

Radioactive waste management: the role of CIEMAT in the Spanish and European R+TD programs for radwaste disposal

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I Introduction

Waste production is a direct consequence of welfare and industrial development. The waste volume produced and the hazards some of them represent have turned out in an appeal to rationale production, recycling and the storage of the final products under safe conditions.

Radioactive wastes have created the greatest social sensibilization, even though they must come from activities closely linked to welfare and development, such as nuclear energy production, industrial and diagnostic or therapeutic medical uses.

The "social" positioning on radioactive wastes is sometimes on the edge of rationality. The military use of nuclear energy, the lack of knowledge dissemination and biased information are giving place to reactions that go beyond the more elementary social and common senses.

Radioactive wastes exist and therefore, they must be adequately managed (recycling or transmutation) or stored, in a safe way. Their potential hazard and long life of some of them impose on our generation the moral obligation to avoid passing the problem to future generations.

The possibility of a permanent storage for high level radioactive wastes on deep geological formations have been intensively studied during the past twenty years in many countries. These studies are generating the knowledge and technical capabilities needed for building and managing a final storage site.

The exercises on long term performance assessment carried out up to date for real or hypothetical scenarios, defined from the knowledge of natural systems, give a reasonable level of confidence on the long term safety of the storage system.

The long term (10^4 - 10^6 years) assessment is the problem arising more doubts on the storage system and decreasing its confidence level. On that sense, the studies on natural analogues have a great relevance for the conceptual contributions, qualitative and quantitative, that they offer to the long or very long term evolution of natural processes and materials.

Any of the options that might be finally chosen for the final waste management, will finally end on the necessity of a definitive storage site.

The design of a Radioactive Waste Geological Storage is based on a multibarrier system concept (Figure 1). This is established so every barrier will provide mutual protection to the others. In the case of failure of any of the barriers all the others will act to solve this failure.

The barriers considered are the spent fuel matrix, the canister filling, the storage canister itself, the bentonite seal, the gallery backfill and the host rock. The ultimate function of the multibarrier system is to avoid or to retard as long as possible the arrival of the radionuclides to the biosphere with the aim of maintaining a dose to the public below the levels of the natural radioactive background for that host rock.

Objectives

The general objective of the Hydrogeochemical Site Characterisation Program of CIEMAT is to study the Long Term Behaviour of the Components of the Radioactive Wastes Storage

