

**PROGRESS AND TRENDS IN THE BELGIAN PROGRAMME ON HIGH-LEVEL
WASTE DISPOSAL**

B. Neerdael, P. Van Iseghem, J. Marivoet, P. De Cannière
SCK•CEN, Boeretang 200, Mol, Belgium

ABSTRACT

Waste and clay characterization in the context of radioactive waste disposal extends over 25 years. For high-level and long-lived waste, no siting procedure has yet been launched in Belgium. The studies on geological disposal are referring to a potential host rock, the Tertiary Boom Clay Formation. At SCK•CEN, special attention has been devoted to the radioactive waste characterization, the migration studies in clay, and the geomechanical behavior of this potential host rock. Performance studies help in assessing the level of confidence that must be achieved at the various stages of the characterization in both near and far fields.

The waste characterization issue rapidly extended towards the study of compatibility of these waste forms in the near field and geological environment, with a view to define acceptance criteria. Present activities include a research and development programme on spent fuel that is becoming, in parallel with ongoing studies on glass, an important issue in the frame of alternative fuel cycle policies in Belgium.

Our capabilities in research on migration are being extended on both experimental and modelling fields by developing electromigration techniques and geochemical coupled transport codes respectively. Other components of the near field, in particular the backfill and sealing materials, remain the subject of extensive in-situ and laboratory testing.

Regarding geomechanics, the feasibility of digging large disposal galleries in a deep, plastic clay has been demonstrated at the Mol site. Attention is now devoted to the quantification of the disturbance affecting the host rock during the construction and operation of a repository. These developments are currently covered by the activities of the Economic Interest Grouping (EIG) PRACLAY, aimed at managing the extension of the HADES Underground Research Facility (URF) and the corresponding experimental programme.

More efforts in the frame of performance assessments were devoted to a systematic scenario selection and analysis. In complement to the deterministic consequence analysis of the identified scenarios, stochastic calculations are carried out for uncertainty and sensitivity analyses. To build confidence in the assessment, a natural analogue study on the potential host clay formation and a palaeohydrogeological study of the aquifer system are carried out.

INTRODUCTION

When considering disposal of long-lived waste in deep argillaceous formations, the host rock is regarded as the main barrier. In Belgium, the Boom Clay Formation has been studied for more than 20 years as a potential host rock for high-level and long-lived radioactive waste.

WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

To assess the safety of a potential repository in clay, SCK•CEN is investigating the different processes able to transport the radionuclides to the biosphere and the factors influencing these processes, starting from the behavior of the waste forms in their geological environment.

Such waste and site characterization work involves the application of a wide range of disciplines as well as novel and innovative science. Multi-disciplinary activities require approaches to be developed for evaluating information or data requirements and priorities in connection with the performance assessment of the whole disposal system. The same philosophy is applied to define the level of understanding and confidence that can or must be achieved at various stages of this characterization work.

The quantification and validation of the interactions between specific materials and components of the disposal system require both laboratory and in-situ experiments at different scales. This is particularly true for the near-field, including the interaction between artificial and natural barriers systems. In-situ tests are performed in the HADES (URF), which, as an underground laboratory in a plastic clay layer, remains unique.

Two large-scale integrated tests are currently carried out in the HADES URF:

- interaction between a simulated waste glass and candidate backfill materials, in the presence of a α and γ radiation field, and a heat field representative for vitrified waste (CORALUS test); and
- sealing experiment installed in a small exploratory shaft (RESEAL test). This low permeability seal will be tested for its water / gas tightness and its mechanical stability.

WASTE FORMS AND PACKAGES

Some highlights of on-going studies are reported in this section for three types of materials: vitrified waste (COGEMA R7T7 & SON68, PAMELA), spent fuel, and container materials.

Recent laboratory experiments on dissolution of high-level waste (HLW) glass lead to the following observations:

- The release of two critical radionuclides, neptunium (^{237}Np) and technetium (^{99}Tc), in different environments appears to be much lower than expected compared to literature data. The outcome of this testing programme, performed in a parametric way, could of course affect the choice of the backfill material.
- The determination of the complexation constant for Np (IV) with humic acids is under study (1). Np complexes formed upon leaching from a HLW glass in clay water, with humic acid and carbonate concentration as parameters, are characterized by Laser PhotoAcoustic Spectroscopy (LPAS) and UltraViolet-Visible Absorption Spectroscopy (UV-Vis) techniques.
- Detailed interpretation of the samples tested in the former CERBERUS experiment (direct contact with Boom Clay at 85°C) indicates a slightly lower corrosion rate in presence of a γ -radiation field (2).

