



WASTE AND DISPOSAL: RESEARCH AND DEVELOPMENT

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Background

The primary mission of the Waste Disposal programme is to propose, develop, and assess solutions for a safe and acceptable disposal of radioactive waste. Geological disposal into clay formations is considered a realistic and adequate solution for the final disposal of high-level and long-lived radioactive waste. For short-lived low-level radioactive waste, as well shallow land burial facilities as deep repositories are examined in Belgium.

In Belgium, deep geological burial in clay is the primary option for the disposal of high-level waste (HLW) and spent fuel. The Boom Clay formation at the Mol site is the reference where field investigations already started in 1975.

Performance assessment studies evaluate the long-term safety of the designed geological or shallow disposal systems by:

- ▣ identifying possible scenarios that might lead to the exposure of man to radioactivity or toxic substances;
- ▣ analysing the consequences of the most relevant scenarios;
- ▣ comparing the estimated concentrations, fluxes, doses and risk with appropriate safety criteria.

Because aquifers are essential components of deep and shallow waste disposal systems, hydrogeological modelling is a critical element for assessing the long-term safety of a repository.

Other key issues or components in this safety assessment are:

- ▣ the waste inventory;
- ▣ the short- and long-term behaviour of waste forms and engineered barriers;
- ▣ the barrier properties of the host formation.

We study the different processes governing the return of radionuclides to the biosphere and the factors influencing these processes. We determine the values of the input parameters for modelling these processes by performing experiments on clay cores and by field testing. The study of the coupling effects in view of validation exercises requires large-scale in situ experiments in underground research laboratories and natural analogue studies.

Objectives

- ▣ to assess the performance and to identify the most influential elements of integrated repository sys-

tems for the final disposal of radioactive waste;

- ▣ to characterise in detail the source term and to assess the compatibility and the performance of the waste forms and other artificial barriers with the clay environment;
- ▣ to determine and understand the migration of radionuclides and gases through the host formation and engineered barriers;
- ▣ to understand the whole hydrogeological system in north-eastern Belgium governing the geosphere transport to the biosphere.

In order to reach these multidisciplinary objectives, each of our projects in its field of expertise needs to fulfil the following objectives:

- ▣ to develop a methodology and associated tools orientated to long-term safety, applicable to different waste types, host formations and disposal concepts;
- ▣ to determine or verify the relevant various physical and chemical characteristics of barriers or waste forms relevant to the Belgian programme;
- ▣ to provide reliable and defensible models and parameters, based on a sound scientific understanding and to collect laboratory and field data according to quality assurance requirements;
- ▣ to test, verify and improve computer codes used in performance assessment calculations of waste disposal concepts or contaminated sites including e.g. processes like the water flow and transport of radionuclides in saturated and unsaturated engineered barriers, aquifers and soils.

Programme

Performance assessments

We develop our performance assessment programme in the framework of the following contracts and research:

- ▣ a multi-year research programme for elaborating the performance assessment of the potential geological disposal of high-level and long-lived radioactive waste in the Boom Clay layer at the Mol site;
- ▣ hydrogeological studies focussed on the further development of the multi-layer regional aquifer model of north-eastern Belgium. We complemented these studies by an internal R&D project in which the spatial variability of the hydraulic conductivity of the Boom Clay is investigated;

- ✎ the BENIPA (BENtonite barriers in Integrated Performance Assessment) project, evaluating the role and harmonising the treatment of bentonite barriers in performance assessments of high-level waste disposal systems;
- ✎ the SPIN (Testing of safety and Performance INDicators) aimed at exploring the applicability of various output variables (concentrations, fluxes, etc) as performance and safety indicators;
- ✎ the BORIS (Building confidence in deep disposal: the borehole injection sites at Krasnoyarsk-26 and Tomsk-7) project, for assessing the adequacy of data and samples from the Russian injection sites to understand the chemical behaviour and migration of radionuclides in the geological environment;
- ✎ performance assessments for the disposal of low-level and medium-level waste in hard rock in the Murmansk area (NW Russia)
- ✎ Complementary performance assessments for site-specific concepts for surface or deep disposal of low-level waste at the nuclear site Mol-Dessel (Belgium);
- ✎ Contribution to the impact assessment of a radium storage facility at Olen (Belgium).

Waste forms and packages

The programme consists of the following activities:

- ✎ the investigation of the chemical stability of cemented waste representative for the conditioned reprocessed BR2 fuel in Boom clay disposal media;
- ✎ the investigation of the effect of the radiolytic degradation of bituminised waste and of the degradation products of contaminated cellulose waste on the solubility of Am and Pu in geological disposal situations;
- ✎ the study of the corrosion mechanisms of high-level waste glass in geological disposal media, with special emphasis for the leaching behaviour of Np and Tc, and for the modelling aspects. In complement to this, the characterisation of Np complexes formed upon interaction with clay water are studied;
- ✎ the determination of the solubility of UO₂ in Boom clay water, emphasising the effect of humic acids and carbonates, and the study of the effect of α -radiolysis on the corrosion of UO₂;
- ✎ the study by electrochemical techniques, of the sensitivity to localised corrosion of stainless steel container materials in geological disposal media. The influence of the most important parameters on the pitting corrosion is considered. To elucidate the time dependence of the corrosion, immersion corrosion tests are carried out;
- ✎ the demonstration by in situ tests of the interaction behaviour between cemented or vitrified waste and Boom clay or backfill materials.