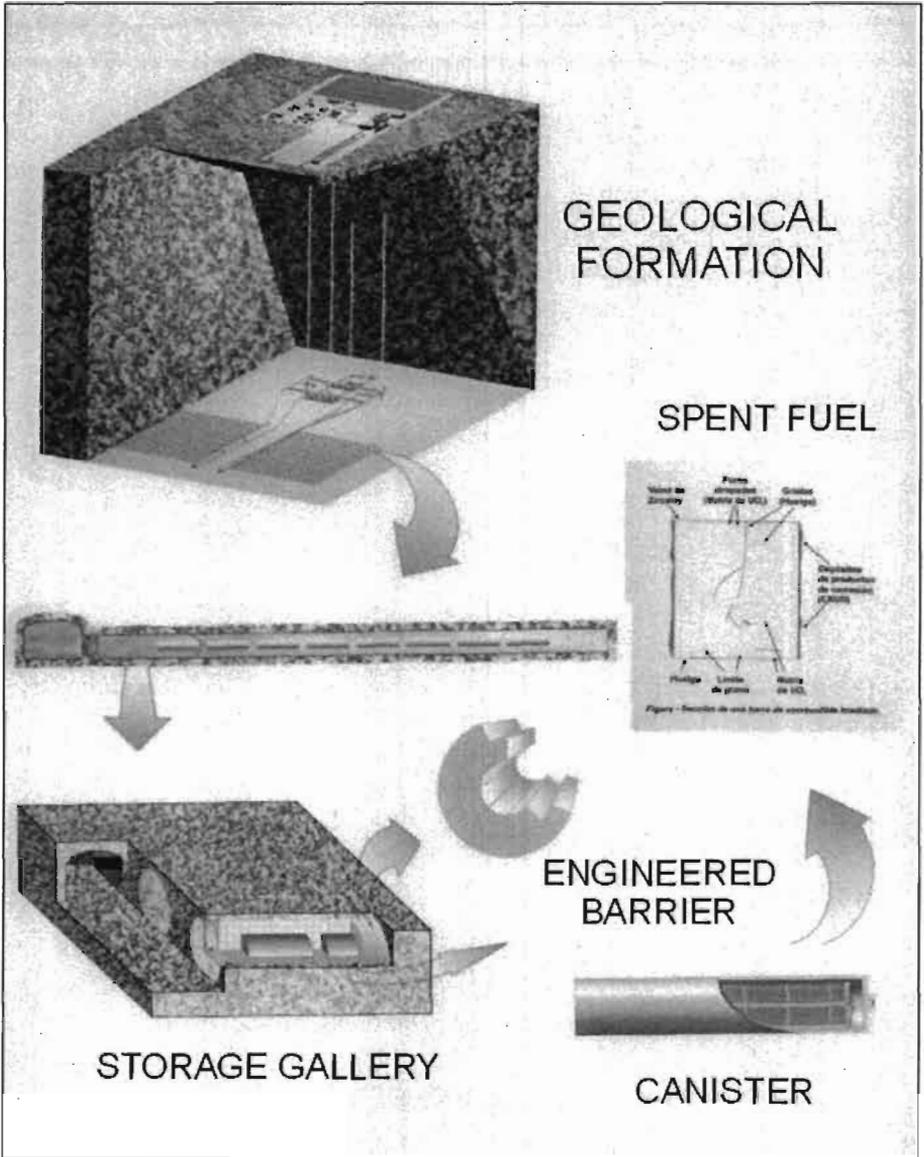


Figure 1
Conceptual design of a radioactive waste Deep Geological Storage system. Multibarrier system option.

Systems. This objective is focused on the study of the processes that determine the behaviour of the backfill and sealing materials, of granitic and clayish rocks and of natural analogues, in order to provide a sound scientific understanding of their role.

In the past four years, this objective has broadened to the study of spent fuel analogues, waste package filling materials and canister corrosion products. For all the materials considered, the most relevant aspect taken into account is their potential capability to decrease radionuclide mobility towards the environment, as they interact with the multibarrier system. This retardation or retention mechanisms may be of physical or chemical nature or a combination of different processes.

Results

The key processes controlling the long term behaviour and safety of a site act in a coupled way in the system as a whole, presenting a specific relevance for each of the barriers considered. The general objective of the Program is to identify those processes, to assess the manner in which they act and to provide a sound scientific understanding of their role. The results being obtained, presented below, are oriented to fulfil this general objective.

Long term stability of the spent fuel

The key process is the solubilization of the fuel matrix, the release of uranium and fission products in an almost dry, reducing, neutral and moderate temperature environment.

The study is carried out on natural UO_{2+x} (Uraninite and Pitchblende) as a fuel analogue, that has been exposed for millions of years to the evolution of a natural geochemical environment. The uraninites and pitchblendes studied correspond to the ore deposits of Oklo (Gabon), Palmottu (Finland) (Perez del Villar, 2000), Margnac and La Crouzille (France), Sierra Aabarrana, Los

Ratones and Mina Fe (Spain). They have been dated between 2000 (Oklo) and 50 Myears (Mina Fe). The more relevant results are the following:

for all the ore deposits it has been considered that most of the UO_{2+x} (Uraninite and Pitchblende) is original, even if it has undergone hydrothermal and/or meteoric weathering processes;

all uraninites and pitchblendes are affected by a relatively intense microfissuration that would had enhanced the weathering processes; weathering has occurred mainly by pseudoisomorphic replacement of the original mineral specie (Uraninite and Pitchblende) by the new Uranium mineral without any apparent transport involved;

the kind of weathering depends on the geochemical environment in which it has happened. In high silica reduced environments Coffinite (U(IV) silicate) has replaced Uraninite or Pitchblende (Oklo, Mina Fe, Margnac and La Crouzille). In a pegmatitic oxidising environment such as Sierra Albarrana the gummities (complex hydrated U(VI) oxides) have partially replaced Uraninite;

all the weathering processes mentioned imply the mobilisation of part of the Uranium from the Uraninite or Pitchblende. The maximum mobilisation happens in the weathering to Uranyl phosphates. The leaching rate and the Uranium concentration in the rock interstitial water in contact with the ore do not vary significantly from those obtained in Uraninite leaching (10^{-9} – 10^{-7} g.yr⁻¹ for weathering ages of 50 and 1 Myears respectively). Those values imply Uranium concentrations of 10^{-7} to 10^{-6} M in the pore water in contact with the mineral;

in the natural reactor of Bangombé (Gabon) the Uraninite is impoverished in ^{235}U due to the fission reaction. However the $^{234}\text{U}/^{238}\text{U}$ and $^{230}\text{Th}/^{234}\text{U}$ ratios, with a unity value, indicate that Uraninite has remained unaltered for at least the last 1.7 Myears. The ^{206}Pb has not been mobilised over the last 740 Myears and the Zr, (Tc), Ru and ^{138}Ba have remained in the Uraninite where the fission occurred. However, Cs and Sr have migrated from that mineral (Gauthier-Lafaye *et al.*, 1999).