As support to laboratory testing, modeling efforts are conducted to investigate the basic glass dissolution process and to assess the influence of secondary phases on the dissolution kinetics. Based on assumptions taken, large uncertainties affect the determination of the lifetime of a glass canister in real disposal conditions, ranging from 17,000 years to several million years.

Studies on spent fuel were first focused on the solubility of uranium dioxide (UO₂) in Boom clay water. The effect of dissolved organic matter (humic acids) and of carbonate on the soluble uranium concentration is investigated. Tests were done under anaerobic conditions and the control of the inert and/on CO₂ atmosphere in the glove boxes requires much attention (3). Another study deals with the effect of α -radiolysis on the corrosion of UO₂ in clay water. In a first approach, UO₂ powder containing ²³⁵U in contact with water was irradiated with thermal neutrons (BR1 reactor, SCK•CEN). In the second approach, we produced the α -dose by doping UO₂ with ²³³U or ²³²U. Interpretation of these tests is in progress.

The susceptibility to pitting corrosion of candidate container materials in Boom Clay water was investigated. The main focus was given to stainless steel AISI 316L hMo, presently the reference material for the waste agency, and the higher alloyed stainless steel UHB 904L, with minor emphasis on some other materials like carbon steel, nickel (Ni)- and titanium (Ti) alloys. As pyrite oxidation products, likely to play a detrimental role for metallic barriers, had been identified in the frame of the CERBERUS test, simulating a COGEMA canister (4), it was decided to investigate the effect of chloride, sulphate, and thiosulphate through electrochemical corrosion tests. Stainless steels, especially the low-alloyed ones, appear to be sensitive to pitting corrosion in presence of high chloride and thiosulphate concentrations in clay water. We currently need to complete these electrochemical corrosion tests at high temperature (140°C), and in anaerobic clay water, before drawing final conclusion (5). The time dependence of the corrosion of the main stainless steels is also studied by performing immersion corrosion experiments.

GEOMECHANICS

Whatever the degree of confidence gained about site characterization before selecting a site, the real disposal conditions require to take into account the disturbance caused to the host medium first by the construction (decompression, fissuration), and later by the operation of the repository (ventilation, heat, radiation). This requires developments in geomechanics going much further than the feasibility of the repository construction itself, like the excavated disturbed zone, the time-dependent effects, the thermo-hydro-mechanical couplings, etc.

The low impact expected in plastic clays during construction and operation of the repository on the global performance of the whole disposal system calls for some demonstration. It is the primary aim of the PRACLAY experiment planned at Mol in the coming years. These investigations could lead to reconsider the disposal concept, including the nature and properties of the engineered barrier system.

Such developments are now largely covered by the activities of the EIG PRACLAY, a consortium between the waste management authority (NIRAS/ONDRAF) and the Nuclear Energy Research Centre (SCK•CEN). This economic interest grouping is aimed at managing the extension of the HADES URF (Fig.1.) and the corresponding experimental programme (6). Another paper in this conference will detail the activities of this consortium.

NEAR FIELD ISSUES

After having considered the source term and the disturbance around the disposal galleries, the role of the engineered barrier system (EBS) consisting mainly of clay-based material used for

backfilling or sealing purposes deserves some attention. This field of investigation, essential in hard rocks where the EBS plays the primary role in the isolation process of the disposal system, is less crucial in argillaceous formations. However, the redundancy principle and safety functions of any repository require high-confinement properties, including convenient swelling pressures and very low hydraulic conductivity. The latter is in the context of Mol about one order of magnitude lower than for the Boom Clay itself.