Near- and far-field studies

These activities are focused on the characterisation of the backfill material (near-field) and the host formation (far-field). More specifically, we can mention:

- ✎ migration tests carried out to study the diffusion of actinides, fission products and the mobility of the dissolved organic matter in the interstitial clay water. We performed experiments with labelled organic matter (OM) to understand the role of OM on the migration of radionuclides. Electrokinetic methods are developed and used, as a technique to reduce the very long time presently needed for the migration experiments, and to study the speciation of the different radionuclides in the reducing clay sediment;
- ✎ large-scale 3-D experiments with tritiated water, with ¹⁴C labelled bicarbonate, and with ¹⁴C labelled OM, installed from the underground research laboratory (URL). To increase the confidence in the modelling results for very long time spans, we study the behaviour of naturally occurring U and daughter products;
- ✎ investigations intended to understand the role of the organic matter on the transport of radionuclides in a reducing, organic rich clay sediment (TRANCOM II project) and to develop a conceptual model to be implemented in performance assessment calculations;
- ✎ a large-scale in situ demonstration and feasibility project (RESEAL II), for developing and assessing backfill and sealing materials and methods. The project consists of a preliminary small-scale sealing test in a horizontal borehole and a large-scale sealing test of a vertical shaft. The validation of models describing water and gas flow through the seal and the near field will also take advantage of this test;
- ✎ the determination of the degree of homogeneity of the Boom Clay with regard to the migration of radionuclides, through detailed sampling over the full thickness of the formation.

Achievements

Performance assessments

We elaborated a detailed performance assessment of the geological disposal of high-level and long-lived radioactive waste for the Mol site. We summarised the main results that have been obtained during the last 10 years in the frame of our contribution to the SAFIR-2 report (Safety Assessment and Feasibility Interim Report) to be presented by the Belgian Agency for Radioactive Waste and Enriched Fissile Materials ONDRAF/NIRAS to the authorities.

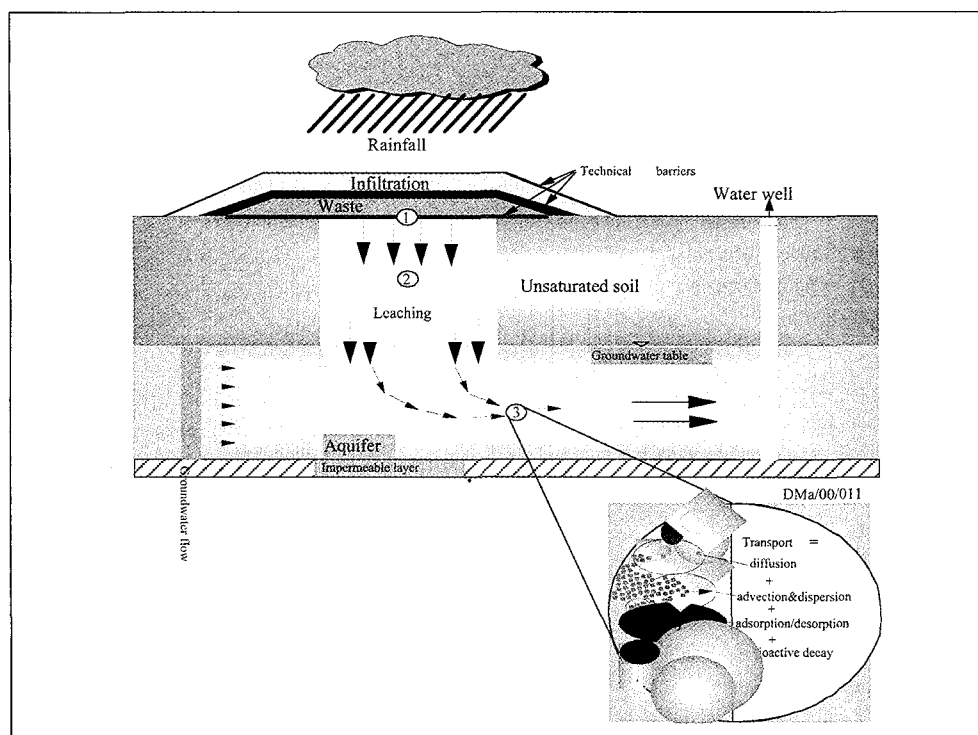
We simulated the transport of radionuclides released from the host clay layer through the Neogene aquifer into the biosphere with the MT3D and MT3D-MS codes, which are integrated in the GMS (Groundwater Modelling System) package. We also carried out a sensitivity study considering the influence of the algorithm used to solve the transport equation, the grid size, the considered boundary conditions and the location of the repository.

SCK•CEN has finalised its contribution to the EC-TACIS project on the improvement of the safety of the radioactive waste management in the North West of Russia (Kola peninsula).

The technology transfer to the Russian partners (i.e. performance assessment methodologies including

numerical models, parameter databases, etc) was the primary role of SCK•CEN before assisting them in performing safety studies for potential repositories in hard rock.