From the results showed above one of the main considerations is that the U mobilisation from natural minerals reflects the behaviour of spent fuel, and the resulting rate of U release from the waste

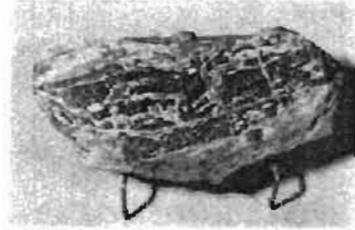
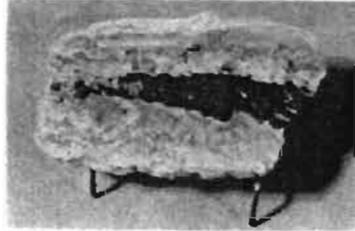
package will be very low with an almost immediate retention on the materials surrounding the waste. This should constitute an encouraging aspect concerning long term radionuclide mobility. In Figure 2 (Roubault, 1962) are shown some of the weathering processes affecting Uranium minerals.

■ Figure 2
Examples of Uranium
minerals weathering
from different
formations.

Top: Henriette mine
(pitchblende oxidized
to autunite).

Middle: Brugeard-
Ouest mine
(pitchblende
to gummite).

Bottom: Margnac II
(pitchblende to yellow
gummite).



Canister filling materials

The function of these materials is to contribute to enhance the canister safety. It is planned to place an additional barrier, taking advantage of the small free space between the fuel elements and the canister inner wall, and several candidate materials are being con-

sidered according to the specific function assigned to them. In ENRESA's conceptual design this barrier would be made of borosilicate glass beads, but the study of other materials that retard the escape of Iodine (^{129}I) and Chlorine (^{36}Cl) from the canisters is also being carried out. Based on the laboratory data (sorption, diffusion, migration) is expected that these two isotopes, not retained in the clay seal and the geological barriers, will contribute the most to the long term average estimated dose, for a million years, in the safety and performance assessment exercises done for a deep geological repository.

Phosphates (apatites) and zeolites have been studied as alternative materials, because they are the natural materials with the higher geochemical contents of Iodine and Chlorine. Apatites also incorporate in their structure a large number of metals and zeolites have a very high ionic exchange capacity.

Partial conclusions from this study indicate that both phosphates and zeolites might contribute to retard the migration of Iodine and Chlorine from the waste canisters. The distribution coefficients for Iodine reach values of 20 ml.g^{-1} in comparison with the zero value assigned to this element in previous Performance Assessment exercises (Stenhouse, 1995). Although the sorption mechanisms are not well defined it has been confirmed that between 75 and 90% of the Iodine remains retained in the solid phase after desorption with a reference granitic groundwater. Pre-treatment procedures to decrease the natural salt content, the homoionisation with reactive cations or the increase of specific area enhance the reactive capacity of these materials (De la Cruz *et al.*, 1999).

Both phosphates and zeolites have shown to be structurally stable under the thermal treatment conditions that they have undergone. The textural modifications detected, increase of surface and rearrangement of the pore distribution seem to stabilise at 150°C after two weeks. However, at 200°C over a year, most zeolites showed an structural alteration between 10 and 30% while for phosphates is negligible. The densities obtained by uniaxial compaction and the granulometric distribution of the natural materials indicate the feasibility of obtaining high density granulates of size above 1 mm. This type of preparation would facilitate the manipulation of the material avoiding the formation of a dusty environment during handling.

Phosphates most unfavourable characteristic is its low thermal conductivity, in the range $0.2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. It would be necessary to adjust this parameter through treatment or mixtures with other materials.

Behaviour of the canister materials

In the Spanish conceptual design of the Deep Geological Storage the canister material will be constituted by carbon steel or copper alloys (ENRESA, 1997 & 1999). In the case of steel there exist both localised and general corrosion phenomena, with an estimated mean rate of about 6.5 mm/ year, (Hernandez Benitez, 1999). Amongst the dominant corrosion products observed in 18 months experiments under saturated bentonite conditions with high HCO_3^- concentrations are Siderite ($\text{CO}_3\text{Fe}(s)$), Magnetite (Fe_3O_4) and Haematite (Fe_2O_3) (Azkarate *et al.*, 2000).

These studies have been carried out for gathering data on canister behaviour, under different corroding conditions. These results will be implemented in the framework of the Geochemical Mock Up test to be carried out along with the FEBEX II Project.

