science and Technology Report EUR 11896, EC NST, Luxembourg, 101 p.

Blomqvist, R. and Ruskeeniemi, T. (1997). *Transport of Radionuclides in a Natural Flow System at Palmottu.* (Progress Report SKB 97-15), SKB, Stockholm, Sweden.

Blomqvist, R., Ruskeeniemi, T., Kaija, J., Ahonen, L., Paananen, M., Smellie, J., Grundfelt, B., Pedersen, K., Bruno, J., Pérez del Villar, L., Cera, E., Rasilainen, K., Pitkänen, P., Suksi, J., Casanova, J., Read, D., Frape, S. (Ed.) (2000). *The Palmottu natural analogue project. Phase II: transport of radionuclides in a natural flow system at Palmottu.* (Final Report EUR-19611), EC-NST, Luxembourg, Luxembourg, 171 p.

Brandberg, F., Grundflet, B., Höglund, L.O., Karlsson, F., Skagius, K. y Smellie, J.A.T. (1993). *Studies of natural analogues and geological systems. Their importance to performance assessment.* Technical Report SKB TR 93-05, SKB, Stockholm, Sweden.

Côme, B. y Chapman, NA. (1986). CEC *Natural Analogue Working Group. First Meeting.* EUR 10315, Brussels, Belgium, 5-7 November, 1985.

Côme, B. y Chapman, NA. (1987a). *Proceedings of the second* CEC/NAWG Meeting. EUR 10671, Interlaken, Switzerland, 17-19 June, 1986.

Côme, B. y Chapman, NA. (1987b). *Natural Analogues in Radioactive Waste Disposal, Proceedings of a Symposium held in Brussels. EUR 11037, 28-30 April, 1987.*

Côme, B. y Chapman, NA. (1989). CEC *Natural Analogue Working Group. Third Meeting*. EUR 11725, Snowbird, near Salt Lake City, USA, 15-17 June, 1988.

Côme, B. y Chapman, NA. (1991). *Proceedings of the Fourth Natural Analogue Working Group Meeting and Poços de Caldas Project Final Workshop.* EUR 13014, Pitlochry, Scotland, 18-22 June, 1990.

Chapman, N.A., McKinley, I.G., Shea, M.E. y Smellie, J.A.T. (1984). *The potential of natural analogues in assessing systems for deep disposal of high-level radioactive waste.* Tech. Rep. KBS 84-16.

Chapman, N.A., McKinley, I.G., Shea, M.E. and Smellie, J.A.T. (1991). *The Poços de Caldas project. Summary and implications for radioactive waste management.* (Technical Report SKB TR 90-24), SKB, Stockholm, Sweden.

Cramer, J.J. and Smellie, J.A.T. (Ed.) (1994). *Final Report of the AECL/SKB Cigar Lake Analog Study.* (Tech. Rep SKB TR 94-04), AECL, Pinawa, Manitoba, Canada.

CSN (2003). Salas, E., Ruiz, M.C. *Estudio comparativo de las evaluaciones de seguridad del almacenamiento profundo de residuos radiactivos de alta actividad en medios cristalinos. (Comparative study of safety assessments of the deep geological disposal of high level radioactive wastes in crystalline media).*

Curtis, D.B. y Gancarz, A.J. (1983). *Radiolysis in nature: evidence from the Oklo natural reactors.* KBS Technical Report, TR 83-10.

De Putter, Th. y Charlet, J.M. (1991). *Analogies Naturelles en Milieu Argileux. Essai de synthèse bibliographique.* ONDRAF, October, 1994.

Dexter, S.C. (Ed.) (1986). *Biologically induced corrosion. Proceedings of the International Conference on Biologically Induced Corrosion.* Houston, EE.UU.

Duro, L., Arcos, D., Bruno, J., Jordana, S., Grivé, M., Pon, J., Castelier, E., Beaucaire, C., Fauré, M.H., Peña, J., Gimeno, M.J., Del Nero, M., Ayora, C., Salas, J., Ledoux, E. and Madé, B. (2000). Blind prediction modelling (BPM) exercise in Oklo. In: Louvat, D., Michaud, V. and von Maravic, H. (Eds.) Oklo working group. Proceedings of the third and final EC-CEA workshop on Oklo-Phase II [CD-ROM]. Cadarache, France, May 20-21, 1999. EC NST, Luxembourg, Luxembourg, pp. 285-306.

Enresa (1997). *Evaluación del Comportamiento y de la Seguridad de un Almacenamiento Geológico Profundo en Granito. (Performance and safety assessment of deep geological disposal in granite)* Technical Publication 06/97, ENRESA, Madrid, Spain.

Enresa (1999). *Evaluación del Comportamiento y de la Seguridad de un Almacenamiento Profundo en Arcilla. (Performance and safety assessment of deep geological disposal in clay)* Technical Publication 03/99, ENRESA, Madrid, Spain.

Enresa (2000). *Análogos arqueológicos e industriales para almacenamientos profundos: estudio de piezas arqueológicas metálicas. (Archaeological and industrial analogues for deep disposal facilities: study of metallic archaeological artefacts)* Technical Publication 00-07, ENRESA, Madrid, Spain.

Gauthier-Lafaye, F. (1996). Introduction to the Oklo problematic. In: Blanc, P.L. and von Maravic, H. (Eds.) Oklo Working Group. Proceedings of the fourth joint EC-CEA progress and final meeting. Saclay, France, June 22-23, 1995. EC NST, Luxembourg, Luxembourg, pp. 5-16.

Gera, F., Hueckel, T. y Peano, A. (1996). *Critical issues in modelling the long-term hydro-thermo-mechanical performance of natural clay barriers*. Eng. Geol., 41, 17-33.

Goodwin, B.W., Cramer, J.J. and McConnell, D.B. (1989). The Cigar Lake Uranium deposit: an analogue for nuclear fuel waste disposal. In: Côme, B. and Chapman, N.A. (Eds.) CEC Natural Analogue Working Group. Third Meeting. Snowbird, near Salt Lake City, USA, June 15-17, 1988. CEC, Luxembourg, Luxembourg, pp. 181-190.

Goodwin, B.W., McConnell, D.B., Andres, T.H., Hajas, W.C., LeNeveu, D.M., Melnyk, T.W., Sherman, G.R., Stephens, M.E., Szekely, J.G., Bera, P.C., Cosgrove, C.M., Dougan, K.D., Keeling, S.B., Kitson, C.I., Kummen, B.C., Oliver, S.E., Witzke, K., Wojciechow (1994). *The disposal of Canada's nuclear fuel waste: post-closure assessment of a reference system.* (Technical Report AECL-10717), Pinawa, Manitoba, Canada, 684 p.

Gumtensperger, M.A., Lindquist, J. (1999). "Public Perception and natural analogues". Report EUR 17543 EN p. 543. Proceedings of the Fourth Conference of the EC on the Management and Disposal of Radioactive Waste, Luxembourg.

Hofman, B.A. (1992). *Isolated reduction phenomena in red beds: a result of pore water radiolysis?*. In Kharaka, Y.K. and Maest, J. (Eds.) Proceedings of the 6th International Symposium on Water-Rock Interaction. Balkema, Rotterdam, Holland.

IAEA (1989). *Natural analogues in performance assessment for the disposal of radioactive wastes.* IAEA Technical Report, 304, International Atomic Energy Agency, Vienna, Austria.

IAEA (1996). *Issues in radioactive waste disposal. Second report of the Working Group on Principles and Criteria for Radioactive Waste Disposal.* TECDOC-909, International Atomic Energy Agency, Vienna, Austria, October 1996.

IAEA (1997). *Regulatory decision making in the presence of uncertainty in the context of the disposal of long lived radioactive wastes. Third report of the Working Group on Principles and Criteria for Radioactive Waste Disposal.* TECDOC-975, International Atomic Energy Agency, Vienna, Austria, October 1997.

IAEA (1999). *Use of natural analogues to support radionuclide transport models for deep geological repositories for long lived radioactive wastes.* IAEA Technical Report 1109, International Atomic Energy Agency, Vienna, Austria.

IAEA (2002). *Geological Disposal of Radioactive Waste.* IAEA Safety Standard Series-Draft Safety Requirements DS 154, 2002.10-14 (Status: for approval, for submission to the Member States)

ICRP (2000). *Radiation Protection Recommendations as Applied to the Disposal of Long Lived Solid Radioactive Waste.* ICRP Publication No. 81, International Commission on Radiological Protection, Pergamon Press, Oxford and New York (2000).

JNC (2000). H12: *Project to establish the scientific and technical basis for HLW disposal in Japan.* Second Progress Report on Research and Development for the Geological Disposal of HLW in Japan.

Lagerblad, B. y Trägardh, J. (1995). *Conceptual model for concrete long time degradation in a deep nuclear waste repository.* SKB TR 95-21, Stockholm, Sweden.

Linklater, C. M. (Ed.) (1998). *A natural analogue study of cement-buffered, hyperalkaline groundwaters and their interaction with a repository host rock.* (Science Report S/98/003), NIREX, Harwell, Oxfordshire, UK, 301 p.

Mckinley, I.G. y Alexander, W.R. (1996). *The uses of natural analogue input in repository performance assessment: an overview.* In: von Maravic H and Smellie J (editors) Natural analogue working group, sixth meeting, Santa Fe, September 1994. CEC Nuclear Science and Technology Report, EUR 16761M 273-2282, CEC, Luxembourg.

Mckinley, IG (1993). *The role of natural analogue in nuclear waste repository performance assessment with particular emphasis on experience in Switzerland.* Reliability Engineering and System Safety, 42, Nos 2-3, 233-246, Elsevier Science Publ.

Miller, W., Alexander, R., Chapman, N., McKinley, I. y Smellie, J. (2000). *The geological disposal of radioactive waste and natural analogues. Lessons from Nature and archaeology.* Pergamon, Amsterdam, vol. 2, 316 p. Waste Management Series.

Miller, W.M. y Chapman, N.A. (1995). *Postcards from the past: archaeological and industrial analogues for deep repository materials.* Radwaste Mag., 2, 32-42.

NAGRA (1994). *Kristallin-I. Safety Assessment Report.* (Technical Report NTB TR 93-22), NAGRA, Wettingen, Switzerland, 396 p.