The first part of the impact assessment study for the radium storage facility at Olen (Belgium) considered one of the important pathways by which the contaminants can reach man, i.e., leaching to groundwater and use of groundwater for drinking and irrigation. Once the conceptual model to be considered in the impact assessment was derived, we could continue with the characterisation for the waste form, the engineered barriers, and the site. Waste characterisation focused on derivation of elemental concentrations under disposal conditions using a geochemical model. Properties of engineered barriers that govern the leaching of contaminants present in the waste, notably percolation of water, sorption, and diffusion, were derived on the basis of literature and construction data. Measurements of hydraulic conductivity and sorption for radium, uranium, arsenic, and lead using cores sampled from the field provided the data necessary for calculating the migration of contaminants in the aquifer sediments. We calculated leaching of radionuclides and non-radiological components towards groundwater using state-of-the-art numerical models of water flow and contaminant



Processes considered in impact assessment calculations of a shallow land disposal systems.

transport. The figure below illustrates the repository system under consideration.

In the framework of the management of L/ILW in Slovenia we provided intensive training of local staff in safety assessment methodology for surface and deep disposal concepts, which included the use of computer codes for groundwater flow and contaminant transport and evaluation of radionuclide migration in the engineered barriers. The co-operation also included consulting for more general aspects of waste management and review of safety assessment reports.

In the framework of the support programme of the Ministry of Economic affairs in the enhancement of nuclear safety in East and Central European countries, we assisted the Slovak Republic in developing their safety assessment studies for geological disposal of spent fuel and surface disposal of LLW at the Mochovce site.

The results of the measurements of hydraulic conductivity of the clay cores taken from boreholes at Zoersel, Mol and Weelde (Belgium) are now available. They confirm the horizontal homogeneity of the Boom Clay formation in the considered area with a slight influence of the depth on the value of the hydraulic conductivity.

A new version of the regional hydrogeological model for the Mol site is in preparation by collecting recent geological data, pumping rates and other hydraulic characteristics.

Waste forms and packages

We can highlight two main actions in 2000. First we organised a successful international topical workshop "Glass in its Disposal Environment" from April 11-14 in Bruges. The workshop assessed a state-of-the-art of the research on dissolution processes of vitrified waste when in contact with potential disposal environments. It also focused the importance of integrated tests, in-situ tests, and on the dialogue with the performance assessment studies for geological disposal. The workshop was attended by some 80 experts from 15 countries. The proceedings of the workshop will be published as a special issue of the Journal of Nuclear Materials in 2001.

The second action is the programme that we initiated, on behalf of the Belgian government, to develop techniques for analysing the high-level waste glass canisters produced by COGEMA in their R7T7 plant in La Hague (France). The programme consists in

developing the non-destructive assay techniques on a real glass canister and the techniques for destructive analysis of a fully active glass sample taken during the vitrification process.

This programme should provide amongst others the inventory and distribution of some specific radionuclides (^{137}Cs , ^{244}Cm) and the thermal output of the vitrified waste canister. Regarding radionuclide inventory, we will investigate the commonly measured radionuclides (as by γ - and α - total analysis), and the radionuclides of importance for the long-term dose-to-man in the disposal concept, like ^{79}Se , ^{126}Sn , ^{93}Zr and ^{107}Pd . Lot of expertise already exists, but we need to pursue a number of analytical procedures, equipments and demonstrations for the final data to be available. The chemical composition of the sample will also be measured.

We also emphasise some on-going actions of general importance for our programme.

We continued our contribution to the Fusion Programme (*see chapter Fusion*), through our measurements of the beryllium reactivity in various conditions, and through a new action on Be-waste. We also continued our participation in the European Network for Quality Checking Facilities for Radioactive Waste Packages (ENTRAP). Specific progress in the working groups on destructive and non-destructive analysis are reported elsewhere (*see chapter Chemical Analyses*).

We further succeeded to obtain an extension of the accreditation by BELTEST for a number of projects within the section. In this frame, we improved our laboratory equipment by adding a mass spectrometer to our thermo-gravimetry differential/ thermal analysis apparatus and by operating a new glove box for Pu-active tests in controlled CO_2 conditions.

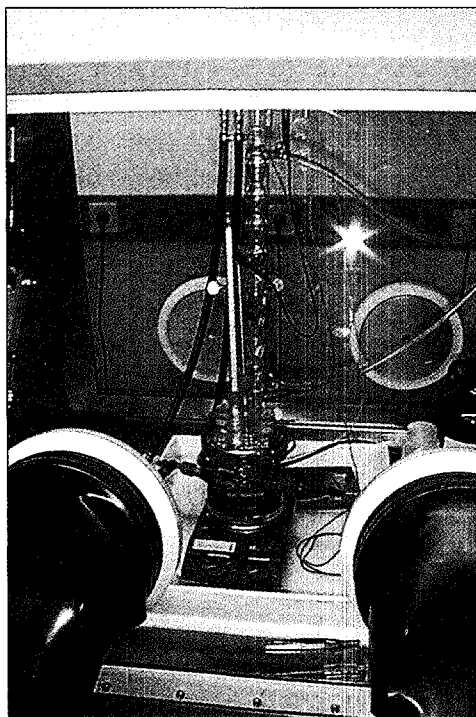
Next, we report on the R&D projects that are dealing with assessing the long-term behaviour of either the container material or the conditioned radioactive waste.