Behaviour of the clay barrier

In relation with this research line has been carried out the FEBEX (Full-scale Engineered Barriers Experiment) project (ENRESA, 2000). This project aims at demonstrating the technical feasibility and studying the behaviour of near-field components of a high level radioactive waste repository in crystalline rock. It consists of an "in situ" test (Grimsel), a "mock-up" (CIEMAT) test and a series of complementary laboratory tests as well as modelling work. In Figure 3 are shown the two experimental set-up for both tests. (Huertas *et al.*, 2000).

The clay barrier functions are the following: to facilitate the transfer of the heat generated by the waste, to act as a low hydraulic conductivity medium between the canister and the host rock, to seal the building discontinuities and fissures of the host rock, to retard the diffusive transport phenomena of the released radionu-

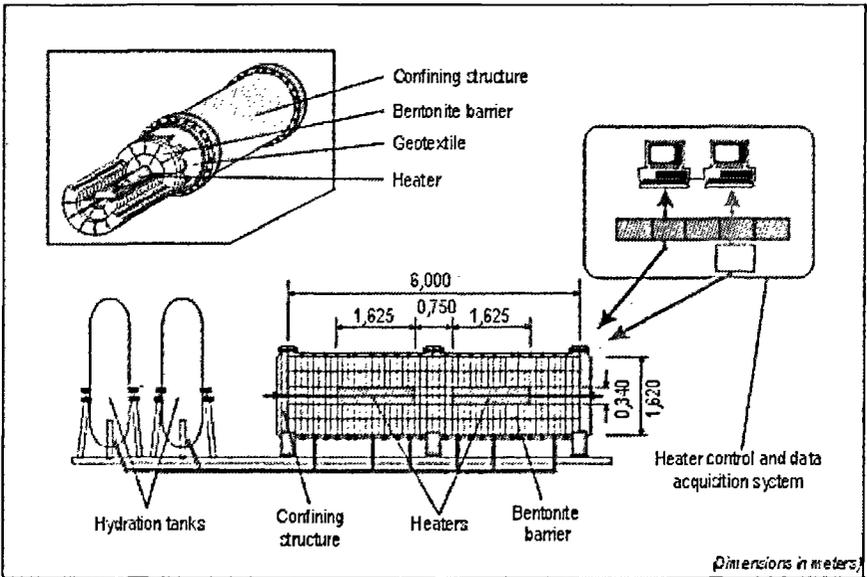
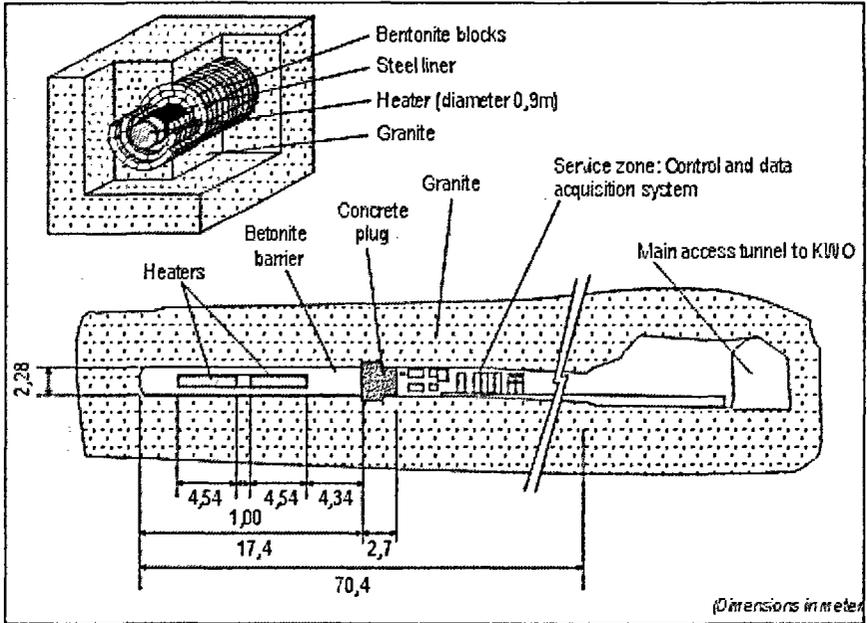


Figure 3

Experimental set-up for FEBEX experiments.

Top: *In situ* experiment at Grimsel Test Site (Switzerland).

Bottom: Mock-up experiment at CIEMAT facilities (Spain).

clides and to provide mechanical support to the host rock protecting the canister of possible shear movements of the geological formation.

The thermal gradient generated by the radioactive decay and the hydraulic gradient of the rock start the heat flow and water transport in the clay barrier. These two processes give place to the mechanical reaction of the bentonite and the chemical interactions in the mineral/water complex system that form the barrier. The four main basic processes (thermal, hydraulic, mechanical and geochemical) function in a coupled manner, producing new mineral phases and gradients and intrinsic modifications of the porous medium, until the general equilibrium of the system is reached.