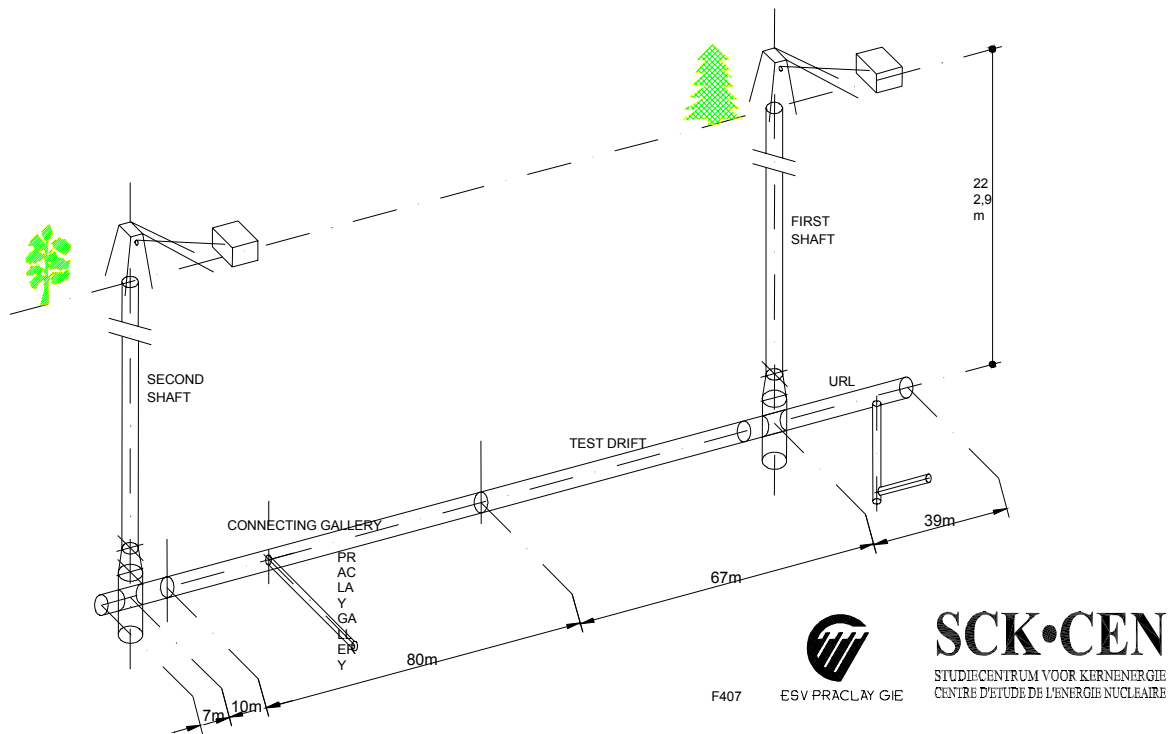


Fig. 1. Underground Research Facility

Recent developments at SCK•CEN are dealing with the definition, characterization and full-scale installation and testing under repository conditions of a mixture of high-density pellets and clay powder. More information is provided further when referring to the RESEAL test, sponsored by the Belgian, French, and Spanish radioactive waste management agencies.

Compacted bentonite blocks were used in the PRACLAY mock-up, simulating at scale 1/1 the Belgian (PRACLAY) reference concept for HLW and subjected since more than 1.5 year to saturation and heating. The current lack of data concerning these materials at high temperatures up to 150°C or even more in case of direct disposal of spent fuel required further developments for both experimental and modeling aspects. This is needed in order to prove that these high temperatures will not jeopardize the safety of the repository on a long-term basis.

A major problem in the context of direct disposal of spent fuel in geological formations is the presence and possible release of long-lived iodine (^{129}I), under the iodide form that is not retarded in clay. The confinement of ^{129}I can be obtained by applying a robust geotechnical

barrier that contains anion scavenger materials. New research is launched to screen candidate scavenger materials and to assess their sorption characteristics and long-term stability (7).

With regard to the definition of waste acceptance criteria, studies are currently conducted to assess the consequences of a modification of clay properties on radionuclide solubility and sorption. The information provided in the next section on the transport of nuclides in clay through complexation shows further that we need to pay attention to the speciation of radionuclides formed in the near-field as it will highly influence the mechanisms of interaction in the far-field.