We achieved significant progress in our *in-situ* project CORALUS (CORrosion of Active gLass in Underground Storage conditions). We will install in the Boom Clay for up to ten years, four modular test tubes to be run at different temperatures, containing inactive and alpha-active SON68 glass samples, several backfill materials and, for two of them, ^{60}Co sources. After the blank test (inactive tube) performed last year for three months at 90°C in order to test and optimise the operation of the active tubes (sampling of interstitial solutions, measurement of *in*

situ pH and redox potential, dismantling operations), we installed in 2000 a second test tube, loaded with inactive and active glass samples. We use a glove box with the valve panel to sample the piezometer solutions and to measure the *in situ* pH and redox values. The temperature of this test tube will be raised to 30°C in the beginning of 2001, a temperature level considered as representative to study the glass corrosion behaviour after the thermal phase of the repository.

We study the susceptibility to localised corrosion (pitting) of the candidate container materials by cyclic potentiodynamic polarisation (CPP) measurements, as described in the previous report. These measurements were performed in:

- synthetic oxidised Boom clay water solutions at 140°C under aerobic conditions;
- synthetic Boom clay water and bentonite water solutions at 16 and 90°C under anaerobic conditions;
- in Boom clay and bentonite slurries at 16 and 90°C under both aerobic and anaerobic conditions.



The laboratory set-up to perform electrochemical experiments under anaerobic conditions. The double-walled corrosion cell is implanted in a glove box.

To perform the CPP experiments at 140°C, a special internal Ag/AgCl reference electrode was designed.

Increasing the temperature from 90 to 140°C caused a drastic drop of the pitting potential - the critical potential for pit nucleation - in all solutions. For the solution containing 1000 mg.l⁻¹ Cl⁻, it decreased from 782 to 511 mV for stainless steel UHB 904L and drops even below the free corrosion potential, i.e. 266 mV, for stainless steel AISI 316L hMo.

For the experiments performed in synthetic bentonite pore-water solutions, the characteristic pitting potentials are consistently more noble under anaerobic conditions. The opposite observation (more noble potentials under aerobic conditions) could be made for the experiments performed in synthetic Boom clay water solutions. This difference in potential is more pronounced for stainless steel AISI 316L hMo than for UHB 904L.

In 2000, we finalised the experimental part of the projects studying the effect of the degradation of cellulose waste and of the radiolytic degradation of bituminized waste on the solubility and sorption of Pu and Am in geological disposal conditions.

Our earlier studies have already pointed out the rather limited effect of chemical degradation products (DP) of cellulose, mainly iso-saccharinic acid (ISA) in anaerobic and alkaline conditions in undisturbed Boom clay. The sorption of Am and Pu onto undisturbed Boom clay is very high, with sorption coefficients between 8.10³ and 3.10⁴ dm³.kg⁻¹ (solid/liquid ratio of 1/5 kg.dm⁻³) and slightly increased by the presence of ISA under a concentrations of 10⁻⁴M. We also found indications that ISA itself strongly sorbs onto Boom clay, but we have no idea yet which solid phase (oxides, clay minerals, organic matter, etc) is responsible for it.

These observations are in line with the results reported by other research groups in similar conditions. In strongly alkaline ones, however, the effect of ISA on the Am and Pu solubility is much more pronounced, resulting in six orders of magnitude higher solubility in the presence of ISA at a concentration of 10⁻³M. On request of ONDRAF/NIRAS, we started therefore a new research project to investigate the effect of ISA on the Am and Pu solubility and sorption in Boom Clay affected by alkaline plume around a repository. The sorption capacity of undisturbed Boom clay for ISA will be investigated more in detail.

One of the potential problems that are related to the geological disposal of bituminised waste is that radiolytically generated water-soluble complexants might increase the solubility and reduce the sorption

of important radionuclides. Our approach to study these processes is similar as for cellulose-based waste: degradation, solubility and sorption tests. We irradiated inactive Eurobitum in contact with representative solutions until a total adsorbed dose of about 5 MGy, corresponding to the alpha-dose accumulated within 10^5 years for a fully active bitumen sample. Oxalate and (bi)carbonate were found to be the main degradation products (DP), but a whole range of other organic molecules was found as well, however at lower concentrations. Referring to its high concentration in Boom clay, we are not considering bicarbonate as an important DP. We found in DP solutions high concentrations of sodium, nitrate, and sulphate, all leached from the waste form. We noticed that Am and Pu solubilities in Boom clay pore water were slightly higher in the presence of 50 mass% of the DP solution. Modelling calculations with the Geochemist's Workbench version 3.1 showed that this increase is not due to the salinity effect of sodium, nor to complexation with nitrate or sulphate. We are now verifying whether the higher solubility can be due to complexation with oxalate. The sorption coefficients increased by a factor of 2 to 5 for Am, and by a factor 1 to 1.5 for Pu, depending on the solid/liquid ratio. We expect that this may be related to the oxidation of pyrite present in the Boom Clay through the production of iron (hydr)oxides having high sorption affinities for Am and Pu. This assumption seems to be confirmed by the decreased nitrate concentrations and the increased nitrite concentrations.

One important remaining question will be whether the DP solution that was obtained by external gamma-irradiation, at very high irradiation doses (10^4 times higher than for active Eurobitum), of inactive Eurobitum in contact with water, is representative for the DP products that will be generated in-situ by internal alpha radiation, at low dose rates, and only partially in contact with water.