NEA (1991). *Disposal of Radioactive Waste: Review of Safety Assessment Methods.* Paris, France.

NEA (1994). ARAP. *The international Alligator rivers analogue project. Background and results.* OECD Nuclear Energy Agency, Paris, France, 77 p.

NEA (1997a). *Regulating the Long-Term Safety of Radioactive Waste Disposal. Proceedings of an International Workshop.* Córdoba, Spain. 20-23 January 1997, OECD Nuclear Energy Agency, 337 p.

NEA (1997b). *Lessons Learnt from Ten Performance Assessment Studies.* OECD Nuclear Energy Agency, Paris, France, 129 p.

NEA (1999a). *Confidence in the Long-term Safety of Deep Geological Repositories. Its Development and Communication.* OECD Nuclear Energy Agency, Paris, France, 80 p.

NEA (1999b). *Geological Disposal of Radioactive Waste. Review of Developments in the Last Decade.* OECD Nuclear Energy Agency, Paris, France, 106 p.

NEA (2000). *Regulatory Reviews of Assessments of Deep Geological Repositories, Lessons Learnt.* OECD Nuclear Energy Agency, Paris, France, 134 p.

NEA (2001). *The Role of Underground Laboratories in Nuclear Waste Disposal Programmes.* OECD Nuclear Energy Agency, Paris, France, 43 p.

NEA (2002). *Approaches and arguments to establish and communicate confidence in safety and the overall results of Integrated Performance Assessments.* (Report in the press).

Neall, F.B., Baertschi, P., McKinley, I.G., Smith, P.A., Sumerling, T.J. y Umeki, H. (1994). *Kristallin-I - results in perspective. (Technical Report NTB 93-23)*, NAGRA, 120 p.

Pellegrini, R., Horseman, S., Kemp, S., Rochelle, C., Boisson, J.-Y., Lombardi, S., Bouchet, A. y Parneix, J.-C. (1999). *Natural analogues of thermo-hydro-chemical and thermo-hydro-mechanical response.* Report EUR 19114, CEC, Luxembourg.

Pérez del Villar, L., De la Cruz, B., Pardillo, J., Pelayo, M., Rivas, P. and Astudillo, J. (1993). El sistema granito-mineralización de Uranio de El Berrocal como análogo natural de un almacenamiento de residuos radiactivos. (The El Berrocal granite-Uranium mineralisation system as a natural analogue of a radioactive waste repository) *Estud. Geol.*, 49(3-4), 187-198.

Pusch, R. and Madsen, F.T. (1995). *Aspects on the illitization of the Kinnekulle bentonites.* Clays Clay Miner., 43, 261-270.

Savage, D. (Ed.) (1995). *The scientific and regulatory basis for the geological disposal of radioactive waste.* John Wiley & Sons, 437 p.

SKB (1999a). *Deep repository for spent nuclear fuel. SR 97. Post-closure safety.* (Technical Report SKB TR 99-06), SKB, Stockholm, Sweden.

SKB (1999b). *SR 97. Processes in the repository evolution. Background report to SR 97.* (Technical Report SKB TR 99-07), SKB, Stockholm, Sweden.

SKI (1991). *SKI Project-90. 2 vols.* Technical Report SKI TR 91/23, SKI, Stockholm, Sweden.

SKI (1996). *SKI SITE-94. Deep repository performance assessment project. 2 vols.* Technical Report SKI R 96/36, SKI, Stockholm, Sweden, 305-660 p.

Smellie, J.AT., Karlsson, K. y Alexander, W.R. (1997). *Natural analogue studies: Present status and performance assessment implications.* J. Contam. Hydrol., Vol. 26.

Smellie, J.A.T, Grundfelt, B., Carlsson, F (1999). *"Natural Analogue (NA) Approach: Have they provide quantitative data applicable to repository performance assessment (PA) and contribute to a better public perception on radioactive waste disposal?"*, p. 243. EUR 19143. Proceedings of Euradwaste 1999, Luxembourg, November 15-18, 1999.

Steadman, J.A. (1986). *Archaeological concretes as analogues.* Technical Report EUR 10671, EC-NST, Luxembourg, 165-171 p.

Trotignon, L., Michaud, V. and Louvat, D. (2000). *Application of the Oklo Natural Analogue Study to Nuclear Waste Disposal Model, Testing and Development.* In: Louvat, D., Michaud, V. and von Maravic, H. (Eds.) Oklo working group. Proceedings of the third and final EC-CEA workshop on Oklo-Phase II {CD-ROM}. Cadarache, France, May 20-21, 1999. EC NST, Luxembourg, Luxembourg, pp. 401-408.

USDOE (1996). Title 40 CFR 191 *Compliance Certification Application (CCA) for the Waste Isolation Pilot Plan.,* DOE/CAO-1996-2184, Carlsbad Area Office.

USDOE (1998). *Viability Assessment of a Repository at Yucca Mountain. Total System Performance Assessment. Vol. 3.* DOE/RW-0508, Office of Civilian Radioactive Waste Management, 523 p.

U.S. Nuclear Regulatory Commission. *10 CFR Part 63, Disposal of High-Level Radioactive Wastes in a Proposed Geologic Repository at Yucca Mountain, Nevada.* Proposed rule 10 CFR Part 63.

Valentini, G., Lombardi, S., Bozzano, F. and Scarascia Mugnozza, G. (Ed.) (1997). *Analysis of the geo-environmental conditions as morphological evolution fac-*

tors of the sand-clay series of the Tiber valley and Dunarobba forest preservation. (Report EUR 17479), EC-NST, Luxembourg, Luxembourg.

Vieno, T. y Nordman, H. (1999). *Safety assessment of spent fuel disposal in Hästholmen.* Kivetty, Olkiluoto and Romuvaara. TILA-99, POSIVA 99/07, Helsinki, Finland.

von Maravic, H. y Smellie, J.A.T. (1994). *Fifth CEC Natural Analogue Working Group meeting and Alligator Rivers Analogue Project (ARAP) final workshop. Proc. of an international workshop,* EUR 15176, Toledo, Spain, 5-9 October, 1992.

von Maravic, H. y Smellie, J.AT. (1996). *Proceedings of the Sixth EC Natural Analogue Working Group (NAWG) Meeting.* EUR 16761, Sante Fe, NM, USA, 12-16 September, 1994.

von Maravic, H. y Smellie, J.A.T. (1997). *Seventh EC Natural Analogue Working Group Meeting. Proceedings of an international workshop.* EUR 17851, Stein am Rhein, Switzerland, 28-30 October, 1996.

Winberg, A. and Stevenson, D. (1994). Hydrogeological modelling. In: Cramer, J.J. and Smellie, J.A.T. (Eds.) *Final Report of the AECL/SKB Cigar Lake Analog Study.* (Tech. Rep SKB TR 94-04), AECL, Pinawa, Manitoba, Canada, pp. 104-142.

Annex A. Relevant performance and safety assessment exercises of different disposal concepts

Most representative integrated performance/safety assessment studies of waste repositores completed in recent decades, indicating the performing organisation and the objective of the study.

Organisation	Main references and other relevant documents	Assessment objectives
AECL Canada	AECL (Atomic Energy of Canada Limited) 1994. Environmental Impact Statement (EIS) on the concept for disposal of Canada's nuclear fuel waste. AECL Report. AECL-10711, COG-93-1. Various publicly available sub-documents The disposal of Canada's nuclear fuel waste: Post-closure assessment of a reference system (Goodwin et al., 1994) The disposal of Canada's Nuclear fuel waste: A study of the post-closure assessment of used CANDU fuel in copper containers in permeable plutonic rocks (Goodwin et al., 1996) AECB (Atomic Energy Control Board) 1995. AECB staff response to the environmental impact statement on the concept for disposal of Canada's nuclear fuel waste. INFO-0585-1. Public.	To support decision-making in relation to future development of the radioactive waste programme in Canada. Deterministic and probabilistic assessment analysis of the reference disposal system to demonstrate the feasibility and safety of the disposal concept. Assessment of alternative concepts: copper container in horizontal galleries. Regulatory review of the EIS.
EC European Commission	PAGIS (Performance Assessment of Geological Isolation System for Radioactive Waste) (Cadelli N. et al., 1988) –5 volumes. PACOMA - Performance assessment of the geological disposal of medium level and alpha waste in a clay formation in Belgium. (Marivoet and Zeevaert, 1990). First Performance Assessment of the Disposal of Spent Fuel in a Clay Layer (Marivoet et al., 1996). Evaluation of elements responsible for the effective engaged dose rates associated with the final storage of radioactive waste: EVEREST project (Cadelli, N. et al., 1996, Marivoet, J. et al., 1997). SPA (SPent Fuel Assessment project) (EC, 1998).	To build a common understanding of the methods applicable to deep disposal performannce assessment considering various types of wastes, repository designs and host rock formations.
ECN, RIVM, RGD Holland	PROSA Prij et al.,.1993	Study of the option for radioactive waste disposal in salt formations.
ENRESA Spain	ENRESA-97 -- Performance assessment of a spent fuel repository in granite (ENRESA, 1997) ENRESA-98 -- Performance assessment of a spent fuel repository in clay (ENRESA,1998)	To develop a methodology for the the performance assessment of disposal systems at a generic site in granitic and clay rocks, respectively
GRS Germany	GSF-91: "Analysis of the long-term safety of disposal concepts with heat producing radioactive wastes" (Buhmann et al. 1991) (in German)	Study and compare different disposal concepts.
HMIP Great Britain	Dry Run 3: Trial assessment of underground disposal based on probabilistic risk analysis (Sumerling ed., 1992)	Testing of the capacities of environmental simulation codes applied to the analysis of a hypothetical low and intermediate level waste repository at the Harwell site.

Continuation. Most representative integrated performance/safety assessment studies of waste repositores completed in recent decades,indicating the performing organisation and the objective of the study.