MIGRATION IN THE FAR FIELD

In the frame of radionuclide migration, percolation experiments made on undisturbed clay cores with redox sensitive radionuclides, or trivalent actinides complexed by organic matter (OM), have pointed out the importance of introducing radionuclides in chemical equilibrium with Boom Clay. When slow chemical reactions occur during transport, it may easily lead to erroneous interpretations of migration experiments. It has been observed in case of slow reduction followed by precipitation for selenium (Se), technetium (Tc) or uranium (U), or in case of slow dissociation / formation of complexes between americium (Am) and mobile or immobile organic matter. Uncontrolled precipitation processes can be interpreted as sorption, leading to non-conservative, high retardation factors. Coupled chemical/transport codes taking into account slow kinetic reactions are very difficult to implement for the determination of migration parameters and, consequently, the preparation of appropriate radionuclide sources in direct chemical equilibrium with the clay is crucial. Very sensitive measurement techniques to directly control the speciation of radionuclides at very low concentrations in the migration experiments are required.

Further, the accurate determination of migration parameters of sorbed radionuclides generally is time consuming. The applicability of innovative migration techniques accelerated by an electrical field was investigated to determine the diffusion coefficients of ionic species in equilibrium with the clay. The electromigration technique also has the advantage that the apparent molecular diffusion coefficient can be derived from two independent methods:

- The dispersion coefficient knowing the dispersion length and convection velocity; and
- The Einstein relation between the velocity of the migrating species and the apparent molecular diffusion coefficient.

The feasibility of the technique, providing the necessary corrections are made for the electro-osmotic flow, has been demonstrated for strontium (^{85}Sr), sodium (^{22}Na), iodine (^{131}I), and tritiated water (HTO) by comparison with classical diffusion tests (8). Electromigration tests have also been successfully performed with calcium (^{45}Ca) to get access to migration parameters in the case of an alkaline plume in clay and to characterize the migration behavior of europium (Eu)- carbon (^{14}C)- OM complexes.

For radionuclides like U, plutonium (Pu), Se, and Am, identified as critical by performance assessment, special emphasis now lies on the dual role of organic matter by developing and demonstrating concepts describing radionuclide migration in a reducing, organic rich clay environment. In the TRANCOM project, the possible influence of an OM-facilitated

radionuclide transport has been assessed through detailed characterization of the organic matter and its interaction with trivalent radionuclides by means of batch and migration experiments. Boom Clay organic matter has been successfully labeled with ^{14}C . Injection experiments with the labeled organic material and with this material contacted with ^{241}Am were performed, stressing the role of colloids, acting as a fast vehicle for a small fraction of the radionuclide (9,10). Further questions remain open and will be subject to additional study. For example, performance assessors need to know whether the behavior of Se can be explained as solubility controlled or whether sorption takes place. The overall interaction of U assumed to be in the tetravalent form needs to be understood. For Pu that might exist in the trivalent form, we need to verify if the strong interaction typical of trivalent lanthanides or actinides applies.

PERFORMANCE ASSESSMENTS (PA)

The performance assessments developed at SCK•CEN consist of two consecutive steps: a systematic scenario analysis followed by consequence analyses for the identified scenarios. The scenario identification process starts with the elaboration of a catalogue of features, events and processes (FEPs) that might influence the behavior of the repository system. The FEPs are screened on the basis of their probability of occurrence and their relevance for the considered host formation, repository concept, and waste types. A distinction is made between FEPs that are about certain to occur, which are treated in the normal evolution scenario, and low probability FEPs. The latter are then classified according to the barrier that is affected. Eventually, groups of altered evolution scenarios are identified and the most relevant sub-scenarios are subjected to consequence analysis.

The consequence analysis is strongly focusing on the normal evolution scenario. The complicated repository system is conservatively simplified to a system consisting of a number of essential barriers that fulfil well-defined safety functions. The supporting Research and Development (R&D) programme has to demonstrate that the engineered and natural barriers will satisfy the assumed design and performance objectives. For each selected altered evolution scenario a deterministic "best-estimate" evaluation of its radiological consequences is made. The deterministic analyses are complemented with stochastic calculations whose results are used for sensitivity and uncertainty analyses to identify the most influential processes and parameters. The possible impact of the various sources of uncertainty, i.e., parameter values, models, and scenario descriptions, on the calculated results is also assessed (11,12).