Therefore, the long term performance assessment requires a deep knowledge of the short term behaviour, identifying and understanding the governing processes and parameters. This is necessary to prepare a conceptual model as a basis of a numerical model, calibrated for natural and experimental evidences, that will guarantee its long term predictive capacity.

The research work on the behaviour of clay barriers is being carried out mainly with the spanish reference bentonite (Serrata de Nijar, Almería) through laboratory experiments and large scale experiments in the "mock-up" (CIEMAT) and "in situ" (Grimsel, Switzerland) tests, that are becoming essential to calibrate the thermo-hydro-mechanical and geochemical coupled numerical models (ENRESA, 2000).

The most relevant results, for the basic processes and function of the barrier previously mentioned are the following:

Thermal behaviour

In a very short time the clay barrier will reach a quasi-stationary state and a very regular temperature distribution, with extreme values of about 100°C in the canister interface and 35°C in the contact with the host rock. The expected mean gradient is on the range 1°C.cm⁻¹.

The heat transfer takes place by convection through water and water vapour phases and during the phase change processes produced by cooling as the distance from the heat source increases.

The characteristic empirical functions of the clay barrier have been set, relating the thermal conductivity with the hydration state, the specific heat with temperature, and the deformation also with the temperature (thermal linear expansion coefficient).

The predictive capacity of the numerical modelling is very good (Gens *et al.*, 1998).

Hydraulic behaviour

The barrier hydration takes place in a radial and centripetal fashion with a relatively homogeneous distribution of relative humidity and presumably of the saturation degree. The construction discontinuities (block joints), the structural heterogeneities and of water supply from the host rock do not seem to produce significant anomalies in the process.

In relatively short time periods the peripheral external annulus, of centimetric depth, reaches a near saturation while the inner annulus in contact with the canister dries out. However, the hydration rate reaches extremely low values, due to the low permeability, and the suction capacity decrease, due to hydration, of the clay. Therefore the clay saturation of the clay barrier will be a process taking several decades. For the mock-up experiment it has been predicted a 12 years time to reach a saturation value of 95%.

The hydraulic processes acting are water flow, water vapour diffusion, gas flow, air dissolution in water and dissolved air diffusion.

Up to this moment, to achieve the best fitting level between numerical predictions and results for the “mock-up” (CIEMAT) and “in situ” (Grimsel) experiments, it has been indispensable to obtain characteristic functions for bentonite; such as permeability as a function of dry density (ρ_d), of the saturation degree and temperature; the retention curves expressing the relationship between suction and the saturation degree at free and constant volumes, or the tortuosity coefficient and gas permeability as a function of dry density and saturation degree.

Although the predictive capacity for the barrier hydration process offers a great degree of confidence, it will be necessary in the next years to optimise the adjustments of the characteristic functions and to deepen in the study of microstructural modifications and their impact on the characteristic parameters of the barrier (UPC-DIT. 1999).

Mechanical behaviour

Processes governing the mechanical behaviour of the barrier are the thermal expansion and the bentonite porosity variations depending on the stress, suction (saturation degree) and temperature fields.

Bentonite has a strong avidity for water, and reacts mechanically, in such a way that in an open system regime volume can increase up to 200%. In a closed system regime this process can develop a swelling pressure during hydration that exponentially depends on the initial dry density (Lloret *et al.*, 1999). For the final dry density ($\rho_d = 1.60 \text{ g.cm}^{-3}$) of the clay barrier the swelling pressure is about 5Mpa.

The first mechanical reaction of the clay barrier is the sealing of the bentonite-rock interface annulus and of the discontinuities between blocks where the free water arrives. From this point the development of pressure is a function of the hydration state, that homogeneously progresses in a radial and centripetal way as indicated: Hydration implies porosity modifications and therefore of a considerable amount of bentonite characteristic parameters (water and gas permeability, thermal conductivity, diffusion coefficient, swelling pressure, etc.).

Prediction of the barrier mechanical behaviour has an acceptable adjustment with experimental results. However, is not yet possible to rigorously analyse the predictive capacity of the models because the total pressure sensors response for the mock-up and "in situ" experiments is under local stress adjustments depending on location and because the hydration state has not produced significant responses for many of them.

The bentonite mechanical behaviour study has followed an extensive experimental programme from which the deformation parameters as a function of effective stresses, suction and temperature. This has allowed the development on the bentonite thermo-plastic-elastic behaviour model.

There have been determined the resistance parameter and the elastic shear modulus and the functions relating swelling pressures with dry density and suction have been adjusted, as well as the deformations under load.