Organisation	Main references and other relevant documents	Assessment objectives
JNC **Japan**	H3: "Research and development on geological disposal of high-level radioactive waste. First Progress report" (PNC, 1993) The second progress report: H12 Project for assessment of feasibility of HLW disposal in Japan (JNC,1999)	Summary of the current status of R&D programme and a basis for the furhter research and development Demonstration of the technical feasibility and safety of the Disposal concept and support for site selection and development of the regulatory framework
NAGRA **Switzerland**	NAGRA 1985 Proyecto Gewähr Kristallin-I Safety Assessment Report (Nagra 1994)	Demonstration of the feasibility and safety of disposal system, in response to a requirement by the Swiss Government Testing and development of the assessment methodology and guidance for the site selection process
NIREX **Great Britain**	Nirex 95: A preliminary analysis of the Groundwater Pathway for a Deep Repository at Sellafield (Nirex, 1995a). Nirex 97: An assessment of the post~closure performance of a Deep Waste repository at Sellafield (Nirex, 1997).	Iterative development of the disposal concept Demonstration of the capacity developed for the performance assessment in candidate site, integrating information from the sites and the R&D programme
NRI, RAWRA, **Czech Republic**	BAZ 97-02, The role of reference system in deep geological repository development (Konopaskova et al,. 1997)	Evaluation of the role of the barriers in the reference disposal system
ONDRAF/NIRAS **Belgium**	SAFIR (Niras/Ondraf 1989) SAFIR 2 (Safety Assessment and Flexibility Interim Report (December 2001)	Study of the radiological impact of waste disposal on Boom Clays Demonstration of the feasibility of the Disposal option in Belgium. Compilation of the results of 10 years of R&D
POSIVA **Finland**	TVO-92 safety analysis of spent fuel disposal. Helsinki, Nuclear Waste Commission of Finnish Power Companies. Report YJT-92-33E.(Vieno, T., Hautojärvi, , A., Koskinen, L. & Nordman, H. 1992) TILA-96. Interim report on safety assessment of spent fuel disposal TILA-96. Helsinki, POSIVA-96-17. (Vieno, T. and Norman, H. 1996) TILA-99. Safety assessment of spent fuel disposal in Hästholmen, Kivetty, Olkiluoto and Romuvaara. TILA-99. Helsinki Posiva, POSIVA-99-07. (Vieno, T. and Norman, H. 1999)	Basis for decision-making regarding the future development of the radioactive waste management programme and the site selection process. Preliminary site studies in 5 candidate sites. Response to the regulatory body's requirement for periodic reports on safety assessment, incorporating site and R&D programme data Basis for a "Decision in Principle" by the Finnish Government on spent fuel disposal and Environmental Impact Assessment and the associated site selection process

Continuation. Most representative integrated performance/safety assessment studies of waste repositores completed in recent decades, indicating the performing organisation and the objective of the study.

Organisation	Main references and other relevant documents	Assessment objectives
SKB **Sweden**	KBS-1, KBS-2, KBS-3 (SKB 1977,1978,1983) SKB-91, Final disposal of spent nuclear fuel. Importance of the bedrock for safety (SKB, 1992) SR-97, Post-closure safety – Main report, Volumes 1 and 2 (SKB, 1999)	Illustration of possible options for waste management and first preliminary assessment concept Development of the safety assessment methodology Demonstration of the feasibility of the KBS-3 concept, testing of the assessment capability and basis for the site selection process
SKI **Sweden**	SKI Project-90 (SKI, 199l) SKI SITE 94 Deep Repository Performance Assessment Project (SKI, 1996)	Development of regulatory assessment review capability, demonstration of the performance assessment methodology Development of regulatory assessment capability, evaluation of the way to assimilate o site specific data in the assessment
US DOE/WIPP **Estados Unidos**	Draft 40 CFR 191 Compliance Certification Application (DCCA) for the Waste Isolation Plant (SNL, 1995).	Allowing for technical discussions with the regulatory authority prior to the submission of a final license application and basis for site or geological media selection
US DOE/ YMP **United States**	Total-System Performance Assessment - 1993: An Evaluation of the Potential Yucca Mountain Repository, R.W.Andrews, T.F. Dael and J.A. McNeish, Las Vegas, Nevada, IN'I'ERA. Iric. March 1994. Prepared for the U.S. Department of Energy/Yucca Mountain Site Characterization Project TSPA-95. TRW Environmental Safety System Inc. "Total System Performance Assessment - 1995: An Evaluation of the potential Yucca Mountain repository." Las Vegas, Nevada, Document nº B00000000-01717-2200-00136 (Rev. 01, Nov-1995) Viability Assessment of a Repository at Yucca Mountain, Total System Performance Assessment (U.S. Department of Energy, 1998)	Development of the safety assessment methodology: treatment of system design and site information and model development Quantitative evaluation of the suitability of the Yucca Mountain site, Demonstration of safety assessment methodology and the feasibility of the disposal as regards compliance with regulatory requirements
US NRC, **United States**	Initial demonstration of the NRC's capability to conduct a performance assessment for a high-level waste repository (Codell et al., 1992) NRC Iterative Performance Assessment Phase 2: Development of capabilities for review of a performance assessment for a high-level waste repository (Wescott et al. Eds., 1995) Iterative Performance Assessment Phase 3: status of activities (Manteufel & Baca, 1995)	Development and demonstration of the performance assessment methodology Development of regulatory assessment capability Development of regulatory assessment capability and of a specifiic regulations applicable to Yucca Mountain

Anex B. Analogies of processes that might occur in a deep geological disposal system

1. DISSOLUTION OF THE WASTE MATRIX (SPENT FUEL OR VITRIFIED WASTES FROM REPROCESSING)

The processes of alteration and dissolution of the waste form, be it spent fuel or high level vitrified waste, give rise to the release of the radionuclides present in them, and consequently play a basic role in geological disposal safety assessment. These processes are closely related to certain of those that take place in the near field, affecting the wastes themselves and the retention of leached radionuclides (precipitation of secondary phases, sorption, etc.), and their treatment is usually integrated into the so-called radionuclide release models.

In the case of *spent fuel*, of all the different sources from which radionuclides may be released (Zircaloy cladding, cladding-UO_2 pellet gap, UO_2 matrix and grain boundaries), the most important is the UO_2 matrix, since most of the activity present in the spent fuel is contained with it. The chemical alteration of the UO_2 matrix may occur as a result of simple dissolution or transformation processes (oxidisation) of the UO_2 into oxides of different stoichiometries. Both processes are closely linked and their coupled actuation will depend fundamentally on the redox state of the system, with UO_2 dissolution rate being higher with more oxidising conditions. Furthermore, associated with the UO_2 alteration process there will be the appearance of secondary phases that might affect the passivation processes and the retention of the radionuclides released.

In view of the gradual formation kinetics involved, the analysis of the uraninite alteration processes and of the formation of secondary phases is a particularly significant aspect in natural analogue studies, and provides information on the evolution of spent fuel under different conditions and the effects on the migration and retention of the radionuclides present in the UO_2 matrix on timescales considered in safety assessment.

In the studies carried out in uranium ore deposits, the sequence of uraninite alteration has been analysed under both reducing conditions (analogues of *Oklo* in Gabon, *Cigar Lake* in Canada and *Palmottu* in Finland) and under oxidising conditions (analogues of *Shinkolobwe* in the Congo, *Koongarra* in Australia and *Krunkelbach* in Germany). The qualitative observations support the long-term stability of the spent fuel during the maintenance of reducing conditions, since under such conditions the dissolution rate of the uraninite is extremely slow.

Furthermore, in the natural analogues studied, the process of coffinisation (the transformation of U (IV) dioxide into a U (IV) silicate as a result of the addition of silica to the uraninite) has been identified as a uraninite alteration process under anoxic conditions. In the models normally used in safety assessment, It is assumed the hypothesis that the UO_2 matrix is stable under reducing conditions, while there has been evidence that the coffinite formed at the expense of the uraninite, although capable of retaining many radionuclides released from the waste, alters rapidly if there is a change in the chemical characteristics of the solution from which it was formed (*Trotignon et al., 2000*). This increases interest in its study in the field of safety assessment.

In the case of *borosilicate glass for high level waste stabilisation*, radionuclide release occurs exclusively from the processes of glass alteration. Glass is a metastable phase that may with time undergo processes of devitrification (crystallisation in the solid state) to form thermodynamically more stable phases, and then phases more sensitive to dissolution than the original glass may be produced. Furthermore, the glass matrix may dissolve and react with groundwater when it comes in contact with it after the canister failure, generating secondary alteration phases with an important capacity to retain previously released radionuclides.

The studies of natural and archaeological analogues relating to borosilicate glass matrixes have provided relatively abundant information on the processes of devitrification, dissolution and the formation of secondary alteration phases (Miller *et al.*, 2000) that attest to the long-term stability of these materials under a wide range of conditions. On the other hand, available data on radionuclide retention in these secondary phases or on the effects induced by radiation are very scarce or even non-existent.

Furthermore, natural analogues have contributed to the development of conceptual models and to the verification of the radionuclide release models and codes used in disposal safety assessments, and provide quantitative data on the long-term dissolution rates of uraninite and natural glasses, which may be of use, at least as ranges for comparison.

2. CRITICALITY

In a disposal system, due to the considerable quantities of actinides contained in the wastes, after a certain period of time there may be an accumulation of fissionable isotopes such as Uranium-235 and Plutonium-239, which, in the presence of neutrons and an appropriate moderator capable of reducing the speed of these neutrons (such as the groundwater that may access the area close to the canister), might cause a self-sustaining nuclear fission chain reaction. The negative implications that this might have for the safety of the disposal system are the generation of fission products, in turn disintegrating into a high number of radionuclides, heat production and, in the most extreme case (when multiplication factor exceeds the unit), the initiation of a divergent and uncontrolled fission reaction that might cause irreparable damage to the disposal system. Criticality is not analysed in the base case or normal evolution scenario of a high level waste repository, but may be contemplated in other scenarios of altered evolution, with canister failure and subsequent ingress of water.

Oklo is the only natural analogue where conditions of criticality were reached, with nuclear fission chain reactions, as a result of which it provides a unique opportunity to study certain processes that cannot be observed in other uranium deposits, such as the transport of transuranic radionuclides and the stability of uranium minerals once conditions of criticality have been reached. Nevertheless, the analogy between the criticality reached at Oklo and that which might affect the spent fuel used in nuclear reactors has certain weak points, such as the fact that the burnup temperature at Oklo, of between 400 and 600 ºC, is lower than that generated in a nuclear reactor, and that the time over which the criticality event occurred at Oklo was much longer than the period during which the nuclear fuel is inside the reactor.