Concerning the corrosion studies on high-level waste glass, we concluded the programme 1996-1999 and launched the programme for the period 2000-2004. Reference glasses are the Cogéma R7T7 SON68 and the DWK/PAMELA SM539. We would like to underline the following results:

- ☒ in pure clay water, diffusion profiles of the mobile elements (B, Na) with increasing thickness develop at the glass surface;
- ☒ in rather dense clay slurries, the corrosion rate decreases with time but remains relatively high

after two years, in spite of the high silica concentration in solution;

- ☒ the presence of a γ -radiation field has no important corrosion enhancing effect on the glass corrosion in diluted clay slurries;
- ☒ the nature of the clay environment influences the concentration of ^{237}Np and ^{99}Tc released in the clay water but not the rather small retention of the radionuclides in the glass surface
- ☒ diffusion experiments with ^{32}Si show that silica is significantly sorbed by Boom Clay;
- ☒ geochemical modelling showed that both studied glasses can in principle form secondary phases when they are in contact with (clay) water with or without clay. In certain cases, the formation of these phases is likely to determine the glass corrosion rate.

We developed a Monte-Carlo model to describe the phenomena that occur during glass corrosion on a molecular level. Estimations of the lifetime of a glass canister using analytical codes highlights the need for further refinement, mainly with regard to the uncertainties in the premises of the model and in certain model parameters.

To support the corrosion studies on Np and Tc doped-glasses, we studied the Np complexes formed during the interaction between Np-doped HLW glass and Boom Clay water, containing large amount of humic substances. Beside leaching tests intended to determine the Np concentration and speciation, we focused mainly this project the complexation of Np(IV) with humic acids as no data concerning the complexation of tetravalent actinides with humic substances under environmental conditions are available at present in the literature. The Np concentration in the leach tests ranges from $5 \times 10^{-7}\text{M}$ to $2 \times 10^{-6}\text{M}$.

The Np species present in the leachates were identified by UV-Vis spectroscopy. Carbonate and humate complexes of Np(V) are mainly observed in the leachates, anaerobic conditions being difficult to maintain in such solutions. Under the real reducing conditions expected in groundwaters for deep formations, Np(V) can reduce to tetravalent Np. The experiments on the complexation of Np(IV) with organic matter were analysed by UV-Vis spectroscopy for low pH value and by a dialysis equilibrium technique at higher pH.

Indeed, due to the high tendency of Np(IV) for hydrolysis and in order to make calculations and interpretations easier, the experiments were started at low pH ($\text{pH} < 1.5$) using the soluble fulvic acid frac-

tion of the humic substances. The complexation constant, estimated using the metal ion charge neutralisation model is found to be equal to $\log \beta = 7.86$ at pH 1 for the complex Np(IV)FA(IV) . The Np(IV) complexation with humic acids at pH 8 (Boom Clay water) is under study with the dialysis equilibrium technique. At this higher pH value, mixed complexes of the form $\text{Np(IV)(OH)}_n\text{HA}$ are expected.

Concerning the determination of the solubility of UO_2 in Boom Clay water (programme 1996-1999), we concluded that the uranium concentration in clay water without clay is determined by the formation of carbonate complexes. Complexes with humic acids are likely to be less important. XPS analyses on the UO_2 surface confirmed these results.

Near- and far-field studies

We continued the time-consuming percolation and diffusion experiments for the measurement of migration parameters in Boom Clay for the radionuclides of major concern. The figure below gives an overview of one of the temperature-controlled laboratories equipped with the oedometer and percolation cells used for the migration experiments. In view of the preparation of an interim safety assessment report for the authorities (SAFIR-II), we provided a critical review of the existing migration parameters.

We studied the influence of an alkaline plume (from concrete used as construction material) and of a sodium nitrate plume (from bituminised waste) by batch tests and migration experiments. We carried out a preliminary test in a batch reactor in presence of a Boom Clay slurry prepared with synthetic clay water, a sodium nitrate solution and a radiolytic bitumen degradation solution. Partial results tend to support

the hypothesis of a potential active role played by denitrifying bacteria present in Boom Clay in the gas generation source term around Eurobitum-type MLW.

In the frame of the international "Mt TERRI" project, we contributed in the characterisation of the hydraulic, gas and migration properties of the Opalinus Clay, a potential host formation in Switzerland. We performed gas migration experiments under isostatic confining conditions on this indurated clay.