Recently, the consequence analyses of the normal evolution scenario have been updated for the different HLW types. These are spent fuel (13), both UO_2 and mixed oxide fuels (MOX), vitrified high-level waste glass, and cladding waste. The research teams and the performance assessment team strongly interacted for the derivation of the values of the model parameters. To improve the traceability, data collection forms were filled in following a quality assurance procedure. The results of the analyses are used in the SAFIR-2 report (Safety Assessment and Feasibility Interim Report), providing the synthesis of the research on geological waste disposal. It will be presented in the course of 2000 by the Belgian radioactive waste management agency to the government.

Confidence building is an important aspect of a performance assessment. Therefore, two projects contributing to the confidence building have been launched:

- The first project studies the Boom Clay as a natural analogue for the migration of actinides (14). More efforts will be devoted to the natural evidences for the long-term barrier capacity of argillaceous sediments for radioisotopes in a European context. The distributions of the uranium/thorium (U/Th) series and of rare earth elements (REE) are measured on cores taken over the whole depth of the Boom Clay layer.
- The second project, called PHYMOL (15,16), is a palaeo-hydrogeological study of the aquifer system surrounding the potential host formation. The aim of this project is to increase the confidence in predictive groundwater flow modeling by using our understanding of groundwater flow during past glaciations. Geochemical and isotopic analyses have been carried out on groundwater samples taken on a regional scale, i.e. several tens of kilometers, to find indications on the groundwater flow patterns during colder periods. The ^{14}C and oxygen (^{18}O) distributions have been relatively successfully reconstructed by making a transport simulation over the last glacial cycle starting about 115,000 years ago.

IN-SITU TESTS

Two large-scale integrated tests are installed in the HADES URF.

- The CORALUS test is studying the interaction between an alpha-active glass and some candidate backfill materials, in the presence of a gamma-radiation field (Cobalt-60 sources) and upon heating (Fig. 2.). Based on an integrated design, the corrosion of this alpha-active glass in contact with clay, in the presence of iron and different types of candidate backfill materials will be followed for several years. A first inactive test tube was successfully installed, operated, and retrieved. The next step in preparation considers the installation of new tubes, which will be loaded with glass samples doped with ~0.85 wt% of NpO_2 , Am_2O_3 or PuO_2 (17)