Information on the Oklo analogue has been used in certain safety assessment exercises (SR 97, USDOE-VA) to analyse the possibility of criticality in a high level waste disposal system, concluding that such an event would be highly improbable either inside or outside the waste canister.

3. RADIOLYSIS

Radiolysis is the process of chemical decomposition of matter due to the impact of high energy ionising radiation. In the near field of a deep geological disposal facility radiolysis produced by radiation emitted during high level wastes radioactive decay might affect the groundwaters, the organic matter and the bentonite. Among these possibilities, the radiolysis of the groundwater is the most important.

The two main water radiolysis processes are ionisation (in which the aqueous cation radical and an electron are generated), and electronic excitation (in which an excited water molecule is produced). Through different chemical reactions, these three species produce the hydroxyl radical, the solvated electron and the hydrogen radical, which in turn may recombine to generate hydrogen gas, hydrogen peroxide and water.

The theoretical models assume that the most important reducing species, H_2, is chemically inert and migrates outside the system (due to its high diffusivity), while the oxidants are more reactive and have lower diffusivity, as a result of which an oxidising redox front is generated and moves slowly from the waste towards the barriers. The extent of the migration of this front is controlled by the balance between the production of oxidants and their consumption by the iron-rich materials in the engineered barriers and rocks in the near field.

The main effect produced by radiolysis in a deep geological disposal system is the alteration of geochemical conditions in the medium surrounding the canister, as a result of the production of reducing and oxidising agents. These oxidising agents may affect the corrosion of the canister, the dissolution of UO_2 and the solubility and speciation of radionuclides, facilitating their migration towards the biosphere.

Water radiolysis has been identified in uranium deposits with high radiation fields, such as *Oklo* or *Cigar Lake*, or in other deposits such as *Cluff Lake* and *Rabit Lake* (Canada) *Shinkolobwe* and *Menzenschwand* (Germany), which represent different geological environments and chemical conditions, this suggesting that it is a common natural phenomenon. Its study in these systems contributes to a better understanding of the process by allowing insight to be gained into the behaviour of radiolysis products once generated, determination of whether these products induce the oxidisation and degradation of uraninite, establishment of what fraction of the total radiation impacts the surrounding water (in the knowledge that the radiation fields in these natural systems will in all cases be far inferior to those that will exist in a high level waste repository), and the development and improvement of models simulating the process.

4. CORROSION

Corrosion is defined as the degradation or dissolution of a material, mainly metals, as a result of reaction with the surrounding medium. In a high level waste disposal system, corrosion is the main cause of canister failure under the repository expected conditions , It may also give rise to secondary alteration phase products and gas generation. Failure as a result of corrosion will be due to localised and/or general corrosion phenomena, which will depend to a large extent on geological medium, the site, the concept, the characteristics of the groundwater and, evidently, the canister material (as well as the welding methods, etc.).

Generalised corrosion occurs uniformly over the entire exposed surface of the material, or much of itn. Localised corrosion takes place in a given area of the material and initiates generally in locations in which there are inclusions or variations in compositions in the crystalline structure. The main types of localised corrosion that can occur under disposal conditions are pitting, stress corrosion, by overlapping and by galvanic coupling. In certain metals a layer of corrosion products may form on the surface of the metal as the corrosion process progresses, this being known as passivation. This process may limit the flow of metallic ions to the solution and reduce the corrosion rate. In this case localised corrosion may take place as a result of destruction of the passive layer.

The corrosion of metal is also possible in association with the action of living organisms, this being known as bacterial corrosion. The factors that determine the presence of these living organisms and their study in natural analogues are described in section 16 of this annex.

Although abundant information has been obtained on corrosion rates from laboratory experiments, it is difficult to extrapolate their results to periods as long as those to be considered in radioactive waste disposal systems. In these experiments a more or less linear dependence of the corrosion rate is assumed with respect to time, a dependence that ceases to be linear when a passive layer is formed on the surface of the metal.

The study of the corrosion process in metallic archaeological analogues (*Kronan Cannon, Tournai Sarcophagus, Spanish archaeological analogues, etc.*) provides a way of quantifying, to the extent possible, the behaviour of a material in response to corrosion over timescales of hundred to thousands of years. By estimating the corrosion rate of the material constituting the archaeological artefact, it is possible to obtain an approximate value of the service lifetime of a canister of similar material. The observations made of archaeological analogues also underline the capacity of certain canister corrosion products to delay the migration of radionuclides in the near field, this being of great relevance for disposal safety assessment.

5. DISSOLUTION – PRECIPITATION OF IMPURITIES IN BENTONITE

The processes of dissolution-precipitation of the accessory components present in the bentonite, along with the processes of ion exchange that occur during the interaction between the groundwater and the bentonite, are especially relevant in high level waste disposal safety assessment, since they are considered to control the geochemical evolution of the waters in the bentonite barrier and, therefore, the radionuclide solubility limits. In addition, the processes of dissolution-precipitation of certain accessory components of the bentonite, such as ferrous sulphides and carbonates, play an important role in bentonite function as a barrier. For example, the presence of certain proportions of pyrite or siderite in the bentonites favours the maintenance of a reducing environment in this barrier.

The impact of pyrite dissolution processes on the maintenance of reducing conditions in the bentonite has been repeatedly contrasted both in the laboratory and in theoretical simulations of water-bentonite interaction processes. These processes have been implicitly considered in many disposal safety assessment exercises, although their importance have been less explicitly referenced, possibly because the calculations of the geochemical conditions in the near field also integrate the effects of anoxic canister corrosion , which are far less effective as redox condition buffers.

In this respect, the effect of the ferrous sulphides and carbonates dispersed throughout the clay halo of *Cigar Lake*, which maintain a reducing and stabilising environment in the uranium deposit, constitutes a clear analogy of the process and a qualitative verification of the extrapolations performed, by means of mathematical simulations, on their long-term effects for the normal or reference scenario.

The processes of dissolution-precipitation induced by a thermal gradient will be commented on in the next section, in relation to cementation processes.

6. PROCESSES OF BENTONITE DEGRADATION: CEMENTATION, ILLITISATION

The bentonite used as buffer, backfill and sealing material in a high level waste disposal system will undergo chemical processes that might cause changes in the capacity of the material to fulfil its functions as a barrier, especially during the thermal transient period. In this respect, the thermal gradient induced in the bentonite barrier causes variations in the solubility of the mineral phases and in reaction rates, favouring processes of dissolution, alteration or the transformation of certain constituents of the bentonite at some locations and precipitation at others. Depending on the intensity of this type of processes, the rheological properties of the bentonite may be negatively affected (increased brittleness, reduction of swelling capacity, etc.), consequently resulting in the deterioration of bentonite buffer properties as a barrier.

The most important processes as regards the long term degradation of bentonite are those of dissolution-precipitation (cementation) that arise as a result of water-bentonite interaction and the transformation of smectite into illite (illitisation).

Processes of *cementation* are defined as the chemical precipitation of mineral phases produced from the interstitial waters of the bentonite in the pores or voids in this material, and are the result of the coupled actuation of thermohydrochemical processes. These processes, and their effects on the mechanical properties of the bentonite, have been studied through laboratory experiments, showing that they would be of little significance and would be controlled fundamentally by processes affecting the accessory mineralogy of the bentonite. However, these results are affected by the very limitations of the working scale of laboratory experiments and by the difficulties involved in their extrapolation to much greater timescales.

This circumstance might be resolved through the studies of silica cementation processes that have been carried out in natural analogues of bentonitic nature, such as *Busachi* (Sardinia) and *Kinnekulle* (Sweden). Quite apart from any conceptual vagueness or alternative interpretations regarding the mechanisms involved, the observations made in these analogues indicate that the intensity of silica cementation processes would not appear to be sufficiently significant as to constitute a relevant problem for the integrity of the barrier, thus confirming the results of laboratory experiences and extrapolating them to timescales of relevance to safety assessment. However, the process has been studied in a very small number of analogues and, in certain of them, the conditions under which the process of silica cementation has occurred might not be extrapolate to the expected conditions in a radioactive waste disposal facility, this underlining the need for the process to be analysed through additional studies.

The transformation of smectite into illite, or *illitisation*, is a process of mineral replacement (in which there is an exchange of the exchangeable cations of the smectite for the potassium ion from the interstitial solutions) that occurs naturally in geological systems subjected to relatively high temperatures and over fairly widely varying timescales, in processes such as diagenesis due to burial, contact metamorphism (igneous intrusions) or hydrothermal alteration.

In a deep geological repository, the process of illitisation associated with the thermal transient period is one of the main degradation processes of the bentonite buffer, and the most probable at high temperatures. This process implies a degradation of the swelling capacity, permeability and cation exchange properties of the bentonite (since these properties are less favourable for illite than for smectite) and might consequently affect the processes of diffusion, sorption and radionuclide retardation in the near field.

As regards the aforementioned geological processes, diagenetics and igneous intrusions are the ones that have been considered analogous to the process of illitisation in a waste disposal system. Regional metamorphism environments are not adequate for use as analogues, since both the temperatures reached and the duration of the material heating period are much more extreme than would be expected in a waste disposal repository. Neither are hydrothermal systems particularly suitable as analogues, since the process of illitisation occurs under bounding conditions (temperature, water-rock interaction, etc.) that are highly variable and not easy to determine (*Miller et al., 2000*).

The number of natural analogues studied that might provide information on this process is considerable: *Kinnekulle, Hamra, Busachi, Burgsvik, Forsmark, Ignaberga, Murakami, Orciatic, Skie Isle and Col de Perthus.* Furthermore, at *Cigar Lake*, the fact that the clay halo surrounding the uranium deposit is made up fundamentally of illite (formed at the same time as the deposit) would be equivalent to a situation in which the bentonite barrier had undergone a process of almost total degradation, and would therefore constitute a unique opportunity to determine the performance of an illitic material as a barrier. The results obtained from the analogue studies, along with those of laboratory experiments on different types of bentonites, suggest that the process of illitisation of the bentonite barrier would require periods of time in excess of those considered in the area of safety assessment.

The study of the illitisation process in natural analogues and systems has provided quantitative data on the intensity or periods of time required for the occurrence of the process under widely varying conditions, and has made it possible to verify the simple kinetic models contemplated for its description, which have been calibrated using data from natural systems (*Hamra, Kinnekulle, Burgsvik*). At the same time, the uncertainties affecting the modelling have been underlined through the use of more sophisticated tools such as reactive transport codes, as well as the potential use of analogues for the verification of this type of tools.