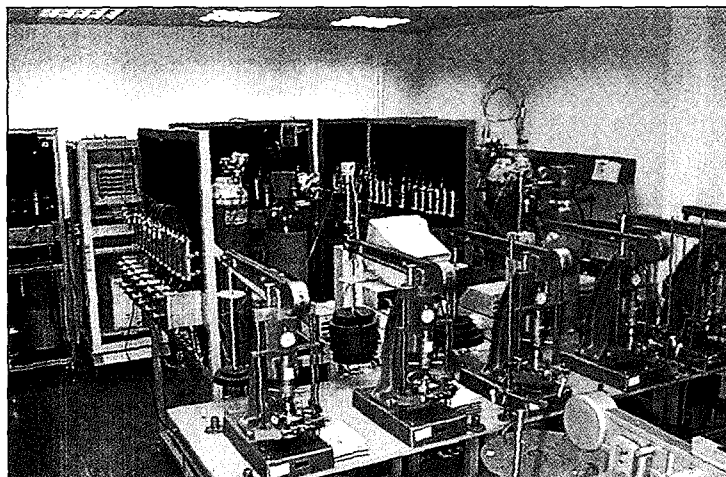
After a detailed assessment and a successful application of the electromigration technique for a direct determination of the diffusion parameters of strong retarded species, we apply it for the study on the migration behaviour of redox sensitive species like uranium. Under the Boom Clay reducing conditions, uranium is expected to be present in its 4+ valence as precipitated UO_2 giving U(OH)_4 (aq) in solution with a low solubility limit. However in bicarbonate rich media and slightly alkaline conditions (Boom Clay), traces of oxygen will very rapidly oxidise uranium to its valence 6+ as $\text{UO}_2(\text{CO}_3)_3^{4-}$, which is highly soluble. Until then, there was no direct proof on the speciation of the uranium under in-situ conditions.

Since it is difficult to control the source conditions at the start of a migration experiment, uncertainty always remains on the initial speciation of the uranium. In this case, *electro-migration* is of great help, considering that the migration behaviour of molecules in an electrical field depends on their charge (speciation).

Electro-migration experiments using prepared sources of $\text{UO}_2(\text{CO}_3)_3^{4-}$ did not show the expected migration behaviour of anionic (non-retarded)

species towards the anode side (positively charged electrode). Most of the uranium did not move and a clear presence of species moving towards the cathode (neutral or positive charged species) was noticed. It is therefore likely that uranium is present in its reduced state UO_2 (c) and U(OH)_4 (aq).

We started in parallel a PhD study to improve the mechanistic understanding of the interaction phe-



Apparatus for the migration experiments.

nomena controlling the uranium behaviour under Boom Clay conditions and to study the interaction with the organic matter (OM). Preliminary results from a first dialysis experiment suggest that the uranium(VI) complexation by OM is limited.

Large-scale hydraulic tests performed in-situ with piezometers installed perpendicular or parallel to the bedding planes of the Boom Clay Formation have confirmed at the metric scale the anisotropy observed onto small clay cores in the laboratory. A factor 2.4 was observed between the horizontal and the vertical component of the hydraulic conductivity.

Several large-scale migration experiments are still in progress in the same piezometers with a conservative tracer like tritiated water (HTO), or with weakly sorbed species ($H^{14}CO_3^-$ and ^{14}C -labelled organic matter). The concentration profiles of HTO measured for more than 13 years in the CP-1 experiment at the northern end of the HADES (High Activity Disposal Experimental Site) URL are still in good agreement with the predictive model calculations based onto migration parameters independently determined in the laboratory on small clay cores.

We completed the first phase of the study of the influence of the dissolved organic matter on the migration of radionuclides (TRANCOM-Clay project). The results showed that the organic matter might serve as well as transport agent, but also as sorption sink due to exchange with immobile organic matter. We do not observe thermodynamical equilibrium between radionuclides complexed by mobile OM and by immobile OM; it is like a quasi-irreversible partitioning is taking place. As a consequence, the concept of operational solubility ($S = 2 \cdot 10^{-13} M$) for a very small non-sorbed fraction ($R = 1$) of trivalent actinide migrating with the mobile OM is used in performance assessment.

In its second phase, the project is precisely focused on the study of the role of the organic matter on the transport of radionuclides in a reducing, organic rich clay sediment, more especially for redox sensitive nuclides (Se, U). For experiments using reactive elements, the integration of solubility concepts into the transport model is required.

In the frame of geochemical and reactive transport modelling, we continue to pursue advanced numerical computer codes. We joined the Industrial Consortium for Research and Education (ICRE) supervised by the hydrogeology program at the University of Illinois, USA. Through this consortium, we have access to several numerical codes and

information exchange with other research organisations and universities. We conducted the first scoping calculation on speciation and solubility of radionuclides in Boom Clay using the computer code The Geochemist's Workbench® provided within the consortium.

To support the numerical modelling and take advantage of data of the highest quality, we developed a software (NEA2GWB) to process and convert the database available at the Nuclear Energy Agency (NEA).

We also devoted efforts to transfer and adapt our modelling expertise to the R&D environmental remediation projects. In this frame, we performed first calculations to model the processes, e.g., ion exchange, and adsorption that are relevant for describing contaminant behaviour in soils.

The need of collecting real in-situ clay water for geochemistry led to the construction of a new concept of non-metallic piezometer. From this device installed from the URL in the Boom Clay, we will be able to measure for the first time the redox potential and the pH under the real pressure conditions of about 1.5 to 2 MPa.

We completed a series of migration experiments, running over more than six years, to study the influence of the ionic strength of the interstitial solution on the diffusion accessible porosity in clays. The analysis of results shows a substantial increase of the diffusion accessible porosity for anions by increasing ionic strength.

We also completed the study of the homogeneity of the Boom Clay on the migration properties (hydraulic conductivity, apparent diffusion coefficient, and diffusion accessible porosity) for non-retarded species. The results show quite homogeneous properties over the entire thickness of the formation.