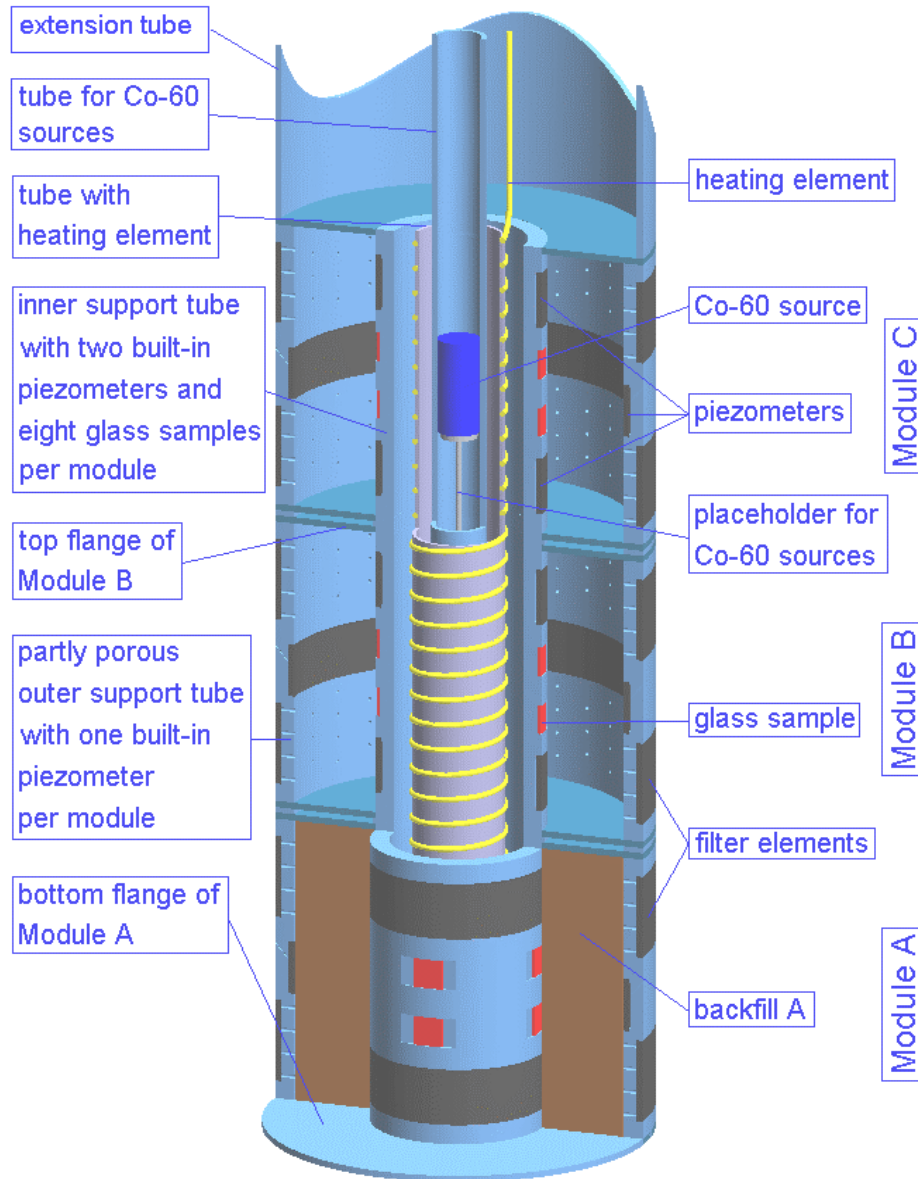


Fig. 2. The CORALUS experiment

- The RESEAL test is intended to study the performance of a seal plug installed in semi-industrial conditions and representative scale from the exploratory shaft of the HADES URF (Fig.3a. and 3b.). This low permeability seal plug (clay pellets) will be tested for its water/gas tightness and its mechanical stability in case of overpressure conditions. The test should, of course, demonstrate that preferential migration of water, gas, or radionuclides through the shaft but also through the interface seal-host rock or even through the excavation disturbed zone around the shaft can be avoided.