7. SPECIATION-SOLUBILITY: SOLUBILITY LIMITS CALCULATION

The speciation-solubility properties of radionuclides are specific to each individual element and also depend on the chemical characteristics of the groundwater present in the geosphere following its interaction with the different barriers in the near field. Consequently, these characteristics depend on the geochemical behaviour of the near field (bentonite, canister and waste) under different conditions (temperature, pH, etc.), depending also on the disposal system evolution or scenario considered.

Calculation of radionuclide solubility limits consists of determining the solid phases that control the concentration of these elements in a fluid phase of defined characteristics. This requires definition of the solution considered, calculation of the scheme of speciation corresponding to each element in this solution, identification of the limiting solid phase most probably controlling the concentration of each element in the solution and calculation of the concentration of the element in equilibrium with this phase, which will be its solubility limit. These calculations imply the use of geochemical models and associated thermodynamic databases. Besides, speciation, in addition to being of fundamental importance as regards solubility, conditions the properties of radionuclide transport in the bentonite and host rock.

The calculation of solubility limits is one of the key points in disposal system safety assessment exercises, since it determines the specific concentration of each radionuclide during its transport from the waste to the biosphere. In most assessment exercises, these calculations are associated with the near field. The estimation of additional solubility limits for the evolution of the transport of these radionuclides in the geosphere is normally discarded conservatively, although in some exercises limits are also defined for this component of the disposal system.

The analogue studies have been used in disposal safety assessment to testing of the conceptual geochemical models, the associated thermodynamic databases and the numerical codes required to describe the migration of radionuclides under repository conditions, and increasing confidence in such models, databases and codes.

"Blind prediction modelling" exercises (BPM)[19], designed to compare the results obtained with different geochemical codes, thermodynamic databases and modelling approaches, have been carried out on numerous natural analogues (*Oman, Poços de Caldas, Cigar Lake, Maqarin, El Berrocal, Oklo and Palmottu*). These have been performed by different working groups with a view to predicting the expected behaviour of trace elements in natural environments. The main contributions made by the BPM exercises as regards analogues are as

19 BPM: Blind Predictive Modelling exercises

follows: (a) identification of the solids and/or processes that control their solubility; (b) quantification of the aqueous concentrations of radionuclides under the different geochemical conditions that may occur in the near and far-field environments; and (c) suitable treatment for their modelling, where the information derived from BPM exercises is extremely relevant.

8. DEGRADATION OF CEMENT: GENERATION AND EVOLUTION OF THE HYPERALKALINE PLUME

The cement and concrete used as a barrier in radioactive waste disposal systems will eventually degrade, by way of a process that will take place in different stages (Lagerblad and Trägardh, 1995) depending on the characteristics of the medium, fundamentally the composition of the groundwaters and host material. These successive stages will condition the development and evolution of the hyperalkaline plume and its effect on the near field. Any local variation in pH as a result of the degradation of cement will have implications for the solubility of the radionuclides, for the metal corrosion rate, for degradation of the bentonite and ultimately for alteration of the host rock.

The conceptual approach to cement degradation considers an initial period, during which the concrete will be altered as a result of contact with atmospheric gases, especially carbon dioxide. Subsequently, the degradation will be governed fundamentally by the concrete-groundwater interaction process, which will modify the composition of both components. An aureole of high pH fluids will be created around the concrete, interacting with the materials in contact with it and affecting fundamentally the bentonite and also the host rock. In addition, the development of the hyperalkaline plume is directly related to other processes, such as radionuclide retention in secondary phases, either as a result of sorption mechanisms or of coprecipitation (see section 14), and the generation of silica colloids at the advance fronts of the plume (see section 16).

The study of cements in archaeological constructions such as the *Acquarossa conduit* (Italy) and *Hadrian's Wall* (England), of ancient industrial facilities or of natural systems such as *Oman* and *Maqarin* (Jordan), analogues of the hyperalkaline environments that may be generated in the near field of a repository containing important amounts of cement, may provide valuable information on the process of degradation of this barrier over time - taking into account their limitations - and allow consideration to be given to the influence of the process of generation and evolution of the hyperalkaline plume on the repositpory performance.

In the aforementioned natural analogues, studies have been carried out on the hyperalkaline waters and their effect on radionuclide transport processes, with "blind predictions" (BMP) performed to check the codes and methodologies used to model equilibrium conditions in a hyperalkaline rock-groundwater interaction system. Maqarin is the best analogue to study the evolution of the hyperalkaline plume and its interaction with the host material, since the alkalinity of its waters is the result of leaching of natural cement minerals and has helped in the conceptual modelling of hyperalkaline plume development.

9. FLUID FLOW

The flow of groundwaters in the geological formation housing a radioactive waste disposal system is a highly important process in fractured media such as granite. In such formations, the fractures, immersed in a matrix of very low permeability, may act as preferential paths in which the groundwater flow concentrates. The characteristics of this flow depend mainly on the geometry and configuration of the system of fractures.

In a waste repository, water flow affects the transport and dispersion of radionuclides and their retardation, since it determines the surface available for matrix diffusion and sorption, and also affects the lithological stresses and temperature and gas transport in the bed-rock.

The flow may be altered locally and/or globally by changes in the engineered barriers or in the rock, variations in recharge/discharge (climate, topography, changes in sea level), the generation and flow of gas, salinity gradients, variations in temperature affecting the density and viscosity of the water, etc. (SITE-94).

There is a significant corpus of knowledge of fluid flow through geological formations, due to the considerable development of hydrogeology achieved in recent decades. The most relevant information on flow processes in the geosphere is obtained fundamentally from site characterisation studies and underground laboratories. However, and in spite of the uncertainties existing in the hydrological boundary conditions, the study of the hydrogeology in geological formations similar to those proposed for a disposal system (analogues) may provide information and help to corroborate predictions regarding important aspects of groundwater flow, such as for example water transit times from the repository to the biosphere.

Hydrogeological studies in natural analogues (*Oklo, Cigar Lake, Palmottu, El Berrocal, Poços de Caldas*) have contributed to the development and checking of groundwater flow conceptual models and numerical codes and to the development and testing of innovative field techniques for hydrogeological characterisation. Furthermore, many of the hydrogeological studies performed in areas other than the purely nuclear are also valid as analogues. In this respect, mention may be made of the contributions made by the field of palaeohydrogeology.

10. ADVECTION AND DISPERSION

The advection process refers to the movement of a substance by the motion of the fluid in witch it is present. Hydrodynamic dispersion is the mass transport that occurs naturally, from areas of higher concentration to others of lower concentration of solute, as a result both of molecular diffusion and mechanical dispersion.

In the geological barrier (far field) of a radioactive waste disposal system, and in the case of fractured formations, the flow of water circulates preferentially through the fractures and the transport in the geosphere of the radionuclides dissolved in the groundwaters is controlled fundamentally by the phenomena of advection and dispersion.

The processes of transport by advection and dispersion are classical processes that have been widely studied in hydrogeology and that may also be investigated in natural systems, in view of the analogy with the processes expected in a waste repository. The study of these processes in natural analogues, among which *Oklo* (Gabon), *Palmottu* (Finland), el *Berrocal* (Spain) and *Maqarin* (Jordan) may be singled out, has provided abundant information on the parameters involved in the advective-dispersive transport of solutes and has allowed conceptual models of such transport to be developed.

11. MOLECULAR DIFFUSION

Diffusion in water is defined as the process of movement of by which matter is transported from certain regions of a system to others as a result of molecular agitation. The diffusive flow (quantity of mass spreading across a unit surface per unit of time) is proportional to the gradient of its concentration.

The molecular diffusion of chemical species via the interstitial waters filling the porous space in a geological medium is limited by the presence of solid phases and by the irregular paths of the pores. For this reason, diffusion in porous media is lower than in pure water. The magnitude of the molecular diffusion in a geological medium depends on porosity, the distribution or pore size and the complexity of the paths. The importance of this process is quantified by a parameter known as the effective diffusion coefficient, which is a property of the system overall: the porous medium, the water in the pores and the species diffused.

In a radioactive waste disposal system, molecular diffusion is the main transport mechanism in the bentonite barrier, and in the case of clay formations also in the geosphere. In the case of fractured media, molecular diffusion in the geological barrier is important fundamentally in the rock matrix (zone of rock weakly or very slightly fractured), and is known as matrix diffusion.

11.1. Diffusion in clay media: bentonite and clay geosphere

As has been pointed out above, in very low permeability media (as clays normally are) molecular diffusion becomes the main mechanism for the migration of the dissolved chemical species. This is applicable to buffer/ backfill materials (bentonite) and to the repository host rock , in the case of clay formations.

The process of molecular diffusion through the bentonite barrier has been studied fundamentally by means of laboratory experiments and in situ, since the control of the boundary conditions and geometry in such experiments is much greater than in natural analogues and, independently of process timescale, the spatial scale of the engineered barrier can be tested in the laboratory. For this reason, the contribution made by analogues as regards the process of diffusion in bentonite is more limited, although it has been studied in some detail in the *Cigar Lake* analogue.

Molecular diffusion in *clay formations* has basically been studied under laboratory conditions, although it has also been analysed in various analogues, such us *Koongarra* (Australia), *Dunarobba* (Italy) and *Loch Lomond* (Great Britain). This last analogue has provided quantitative data for the diffusion of certain species of interest, and it has showed that the diffusion coefficients obtained in laboratory tests are always greater than those of natural analogues, and consequently conservative from the point of view of disposal safety assessment.

11.2. Diffusion in the rock matrix

In fractured geological formations with low permeability, the permeability of the fractures is orders of magnitude higher than in the rock matrix, this giving rise to what is known as dual porosity systems. The diffusion of solutes in the rock matrix around the fractures occurs mainly in a system of connected pores and microfractures and may be significant, justifying the need to consider dual porosity models in order to simulate transport in these media.

In a radioactive waste disposal system in a fractured formation, such as granite, the main matrix diffusion effect consists of radionuclide retention by the rock matrix, causing both a decrease in maximum activity and radionuclide retardation in their movement towards the biosphere.