Natural occurring REE, U, Th and their radioactive isotopes in the Boom Clay can be considered as natural analogues of critical elements for the long-term safety of a radioactive waste repository. The study of the long-term behaviour of these trace elements and radionuclides naturally occurring in the Boom Clay allows us to get information in realistic geological conditions and over geological time-periods relevant for the assessment of the safety of the disposal. This study is meant to increase the confidence in the long-term model predictions of the radionuclide migration.

The mineralogical, geochemical and radiochemical composition of the Boom Clay was studied over the entire depth of the clay deposit. Special attention was paid to the U-rich interval at the base of the Putte Member and the 'Double Band', which is the most silty layer of the Boom Clay and therefore a potential zone of higher permeability and porewater mobility. In this frame, in the near future, a new borehole will be drilled from the URL down to the base of the Putte Member.

In the frame of the RESEAL project, we started early May 2000 the artificial hydration of the shaft seal, installed in September 1999. We monitored the water intake, the relative humidity (suction), the swelling and the pore water pressure evolutions and the effect of the bentonite swelling on the host clay. The phase II of this project, started in September 2000, mainly includes the follow-up of the seal hydration, the seal-testing phase, the global interpretation of the experiment requiring also the modelling of hydro-mechanical and migration tests performed at the surface laboratory on the powder/pellets mixture used for the seal.

Perspectives

The main activities described in the previous sections, for the different fields of expertise, will continue in 2001. This is typical of research on waste disposal referring to time-consuming laboratory experiments, to long-term and large scale integrated experiments as well as to the modelling developments required for the iterative assessments of the safety of the disposal system.

Regarding more specifically performance assessments, the studies for the geological disposal of reprocessing waste in the Boom Clay layer at the Mol site will now mainly focus on the scenario development. A detailed catalogue of features, events and processes that have the potential to influence the behaviour of the repository system will be completed. A systematic, transparent and well-documented approach will be applied to identify and to define the most relevant evolution scenarios. Additional consequence analyses will be made for medium-level waste types conditioned in bitumen. The updated version of the regional hydrogeological model will be used to evaluate the necessity of performing an additional data acquisition campaign.

In the field of low-level radioactive waste we will mainly focus on the performance assessment studies for the site-specific disposal concepts referring to the

Mol-Dessel region. The issue of gas generation owing to anaerobic corrosion of steel and the concomitant gas transport in disposal galleries, Boom Clay, and aquifers for the deep disposal concept will also be investigated. We expect that our collaborations with East and Central European countries will continue and develop in the future, especially with Slovenia and Slovakia by supporting and advising them in their performance assessments of designed facilities for the disposal of low-level radioactive waste.

Through a post-doctoral research project, we will contribute further to the development and evaluation of a generic coupled geochemical transport model. This model characterises key processes of radionuclide behaviour in the soil (and other substrates) and the soil-plant environment, the groundwater, and the engineered and geological components of repositories of radioactive waste. We will test the new code on well selected cases (i.e., data sets) dealing with waste disposal issues (e.g., the effect of an alkaline plume on the behaviour of the Boom Clay or the migration of organically complexed radionuclides in a clay environment) but also environmental problems related to uranium mining and milling activities. We will also apply our experience gained in site characterisation and in flow and transport modelling in aquifer systems and variably saturated soils to the SCK•CEN's programme on environmental restoration and site remediation.

As already mentioned, 2000 was a turning-point for several large research programmes dealing with waste forms and packages like, laboratory and modelling studies on glass dissolution, cellulose degradation, radiolytic degradation of bitumen, container corrosion, and UO_2 solubility. One of the new projects to be started in 2001 is the corrosion study of doped UO_2 . Different hypotheses and processes still need to be clarified for glass, such as the effect of the glass on the clay environment. New in-situ tests will start or extend. Within the CORALUS project, we will install two new test tubes to be operated at 90°C for 2 to 4 years, using ^{60}Co sources.

The continuation and follow-up of the migration programme is straightforward and the influence of the dissolved organic matter on the migration of radionuclides will be continued in a second phase for the TRANCOM project. It will be more focused, according to the conclusions of the last sensitivity exercise of performance assessment, on redox-sensitive elements as selenium, technetium, and uranium, taking into account their impact for the dose-to-man

in the long-term. The project is focused on the role of organic matter in the reduction, precipitation, complexation and sorption processes of Se, U, Pu and Am.

The RESEAL project will be continued for 3 years. After saturation of the seal, parameters like overall hydraulic conductivity, gas breakthrough pressure, hydraulic conductivity of disturbed zone and iodine diffusion coefficient in both seal and excavation disturbed zone will be measured and used for further analysis and interpretation.