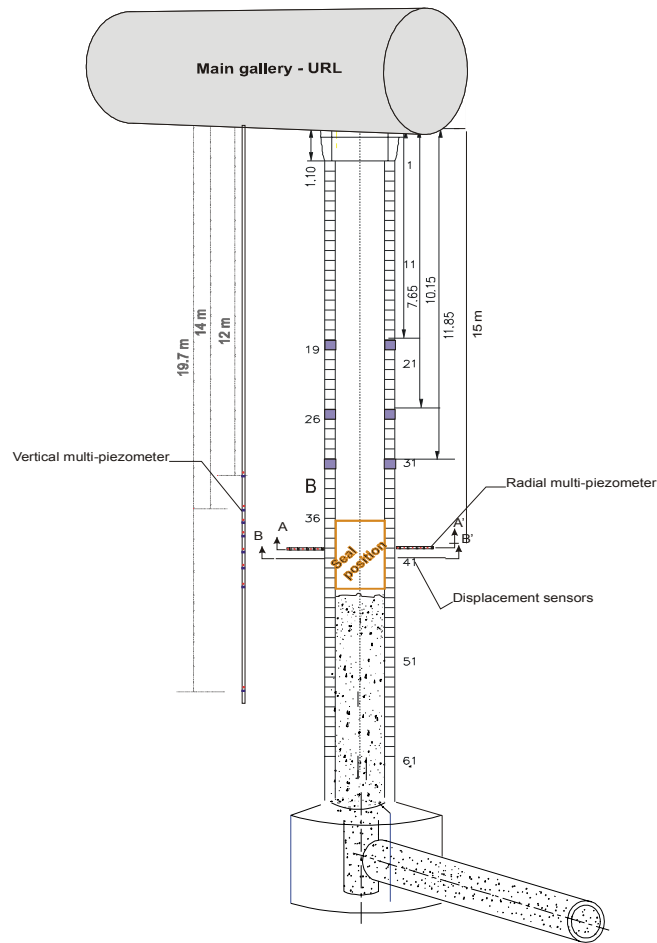


Fig. 3a. Position of the seal in the shaft

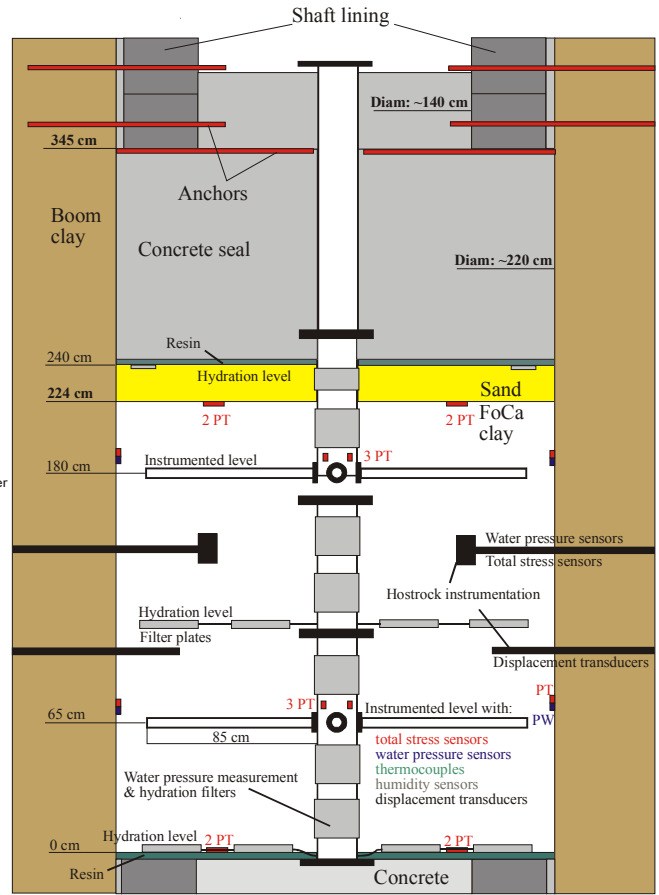


Fig. 3b. General overview of the instrumented seal

NEW RESEARCH TOPICS AND TRENDS

The new 5-year framework programme on nuclear safety of the European Commission and the enlarging information exchange and co-operative actions within different international networks, including Eastern European countries and New Independent States, highlight new topics and trends for the near future.

Much knowledge has been gained, e.g. on glass and container corrosion, or is being gained on spent fuel, but other waste types (spent fuel from material testing reactor, dismantling waste, waste from site restoration and clean-up) will require new characterization approaches, processing and conditioning techniques or management scenarios. Waste characterization should be continued and enlarged with respect to process control and verification, including standardization of methods and procedures.

The long-term performance of most of the components of the disposal system still requires to be confirmed and, to some extent, demonstrated.

WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

Coupled geochemical transport models are required for data interpretation and performance assessment studies, particularly in case of multicomponent transport.

The impact of retrievability on disposal concepts needs to be determined in order to assess the technical, managerial, and economical consequences. The development of monitoring systems may in this frame become an important issue for investigation in URF's.

We have further to fill the gap between the different stakeholders of the public and the decisionmakers to better support public acceptability. SCK•CEN has therefore recently launched different activities dealing with human and social sciences like communication, risk perception, ethics, etc.

CONCLUSIONS

The performance assessment of the Belgian reference repository for HLW disposal is a strongly iterative process. Initially, the main objective of the assessment was to understand the behavior of the integrated repository system and the role of the various barriers. Performance assessment is now more and more contributing to identify research priorities in both waste and site characterization programmes. As more results from the characterization programmes became available, the level of detail and the confidence of the performance assessment is significantly increased.

Over the last ten years, many developments were carried out for basic research as pointed out in the previous section. However, new issues are evolving and will be addressed in the near future. To the already listed topics, we could add e.g. the definition of new safety and performances indicators like concentrations and fluxes in the geosphere, abstracting the influence of the biosphere, and consequently building confidence in the safety assessment.

Many methodological developments have been realized on sampling, sample conservation, and characterization of materials, both in the framework of the migration programme and also for the recent study of Boom Clay as a natural analogue as hereabove mentioned. The evaluation of the SAFIR II report will lead to the definition of research efforts needed to cover the missing information with a view to propose a safe and acceptable solution. To increase public acceptance, more full scale demonstration will be needed.