There are many natural systems in which the process of matrix diffusion may be studied: 1) systems with the remobilisation (transport) of natural uranium from the interstitial water in the rock matrix to the nearby transmissive fractures; 2) adjacent rocks with fractures in which the groundwater changes its ionic strength; and 3) systems with redox fronts migrating from the fractures towards the interior of the rock matrix as a result of interactions between the dissolved oxygen and the surfaces of the fractures, where there is circulation of oxidising water with a low degree of mineralisation. In these systems it is possible to study the redistribution of the natural elements of interest in the rock adjacent to the conductive fractures, or the diffusion of radionuclides in non-fractured rocks around uranium deposits, produced over timescales comparable to those addressed in the evolution of a disposal system.

Matrix diffusion is one of the processes most widely analysed in natural analogue studies (*Palmottu, Poços de Caldas, El Berrocal, East Bull Lake* -Canada-) and, therefore, one of the processes most widely contrasted on the basis of such studies. However, it is a process that is difficult to measure and quantify. The study of this type of analogues makes it possible to gain insight into the factors that regulate this process, such as the volume of interconnected porosity, the importance of grain boundaries, microfractures and the mineralogy of the matrix, obtain ranges of values of apparent diffusivity (incorporating the effects of various processes: diffusion, sorption-desorption and alpha recoil), along with the corresponding uncertainties, identify the relative importance of matrix diffusion compared to other retardation mechanisms and check process models.

As regards quantitative data on matrix diffusion obtained from natural analogues, in these systems it is used the concept of diffusion penetration depth, in reference to the thickness of the adjacent rock affected by the process. However, this concept is not a parameter of any process but the result of matrix diffusion and sorption in a given system, which depends on a series of factors: time, the physico-chemical properties of the diffusing chemical species or radionuclide, the chemical characteristics of the interstitial water and temperature. For this reason, *Miller et al. (1994)* suggest that the values proposed in relation to diffusion penetration depth be taken with some precaution.

12. WATER-ROCK MATRIX INTERACTION PROCESSES

When referring to water-rock interaction processes in the geosphere of a radioactive waste disposal system, we mean the set of processes that conditions the characteristics and evolution of the waters that will come into contact with the different barriers of the system as well as the evolution of certain mineralogical characteristics of the solid medium due to interaction with the groundwaters and of interest from the point of view of radionuclide retention.

Although many of the other water-solid interaction processes analysed individually (speciation-solubility, precipitation-coprecipitation-dissolution, ion exchange, sorption, matrix diffusion, colloids, etc.) may be included in the two aforementioned generic processes, the jointly approach to water-rock matrix interaction process is due to the fact that these processes are formally considered in safety assessment exercises when referring to geochemical processes in the geosphere. These processes will limit both the behaviour of the materials in the engineered barriers and the transport of radionuclides through the geosphere.

Given that the observations of water-rock interaction processes in the hydrogeochemical system of natural analogues are equivalent to the studies required for the geochemical characterisation of a waste disposal site, the first ones may be incorporated into such studies and complete them. This evidently requires that in the geological disposal programme candidate sites for the repository were selected and that there be an important similarity between the nature of the analogue and the selected geosphere.

Natural analogue studies have contributed to the development and verification of conceptual models for the processes of dissolution-precipitation of secondary mineral phases, underlining the importance of these processes in the maintenance of hydrogeochemical stability in fractured media (*Cigar Lake, Poços de Caldas*) and their transcendental participation in the retention of the radionuclides transported by the water (*Palmottu, El Berrocal*). Furthermore, the application of geochemical modelling methodologies in analogue studies, with the use of a wide range of codes, as well as "blind prediction" exercises, allows the verification of the applicability of geochemical and reactive transport codes and thermodynamic databases, thus contributing to increasing confidence in them.

13. RETARDATION PROCESSES: SORPTION AND PRECIPITATION/COPRECIPITATION

Retardation is a measure of the retention suffered by a reactive chemical species in dissolution in a dynamic system as a result of interaction with the solid phase (for example by sorption or precipitation). It may be expressed by means of the coefficient of distribution, K_d, which relates the concentration of the radionuclide sorbed on solid phase, C_S, and the concentration sorbed in a solution in equilibrium with the solid, C_L.

In a waste disposal system, the majority of the radionuclides released from the waste[20] to the groundwater will interact with the materials of the disposal system and with the surfaces of the rock in contact with the water, or will be subject to changes in the behaviour of the solution due to the modifications caused by water-rock interaction along the flow path. These processes may delay their movement with respect to that of the water, not only slowing down progress through the system but also reducing their concentration in solution, leading to situations of equilibrium or even promoting their removal from the solution due to incorporation in solid phases. These retardation processes are, therefore, favourable from the point of view of the safety of the disposal system.

The following are among the retardation mechanisms identified in natural systems:

- chemical retardation mechanisms: sorption (adsorption and ion exchange), precipitation and coprecipitation
- physico-hemical retardation mechanisms: diffusion in the secondary porosity of the rock matrix, molecular filtration and ion exclusion.

Sorption is the interaction process of dissolved chemical species with the solid surface. It generically encompasses adsorption and ion exchange and takes place to counteract the charge defects that occur in solids.

Precipitation and *coprecipitation* are processes of fixation or structural immobilisation of solutes in a new mineral, and are generally considered to be irreversible. Precipitation consists of the placing of an element in a regular position of the ions of that element in a simple solid phase. In the case of coprecipitation, the new mineral is a mixed solid phase, in which the trace element occupies an atomic position that would normally be occupied by an ion of a different element.

Sorption is the mechanism considered to dominate the retardation and retention of solutes. However, it is very difficult to distinguish between the different

20 Except elements such as tritium, ^{36}Cl and ^{129}I, which are not retained by any process in their migration towards the biosphere.

processes that cause radionuclide retardation: sorption, precipitation-coprecipitation, matrix diffusion. This implies that occasionally a given effect cannot be assigned with any certainty to a single process.

The study of the surroundings or uranium deposits helps to gain insight into the factors that influence these retardation processes of mobilised radionuclides (or, for example, of rare earth elements, chemical analogues of certain actinides) in the different types of mineral substrates present in any geosphere, clay or granite, and for a wide range of conditions of the solution (pH, ion strength, concentration of carbonate...), and also develop predictive models adequately describing the delay of such radionuclides under these conditions. In certain cases it has even been possible to determine values of coefficients of distribution *in situ,* and although these results depend entirely on the place analysed, they may give an idea of the parametric ranges and orders of magnitude, with their associated uncertainties.

The process of *sorption in bentonite* has not been considered in depth in natural analogues, although there have been studies of *sorption in clay formations (Loch Lomond) and sorption in the materials filling granitic fractures (Palmottu, El Berrocal)*, which are responsible for the processes of retardation and sorption in fractured media.

The absence of reliable data on the processes of solute immobilisation by *precipitation-coprecipitation* has prevented the consideration of such processes in safety assessment calculations. Analogue studies *(Oklo, El Berrocal, Palmottu, Koongarra, Poços de Caldas, Tono Mine, Needle's Eye, Broubster and Maqarin)* have provided evidence supporting the conceptual model of the process in the three sub-systems in which may take place under the evolution expected in a waste repository (canister corrosion products, cement degradation products and minerals filling fractures in the geosphere). Furthermore, the study of analogues has provided valid information on the most feasible phases controlling radionuclide behaviour in solution , and has allowed testing of different models and calculation codes of this process.

14. REDOX STATE AND REDOX FRONTS

The *redox state* of an aqueous system is defined by the concentrations of all oxidised and reduce species present in it. The characterisation of the redox state of a natural hydrogeochemical system is an intensive methodological and parametric task due to the multiple processes by which it is conditioned and to the specific problems involved in determining certain fundamental data, such as Eh.

Redox fronts are created at the boundary between two interacting systems with different oxidising environments. There is a close relationship between the redox state of a system and the propagation of redox fronts within it.

In a waste repository redox fronts may be generated in the near field and the geosphere for different reasons and at different moments in its evolution. The main causes of these redox fronts development are: 1) radiolysis of the groundwater in the area surrounding the canister, leading to the appearance of oxidising conditions close to it and to a change to reducing conditions at a greater distance; 2) the ingress of air and oxidising waters during excavation and construction of the repository and prior to its closure, which will lead to the oxidisation of the exposed rock surfaces; and 3) the arrival of oxidising recharge waters that, on seeping in, cause a drastic modification of the reducing environment of the repository, creating a front at the interface with the deeper reducing waters. The most significant of these causes is the radiolysis of the groundwaters.

The net movement of the redox front is controlled by the balance between the production of oxidising agents by any of the aforementioned processes and their consumption by the iron-rich materials in the engineered barriers (steel canister, backfill material such as bentonite) and the geological barrier. The interest in better understanding of the development and propagation of redox fronts springs from the fact that the solubility and speciation of many radionuclides (U, Np, Pu, Tc, etc.) are strongly influenced by the redox state of the system. Consequently, their potential migration or retardation changes radically on crossing a redox front.

The work performed on natural analogues (*Palmottu, Oklo*) for the characterisation of redox processes, which has been undertaken as an integral part of the study of water-rock interaction processes, has contributed to the refinement and improvement of the methodologies available for the in situ determination of the redox potential in groundwaters, and has provided information on different conceptual models relating to the characterisation and evolution of the redox state in natural systems.

The study of redox fronts in natural systems such as uranium deposits (*Poços de Caldas, Oklo, Cigar Lake*) may help to understand their dynamic behaviour in different scenarios. Analogues may provide information on aspects such as the description of the set of processes associated with the development of redox fronts (mineralogical and textural changes), the identification of the parameters influencing the evolution and migration of such fronts, their rate of propagation, the development and improvement of conceptual models of the evolution of redox fronts in systems controlled both by advection and by diffusion (clays), the identification of the limits or shortcomings of these models (consideration of the presence of organic matter, etc.), the redistribution or migration of elements of interest in the repository in a redox front and the phases that control their dissolution, etc.

15. COLLOID GENERATION AND TRANSPORT

Colloids are solid particles dispersed in waters and with sizes ranging from 1 micron to 1 nm. In view of their small size, these particles have a very large specific surface (10^3 m^2/g), this meaning that their physico-chemical behaviour is controlled by surface reactions and that they have a high capacity to adsorb dissolved elements.