Geochemical behaviour

The geochemical evolution of the clay barrier is produced by the heat flow from the canister and the water flow coming from the rock (granite). At short and medium term the effect of the granitic water chemistry on the geochemistry of the clay barrier is not significant.

The bentonite minor components solid phases that govern the geochemical processes by interacting with the hydration water are Halite Gypsum, Dolomite and Chalcedony. The dissolution kinetics of other mineral components is so slow that they would only affect the system on a very long term.

Experimental data indicate that during the hydration transitory state Cl^- and SO_4^{2-} are transported by the hydration front. As a conservative ion, Cl^- moves accordingly to the effective porosity determined with HTO, while SO_4^{2-} movement is retarded and its concentration levels in the advancement front must be regulated by its equilibrium with Gypsum. Precipitation of Anhydrite occurs in contact with the heat source.

Bicarbonate concentration is controlled by dissolution/precipitation of Calcite and it remains almost practically constant in time for the duration of the experiments. Na^+ , Ca^{2+} , Mg^{2+} and K^+ show a similar trend of mobilisation and concentration around the heat source. However, the analyses of their stoichiometric relationships with their corresponding anions and of the composition of the bentonite ionic exchange complex show that during transport the exchange reactions with the smectite have occurred. The general trend indicate that the smectite is enriched in Ca^{2+} and Mg^{2+} released by carbonates dissolution and loses Na^+ and K^+ . This is a very favourable aspect because it would exclude the possibility of smectite illitisation.

The ionic strength of the interstitial water varies from 0.07 M in the inlet zone to 1.2 M at the hottest zone, during the hydration transitory regime.

Bentonite buffers pH at values close to 8 and the Eh must evolve towards negative potentials when the residual Oxygen from building and emplacement is consumed.

Diffusion is the dominant transport process once saturation is reached. This fact, together with the concentration gradients created

by precipitates dissolution gives place to salt redistribution and their leaching by the granitic waters (Cuevas *et al.*, 1998).

The estimation of the barrier interstitial water evolution time seem to indicate that the bentonite water salinity will be maintained at a level similar to the initial up to 5000 or 10000 years. So, this time interval will be higher than the estimated lifetime of the canisters.

The geochemical processes described do not seem to affect the basic properties of specific area, hydraulic conductivity and swelling pressure of bentonite, according to the measurements performed after the thermal-hydraulic treatment.

The clay barrier is an efficient filter for the colloids that can be generated as a consequence of fuel alteration. On the other hand, the high ionic strength of the interstitial water and the pH, close to the point of zero charge (PZC) of many interesting colloids, provokes their immediate aggregation. A different situation occurs at the clay barrier/granite interface, where bentonite and silica colloids can be present and are very stable under the physicochemical conditions existing in the granitic formation.

The radionuclides will migrate through the barrier by diffusion, and their movement will be retarded due to sorption reactions with the clay surface (adsorption/desorption, precipitation, ion exchange, colloid filtration, etc.). Diffusion data obtained in laboratory experiments with compacted bentonite at the barrier density vary from 10^{-11} m²/s for tritiated water (HTO) to 10^{-14} m².s⁻¹ for Re(VI), as ReO₄²⁻, and Tc(VII), as TcO₄⁻. Sorption experiments performed on loose bentonite (batch method) have demonstrated that Eu(III), Nd(III) and Th(IV) result completely retained by bentonite; Cs, Sr and Co have high values of the distribution coefficient, K_d (Yllera *et al.*, 1998); U(VI) is slightly retained, and Re(VI), Tc(VII) and I⁻ are not sorbed at all (Garcia *et al.*, 1999A). In Figure 4 are shown some of the different type of diffusion experiments carried out in clay materials.

It is clear that there it has been a great advance in the knowledge of the integrated behaviour of the clay barrier. There are still some uncertainties, that might be solved in the next research programme. On the other hand, the geochemical models have not been calibrated for the evolution of the key parameters in the large scale experiments. With this aim it has been proposed the establishment of a geochemical mock-up experiment.