Partners

		CEBELCOR	Centre Belge d'Etude de la Corrosion (Brussels, Belgium)
		CFG	Compagnie Française pour le Développement de la Géothermie et des Energies Nouvelles (Orléans, France)
		CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Madrid, Spain)
		CIMNE	Centro Internacional de Metodos Numericos en Ingenieria (Barcelona, Spain)
		CNRS	Centre National de la Recherche Scientifique (Paris, France)
-	Chalmers University of Technology (Göteborg, Sweden)	ENRESA	Empresa Nacional de Residuos Radioactivos, S.A (Madrid, Spain)
-	Clay Technology Lund AB (Lund, Sweden)	ENSCP	Ecole National Supérieur de Chimie de Paris (Paris, France)
-	Lawrence Berkeley National Laboratory	ERM	Etudes-Recherches-Materiaux (Poitiers, France)
-	Technical University of Clausthal (Germany).	FZK	Forschungszentrum Karlsruhe (Karlsruhe, Germany)
-	Tractebel (Brussels, Belgium)	Galson	Galson Sciences Ltd. (Oakham, UK)
-	Università di Roma "La Sapienza" (Rome, Italy)	GI	Geotechnisches Institut (Bern, Switzerland)
-	University of Aberdeen (Aberdeen, United Kingdom)	GRS	Gesellschaft für Anlagen- und Reaktorsicherheit (Braunschweig, Germany)
-	University of Birmingham (Edgbaston, UK)	ICRE	University of Illinois (Illinois, USA)
AEA	AEA Technology (Harwell, UK)	IPSN	Institut de Protection et de Sûreté Nucléaire (Fontenay-aux-Roses, France)
ANDRA	Agence Nationale pour la gestion des Déchets Radio-Actifs (Châtenay-Malabry, France)	ISMES	Instituto Sperimentale Modelli E Strutture (Bergamo, Italy)
ARMINES	Association pour la Recherche et le Développement des Méthodes et Processus Industriels (Paris, France)	ISTec	Institut für Sicherheitstechnologie (Germany).
BGD/SGB	Belgische Geologische Dienst/Service Géologique de Belgique (Brussels, Belgium)	KEMA	KEMA Nuclear (Arnhem, the Netherlands)
BGS	British Geological Survey (Nottingham, UK)	KULeuven	Katholieke Universiteit Leuven (Leuven, Belgium)
CEA	Commissariat à l'Energie Atomique (Paris, France)	LU	Loughborough University (Loughborough, UK)
CEA Valrhô	Commissariat à l'énergie atomique (Cadarache and Marcoule, France)	NAGRA	National Cooperation for the Disposal of Radioactive Waste (Switzerland)
		NRG	Nuclear Research Group (Petten, the Netherlands)

PSI	Paul Scherrer Institute (Villingen, Switzerland)	M. De Craen, D. Delleuze, G. Volckaert, A. Sneyers, M. Put, "U-Th series disequilibrium studies on Boom Clay, a natural analogue study of radionuclide migration in argillaceous sediments". <i>Földtani Közölym</i> 130/2, pp. 219-228, Budapest, 2000.
QuantiSci	QuantiSci Limited (Henley-on-Thames, UK)	
TVO	Teollisuuden Voima Oy, Development Office (Helsinki, Finland)	B. Grambow, A. Loida, A. Martinez-Esparza, P. Diaz-Arocas, J. de Pablo, J.L. Paul, G. Marx, J.P. Glatz, K. Lemmens, K. Ollila, H. Christensen, "Source term for performance assessment of spent fuel as a waste form", EUR 19140, 2000.
UCL	Université Catholique de Louvain (Louvain-la-Neuve, Belgium)	
ULB	Université Libre de Bruxelles (Brussels, Belgium)	M. Gysemans, H. Moors, "Determination of ⁷⁵ Se, ⁹⁵ Zr, ²³⁷ Np and ²⁴¹ Am activities in Boom Clay samples from laboratory migration experiments using gamma-ray spectrometry". <i>Applied Radiation and Isotopes</i> 53, pp. 209-213, 2000.
ULg	Université de Liège (Liège, Belgium)	
UPC	Universitat Politecnica de Catalunya (Barcelona, Spain)	L. Hardy, I. Wemaere, I. Van Keer, J. Marivoet, X. Sillen, "Transport modelling of environmental isotopes within performance assessment of a radioactive waste repository in clay". <i>Proc. Int. Conf. on Tracers and Modelling in Hydrogeology</i> , Liège, IAHS Publ. No. 262, pp. 263-268, 2000.
VITO	Vlaamse Instelling voor Technologisch Onderzoek (Mol, Belgium)	
VTT Energy	Valtion Teknillinen Tutkimuskeskus (Helsinki, Finland)	
VUB	Vrije Universiteit Brussel (Brussels, Belgium)	K. Lemmens, V. Pirlet, M. Aertsens, N. Malengreau, P. De Cannière, P. Van Iseghem, "Experimental and modelling studies to formulate a nuclear waste glass source term in representative geological disposal conditions", 403-406 EURADWASTE '99, Radioactive Waste Management Strategies and Issues. EUR 19143, Ed. C. Davies, 2000.

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Customers

ONDRAF/NIRAS	Belgian Agency for Radioactive Waste and Enriched Fissile Materials (Brussels, Belgium)	D. Mallants, A. Dierckx, L. Wang, G. Volckaert, T. Zeevaert, A state-of-the-art methodology for impact assessment of covered uranium mill tailings. <i>International Conference of "Radiation Legacy of the 20th Century: Environmental Restoration (RADLEG-2000)</i> , Moscow, 11 pp, 2000.
UM	Union Minière (Olen, Belgium)	B. Neerdael, M. De Craen, "Repositories for high level waste in argillaceous formations – Geological key questions". <i>Acta Geologica Hungarica</i> , 43/2, pp.115-134, 2000.

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