Interest in the process of colloid generation and transport in a radioactive waste disposal system resides in the fact that radionuclides may be sorbed onto these particles being an additional transport mechanism, which under certain conditions may be more efficient (faster) than that of solutes. Furthermore, the radionuclides sorbed onto the colloidal phase may be excluded from possible retardation or retention effects. In addition, the sorption of radionuclides on colloids in the near field may give rise to concentration gradients promoting the leaching of radionuclides from the waste.

The processes of colloid generation in a repository system are not only those that occur in the natural geological medium of the repository and give rise to the colloids initially present in the groundwater; there are also additional sources of colloids associated with the degradation of the different engineered barriers (waste, canister, bentonite barrier and cement or backfill materials). The colloidal transport of radionuclides is a complex phenomenon whose net effects (the proportion of radionuclides transported through the geosphere that reaches the biosphere) depend on a large set of physical and chemical processes acting in a coupled manner with respect to which there are often significant knowledge gaps.

The study of colloids has been addressed in numerous natural analogues, such us uranium ore deposits (*Oklo, Cigar Lake, Palmottu, Poços de Caldas, Koongarra, El Berrocal*) and hyperalkaline media (*Maqarin*), as well as in anthropogenic analogues (*Nevada Test Site*). The main contribution made by natural analogues to the evaluation of the importance of colloid populations in radionuclide migration centres on the verification of conceptual models of colloid generation in engineered barriers, on the types, concentrations and stability of colloids in the geosphere, on their reactions with the radionuclides and the degree of reversibility of these reactions, and on the mobility and transport of colloid phases. They have also provided quantitative data on certain of the processes involved in the colloidal transport of radionuclides, such as for example the case of the correlations established between colloid concentrations and different factors determining their stability.

Although the observations carried out in natural analogues are not free from important uncertainties, as a result of the complexity of colloidal transport, they have contributed to increasing the understanding of a significant number of the processes involved and to the definition of fundamental directives for research, as well as to verification of the codes and increasing confidence in them.

16. MICROBIAL PROCESSES

There is evidence of the existence of microorganisms in the deep rock/groundwater interaction systems and, although these populations are not as large as those present in systems close to the surface, their effects on groundwater chemistry may be significant.

In the environment of a deep geological disposal system, it is possible that, in addition to the microorganisms that may exist under natural conditions, others may be incorporated during the facility construction phase. The feasibility and possibility of the subsistence of these microorganisms, which may act as catalysers for redox reactions and the formation of secondary minerals, will depend on the radiation fields created, the competition for water with the bentonite and the availability of sources of energy and nutrients. In crystalline formations, the existence of these organisms at great depths is possible thanks to the recharge of hydrogen as the main energy source and of CO_2 as the main nutrient.

A large number of bacteria consume oxygen during the process of organic matter oxidisation. This may have a beneficial effect during the initial phases of disposal, by contributing to the establishment of reducing conditions in the engineered barriers. However, microbial activity, with the associated production of organic complexing agents, may increase the solubility of the radionuclides in the near field and reduce their sorption capacity, possibly giving rise to the formation of gases and sulphides in the engineered barriers, thus reducing their effectiveness. Furthermore, microorganisms are mobile particles that may have a high radionuclide sorption capacity, possibly selective and irreversible, favouring the transport of radionuclides to the biosphere.

The study of microbial populations and the importance of the microbial processes present in different types of geological environments have been addressed in most of the more relevant natural analogues, mainly in uranium deposits (*Palmottu, Poços de Caldas, El Berrocal*) and in hyperalkaline media (*Oman, Maqarin*). The studies have focused on three fundamental aspects: 1) the types of microorganisms and their effects on hydrogeochemical conditions and radionuclide migration; 2) the tolerance of the microorganisms to the extreme radiation, temperature and alkalinity conditions that will prevail in the near field of the disposal system; and 3) the availability of nutrients and energy sources for the microorganisms.

17. COUPLED PROCESSES

The following processes are involved in the quantification of releases to the biosphere of radionuclides from a radioactive waste disposal system:

1. Thermal processes (T) associated with temperature variations. These variations may be natural in origin (geothermal gradient) or may be induced by the heat generated by the wastes during the initial phases of disposal.
2. Mechanical processes (M) relating to the properties and mechanical behaviour of the engineered barriers and the geological barrier.
3. Hydrodynamic processes (H) associated with fluid flow (gases and water).
4. Mass transport processes (Tr) or processes of solute and radionuclide transfer through fluid phases.
5. Geochemical processes (G) determining the geochemical evolution of the interstitial waters in the engineered and geological barriers, which ultimately condition the mobility or retention of radionuclides.
6. Radiological processes (R) associated with the radiation emitted by the radionuclides, which may affect the properties of the fluid (radiolysis) and heat generation.

From analysis of the evolution of a disposal system it may be deduced that the thermal (T), mechanical (M), hydrodynamic (H), solute transport (Tr), geochemical (G) and radiological (R) processes are intertwined, and should, therefore, be analysed together. Originally, the term "*coupled processes*" was coined in thermodynamics to refer to the coupling that exists between processes of a different nature.

The study of coupled and reactive transport processes has undergone considerable development in recent years, partly as a result of R&D work carried out within the framework of natural analogue projects and, fundamentally, due to R&D activities associated with waste disposal development.

The field in which the highest degree of development has been achieved in relation to coupled processes is that referring to the coupling of hydrodynamic processes (water flow), solute transport and geochemical processes, also known as reactive transport models. One of the most illustrative examples of the coupled nature of hydrodynamic and geochemical processes is redox front migration. Furthermore, the processes of interaction between groundwaters and cement give rise to hyperalkaline plumes (with very high pH values), the analysis of which requires that coupled reactive transport models be considered. Reactive transport models have been developed in certain analogue studies, such as *Oklo, Maqarin* and *Poços de Caldas*, and have allowed for: 1) the development and verification of new reactive transport codes; 2) study of the genesis and behaviour of redox fronts; and 3) the contrasting of different conceptual flow and geochemical models.

Anex C: Index of abbreviatons

Abbreviation	***Complete name***
AEA-Harwell	Atomic Energy Authority
AECL	Atomic Energy of Canada Limited
ANDRA	Agence National pour la Gestion de Déchets Radioactifs
ANSTO	Australian Nuclear Science and Technology Organization
ARAP	Alligator Rivers Analogue Project
BGS	British Geological Survey
BPM	Blind Predictive Modelling
BRGM	Bureau de Recherches Geologiques et Minieres
CEA/IPSN	Commisariat à l'Energie Atomique, Institute de Protection et de la Sûreté Nucléaire
CEC	Commission of the European Community
CIEMAT	Centro de Investigaciones Energéticas Medioambientales y Tecnológicas
CLMC	Cigar Lake Mining Corporation
CNWRA	Center for Nuclear Waste Regulatory Analyses
COGEMA	Compagnie Genérale des Matières Nucléaires
CRWM	Civilian Radioactive Waste Management
CSH	Calcium Silicate Hydrates. In accordance with the standard of the cement industry C=CaO; A=Al^2O^3; S=SiO^2; H=H^2O.
CSN	Consejo de Seguridad Nuclear
DGD	Deep Geological Disposal
EA	Environmental Agency
EC	European Commission
EC-NST	European Commission Nuclear Science and Technology
EMP	École de Mines de Paris
EPA	Environmental Protection Agency
ENRESA	Empresa Nacional de Residuos S.A.
ETSECCPB-UPC	Escuela Técnica Superior de Ingenieros de Caminos Canales Puertos de Barcelona – Universidad Politécnica de Cataluña
Gy	Thousands of millions of years
GSF-NWDR	Geological Survey of Finland- Nuclear Waste Disposal Research
GTK-YST	Geologian Tutkimuskeskus - Ydinjätteiden Sijoitustutkimukset
GRWP	General Radioactive Waste Plans
HLW	High Level Wastes
HMIP	Her Majesty's Inspectorate of Pollution
HREE	Heavy Rare Earth Elements; heavy lantanides
IAEA	International Atomic Energy Agency
IREE	Intermediate Rare Earth Elements; intermediate lantanides
ITGE	Instituto Tecnológico Geominero de España
JAERI	Japan Atomic Energy Research Institute
JNC	Japan Nuclear Cycle Development Institute
JRCCEC	Joint Research Center of the Commission of the European Communities
Ky	Thousands of years
LANL	Los Álamos National Laboratory
LBNL	Lawrence Berkeley National Laboratory
LLNL	Lawrence Livermore National Laboratory
LREE	Light Rare Earth Elements; light lantanides
My	Millions of years
NAGRA	Nationale Genossenshaft für die Lagerung Radioaktiver Abfälle (National Cooperative for the Disposal of Radioactive Waste)
NAWG	Natural Analogue Working Group
NEA OECD	Nuclear Energy Agency - Organisation for Economic Co-operation and development
NIREX	United Kingdom Nirex Limited
NRC	Nuclear Regulatory Commission
NUREG	U.S. Nuclear Regulatory Commission
OECD/NEA	Organisation for Economic Co-operation and Development / Nuclear Energy Agency
OH	Ontario Hydro
ONWI	Office of Nuclear Waste Isolation
PCS	Photon Correlation Spectroscopy
PNC	Power Reactor and Nuclear Fuel Development Corporation, Japan
PSI	Paul Schrreder Institute