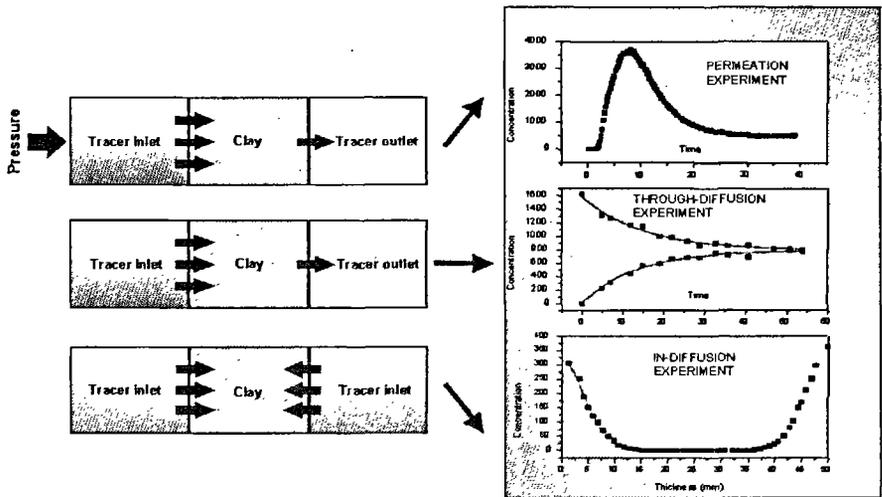


Figure 4
Different diffusion experimental set-up used
for geological material characterization.

Geosphere behaviour

The role of the geological formation for site emplacement is the chemical and mechanical protection of the other barriers and to retard the radionuclide transfer to the geosphere. This is achieved as a consequence of the structural, hydraulic and geochemical properties of the geological barrier.

According to the above mentioned functions, the study of the geological formation is mainly focused on the obtention of a structural model of the discontinuity network, acting as potential flow paths; a groundwater flow model, a geochemical rock-water interaction model and a transport model.

The program mainly addresses the geochemical model, and provides the basic parameters related to the radionuclide transport. In the current program, work is being carried out in granitic, clay and natural analogue formations.

Some of the most remarkable results are presented in the following section:

Granitic formations

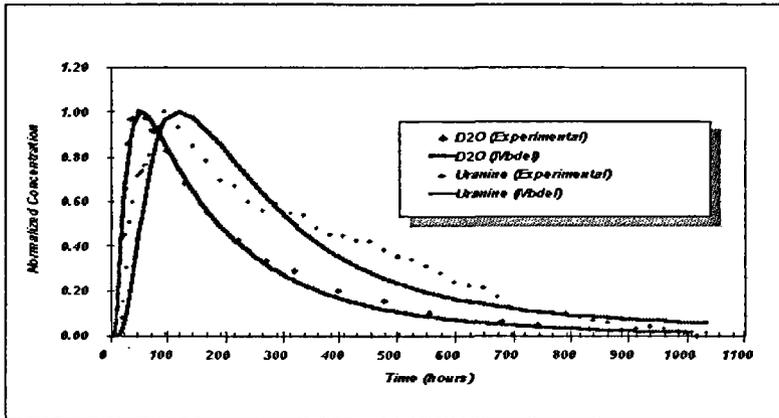
The first research work in this field made in Spain was started in 1989 with the El Berrocal Project (Rivas *et al.*, 1997). The project was carried out in a granitic pluton that hosts a number of U mineralisations, which were mined over a period of several years, although the mines were definitely abandoned by the late 1960's. The project was an integrated international exercise in geological, geochemical, and hydrogeological characterisation and had the aim of understanding and modelling the past and present-day migration processes that control the behaviour of distribution of naturally occurring radionuclides in a fractured granitic environment.

Work at the El Berrocal during the project was extensive. It included geological mapping, structural analysis, mineralogical, litho-geochemical and hydro-geochemical investigations, considering both stable and radioactive isotopes, colloid and microbial studies. Several methodologies and instrumentation were developed for *in situ* tracer tests, application of downhole probes, hydrogeological characterisation and coupled flow and transport modelling. The fieldwork was supported by a wide laboratory program, which generated valuable data for the interpretation of those phenomena observed at natural scale. In Figure 5 are shown the experimental and modelled results for the first tracer test performed between boreholes S-13, S-15 and S-2.

This research constitutes the basis for implementing a methodology that is used for characterisation of crystalline formations affected by U mobilisation processes. Specially in the case of restoration programs for abandoned mines, or mill tailings environmental impact studies (Project Ratonces, Project MATRIX (Perez del Villar *et al.*, 2000), etc.).

The deep groundwater (\cong 500 m) of the reference granite (Gomez *et al.*, 1999) used for ENRESA 2000 Performance Assessment exercise has a sodium/bicarbonate composition presenting a residence time of about 20,000 years. The pH is regulated by the equilibrium with carbonates with neutral or slightly alkaline values. The measured redox potential is regulated by the reaction of the redox pair Ankerite (15% Fe)/Goethite presenting a value of -175 mV.

The processes controlling the concentration of the major components of the groundwater are dissolution/precipitation of carbon-



E El Berrocal Project: results of the *in situ* Tracer Test

I Figure 5

Tracer test modelling and experimental results in El Berrocal granite. First tracer injection (D_2O and Uranine)

ates, the ionic exchange (Ca/Na and Mg/Na), montmorillonitisation and precipitation of goethite-like iron oxide.

Given the redox potential of this groundwater, uranium concentrations are below the detection limits, even in the presence of residual Uranium mineralization existing in the granite. Those characteristics are considered as highly favourable to minimise the radionuclide mobilisation processes.

The sorption parameters obtained, under oxic conditions, indicate that more than 90% of Cs, Co and Eu and about 30% of Sr and Se(IV) are retained by the granite minerals, while Tc is not retained at all (Garcia *et al.*, 1999b).

Clay formations

The first studies on clay formations for their characterisation to act as geological barriers have been carried out.

The Spanish reference clay formation has a thickness close to 280 m., being very homogeneous. These sediments have a lacustrine origin,

and are limited, at top and bottom, by fluvio-alluvial sediments and lacustrine-palustrine carbonates, respectively.

Illite and smectite represent more than 80% of the mineralogical composition of the formation. The minor minerals that regulate the groundwater characteristics are Calcite, Dolomite and Gypsum. The organic matter content is near 1.5%.

The values of total cation exchange capacity, specific area and hydraulic conductivity are coherent with the mineralogical composition. As a whole, these values provide this formation with a high index of favourability as a site emplacement host rock.

The interstitial waters have a sodium/sulphate nature with pH values around 7.5. The geochemical modellisation is being performed.

The radionuclide retention capacity is higher than for the reference granite. For Cs, Sr, Co and Eu the value of the distribution coefficient increases in one or several orders of magnitude.

In the Mt. TERRI Project (Thury & Bossart, 1999), intercomparison exercises have been carried out for the extraction and analyses of the interstitial water, the geochemical modelling and the determination of diffusion coefficients $1.3\text{-}2.3 \cdot 10^{-11} \text{ m}^2\cdot\text{s}^{-1}$ for HTO and $2.3\text{-}3.0 \cdot 10^{-12} \text{ m}^2\cdot\text{s}^{-1}$ for I⁻ (Yllera *et al.*, 2000). All this work has been carried out with the Mt TERRI Opalinous Clay formation, a very consolidated clay of marine origin.

As a consequence of the start of this research line on bentonite engineered barriers, a participation in the GMT Project exists, financed by Japan, to study the gas transport in the reference Japanese bentonite. Also, in the framework of the FEBEX II Project, gas transport essays in the “in situ” (Grimsel) and in the “geochemical mock-up” experiments have been proposed.

Radionuclides behaviour in the geosphere (natural analogues)

In the host rock of the Bangombé (Gabon) reactor, the Uranium has its natural isotopic composition, thus, indicating that the Uranium from the reactor has practically not been mobilised. Neither are there evidences of isotopic data that indicate Sr or Mo retention.

The fissionogenic Ba and the Ba decaying from Cs have migrated from the reactor area and have been retained in the phyllosilicate-rich zones. There is also evidence of Zr migration and retention in the host rock, although it has not been possible to determine the minerals where this element was incorporated.

Based on the fluid inclusion analyses, the different fluid phases and the formation temperatures of the mineralization and weathering processes identified in the host rock of the natural reactor are known.

In the MATRIX project (Perez del Villar *et al.*, 2000), once the structural framework of the Boa fault was established, and knowing the weathering and Uranium distribution in the exploitation front of that fault, the drilling of four boreholes was carried out to study the Uranium mobility as a function of the oxidising-reducing environment.

1 Conclusions

In Spain, for the last fifteen years, it has been studied the problem of radioactive waste Deep Geological Storage. In that time it has been achieved a great advancement on the understanding of components and processes that must initially regulate the behaviour of the storage system. International research has progressed too in the same direction.

The research on the problems arisen from the storage activity, gives place to new questions on the system. Despite this situation, the solution of the problems from the scientific or technological point of view in which they are approached, it produces a reasonable confidence on the storage safety and viability. This consciousness is somehow supported by the Safety and Assessment exercises (PA) carried out on several geological formations.

Several important uncertainties exist, and their resolution will come through the unification, in experiments under natural conditions at a real scale, of the basic knowledge generated on the laboratory experimental programs and the theoretical knowledge developed for the modelling of natural processes.

On the other hand, all the methodologies and knowledge obtained in the field of radioactive wastes is applicable to any of the environmental fields that appear every day. In that way CIEMAT has participated in several projects involving both natural radionuclides of even metallic contaminants. One of these cases was the study of the contaminated soils from the Aznalc6llar area, after a mining slurry spill at the end of April, 1998.

Aknowledgements

The different research programs reflected in this document have been carried out in the framework of ENRESA R+D programs since 1988, and of the consecutive Framework Programs of the European Union.

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