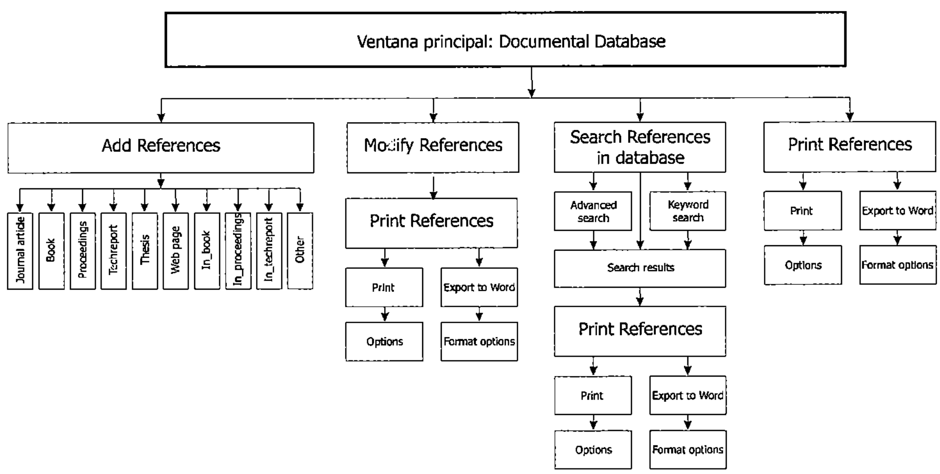
R&D	Research and Development
REE	Rare Earth Elements; Lantanides
SCK/CEN	Studiecentrum voor Kemenergie/Centre d'Etude de L'Energie Nucleaire
SEM	Scanning Electron Microscopy
SKB	Svensk Kärnbränslehantering AB (Swedish Nuclear Fuel and Waste Management Co)
SKI	Statens Kärnkraftinspektion (Swedish Nuclear Power Inspectorate)
SI	Saturation Index
SNL	Sandia National Laboratory
STUK	Center for Radiation and Nuclear Safety
SYNROC	Synthetic Rock
TEM	Transmission Electron Microscopy
TVO	Teollisuuden Voima Oy
UCM	Universidad Complutense de Madrid
UDC	Universidad de La Coruña
UKAEA	Environmental Agency of England and Wales
UKDoE	U.K. Department of Environment
UPC-CSIC	Universidad Politécnica de Cataluña-CSIC
USA	United States of America
US EPA	United States Environmental Protection Agency
US-DOE	United States Department of Energy
USGS	United States Geological Survey
USNRC	United States Nuclear Regulatory Commission
UZ	Universidad de Zaragoza
VTT	Technical Research Center of Finland
XRD	X-Ray Diffraction.
y	Years
YJT	Nuclear Waste Commission of Finnish Power Companies
WIPP	Waste Isolation Pilot Plant

Anex D:
Documentary database

The documentary database attached hereto in CD-ROM form has been developed specifically for this project, with a view to compiling all the bibliographic references consulted or quoted during the project, part of which appear in the bibliography of this document. The database currently has some 1,800 entries.

Given that the project requirements regarding organisation and search for and inputting of data did not correspond to any commercial programme, a specific Access" database has been designed for storage of the information compiled during the project. The technical documentation on the way in which the database is organised internally is included in the User Manual on the CD-ROM. Figure DB-1 summarises the hierarchy of the DDB windows, while DB-2 shows the main window.

Figure DB-1. Organisation of the Documentary Database developed for the project.



The main function of the documentary database (from hereon the DDB) is the systematic and homogeneous storage of all the bibliographic documentation relating to the study of natural analogues and the safety assessment exercises consulted. In order to increase the usefulness of the information stored, the DDB incorporates not only the bibliographic data required to correctly quote all the bibliographic references that appear in the documents associated with the project, but also a series of additional fields describing in detail the type of information contained in each reference.

With a view to meeting the requirements regarding homogeneity in the inputting to the database of references by the various independent working groups, 10 different types of bibliographic references have been defined, as follows:

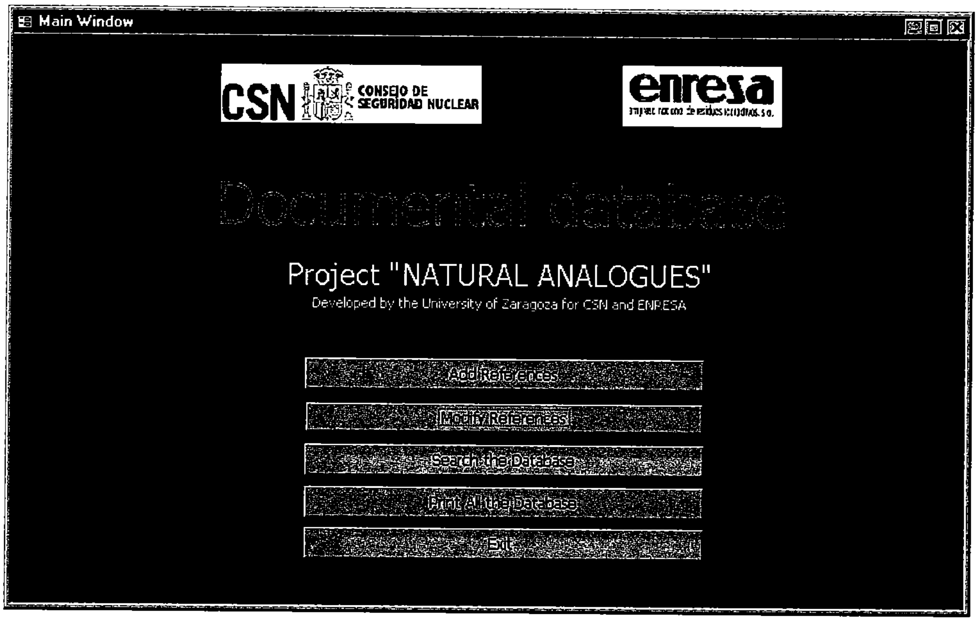


Figure DB-2. Main DDB window.

- **Article:** work published in a journal.
- **Book:** text book, subject-specific work, series of volumes or complete volume of a bulletin.
- **Proceedings:** proceedings of a conference, congress, meeting, workshop, etc.
- **Technical report:** work published by a university, research or regulatory organisation, company, etc.
- **Thesis:** doctorate thesis, or thesis for graduation, a master's degree, etc.
- **Electronic document:** this type of reference serves for all electronic references that may be consulted on line, both via Internet and HTML documents, and via telnet, ftp or gopher. Publications on CD-ROM are not included unless they can only be consulted on line.
- **In book**: a chapter or section, with its own authors, within a book type reference.
- **In proceedings:** an article (or summary) included in the proceedings of a conference, congress, workshop, etc. (proceedings type reference)
- **In technical report:** an article, chapter or section, with its own authors, within a technical report (technical report type reference).
- **Others:** a reference not assigned to any of the previous categories.

Each type of bibliographic reference has its own information requirements, these being included in the fields that appear in the windows corresponding to each type of reference. In these windows, the fields that must necessarily be filled in are shown with a white background, while those that are optional are shown in grey. In this way, there is a guarantee that all the references have a minimum amount of common information, regardless of the working group that has included the reference in the database. As an example, figure DB-3 shows the window corresponding to the "Technical Report" type reference. The DDB has been designed in English, for this reason, all the information that appears in the figures shown here, extracted directly from the application, appears in this language.

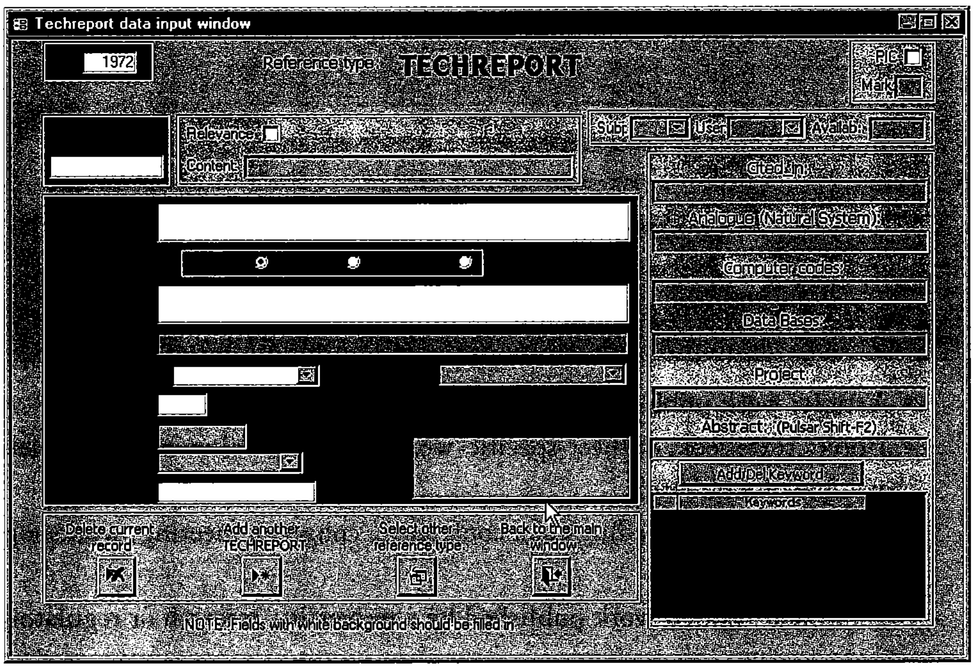


Figure DB-3. Window for the inputting of information for a "Technical Report" type reference (Techreport in the figure). The strictly bibliographic fields are framed in green, while those shown with an orange background are for additional information. In all cases, a white background indicates that the field must necessarily be filled in, while a grey background indicates that this is optional

There are 41 field in total in the DDB, grouped in three categories: (1) control fields, used especially for the internal organisation of the database; (2) bibliographic fields, which include the information necessary for each reference to be correctly quoted; and (3) information fields which, as the name indicates, include information on the contents of each document. In Figure DB-3, the control fields are included in grey areas (upper left-hand section), the bibliographic fields in green and the information fields in orange. The name and definition of each of the 41 fields may be found in the User Manual.

In addition to incorporating bibliographic references, the DDB allows for the following:

- Modify references. This option allows edition and modification of previously input references.
- Search for references. This option allows searches to be carried out in relation to any field of the database.
- Print references. This option makes it possible to print (1) the entire database, (2) the references selected by an automatic search process, (3) manually selected references, (4) the reference displayed on screen.

The DDB has a powerful reference search tool organised on three levels:

- Simple search: the user inputs a word or a literal sentence and the database takes charge of the rest.
- Advanced search: the user inputs one or several words or literal sentences linked by logic operators and the search is performed in specific fields selected by the user.
- Search by key words. the DDB has a "closed" set of key words organised in seven groups (sub-systems, materials, parameters/data, processes, modelling, safety assessment and geological features). These key words are added to each reference when it is incorporated into the database. Working with a limited set of key words allows for the easy recovery of references sharing one or several key words during searches.

A final feature to be underlined is the fact that the DDB makes it possible to print the references contained in the databases in different formats. This printing may be accomplished from the database itself, using a series of specifically designed forms, or from Word, exporting the references selected to a document created in this word-processing tool. The advantage of this option is that the references may be printed directly with the definitive format in which they appear in the bibliography of each of the project documents.