REFERENCES

1. V. PIRLET, P. VAN ISEGHEM, A. DIERCKX, and C. MARQUARDT, "The investigation of the Np complexes formed upon interaction of high level waste glass and Boom Clay media", Scientific Basis for Nuclear Waste Management XXII, Boston, 30 Nov. – 4 Dec. 1998, (1998).
2. P. VAN ISEGHEM, E. VALCKE, B. KURSTEN, and A. LODDING, "In situ testing of the chemical durability of vitrified high-level waste in a Boom Clay Formation in Belgium: the effect of gamma radiation", 7th Int. Conference on RAdioactive Waste Management and Environmental Remediation (ICEM 99), Nagoya (Japan), 26-30 Sept. 1999, (1999).

WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

3. K. LEMMENS, C. CACHOIR, and P. VAN ISEGHEM, "Static dissolution of UO₂ in interstitial Boom Clay water", Scientific Basis for Nuclear Waste Management XXII, Boston, 30 Nov. – 4 Dec. 1998, (1998).
4. L. NOYNAERT, P. DE CANNIERE, B. KURSTEN, D. DE BRUYN, P. VAN ISEGHEM, and G. VOLCKAERT, "Study of the effects of heat and radiation on the near field of a HLW or spent fuel repository in clay – The CERBERUS project", EURADWASTE'99, Luxembourg, 15-18 Sept. 1999, (To be published).
5. B. KURSTEN, and F. DRUYTS, "Influence of thiosulphate on the pitting behavior of candidate container materials for the disposal of HLW in Boom Clay", Submitted to NACE 2000.
6. B. NEERDAEL, and J.P. BOYAZIS, "The Belgium underground research facility: status on the demonstration issues for radioactive waste disposal in clay", Nuclear Engineering and Design 176, pp 89-96, (1997).
7. N. MAES, A. DIERCKX, X. SILLEN, L. WANG, P. DE CANNIERE, M. PUT, and J. MARIVOET, "Sorption of ¹²⁹I on activated carbon", 7th International Conference MIGRATION'99, Lake Tahoe, USA, 26 Sept. – 1 Oct. 1999, (To be published).
8. N. MAES, H. MOORS, A. DIERCKX, P. DE CANNIERE, and M. PUT, "The assessment of electromigration as a new technique to study diffusion of radionuclides in clayey soils", Journal of Contaminant Hydrology 36, pp. 231-247, (1999).
9. A. DIERCKX, P. DE CANNIERE, A. HALL, A. MAES, N. MAES, M. PUT, J. VAN DER LEE, L. WANG, and P. WARWICK, "The behavior of ²⁴¹Am – ¹⁴C labelled organic matter complexes in intact Boom Clay cores", 7th International Conference MIGRATION'99, Lake Tahoe, USA, 26 Sept. – 1 Oct. 1999, (To be published).
10. A. DIERCKX, M. PUT, L. WANG, H. MOORS, N. MAES, and P. DE CANNIERE, "The influence of organic matter on radionuclide transport – The TRANCOM-Clay Project", EURADWASTE'99, Luxembourg, 15-18 Sept. 1999, (To be published).
11. J. MARIVOET, I. WEMAERE, P. ESCALIER DES ORRES, P. BAUDOIN, C. CERTES A. LEVASSOR, J. PRIJ, K.H. MARTENS, and K. ROHLIG, "The EVEREST Project: Sensitivity analysis of geological disposal systems", Reliability Engineering and System Safety, 57, pp. 79-90, (1997).
12. J. MARIVOET, G. VOLCKAERT, I. WEMAERE, and D. MALLANTS, "Approaches for treating uncertainty in the long-term performance assessment of a geological repository in clay", Proc. IAEA Conf. on Topical Issues in Nuclear, Radiation and Radioactive Waste Safety", Vienna, 30 Aug. - 4 Sept. 1998. IAEA, Vienna, TECDOC-1031, pp. 1-9, (1998).
13. X. SILLEN, J. MARIVOET, and G. VOLCKAERT (1999), "Spent Fuel Performance Assessment: the Mol site (SPA project)". SCK•CEN, Mol (report in preparation)
14. M. DE CRAEN, D. DELLEUZE, G. VOLCKAERT, A. SNEYERS, and M. PUT, "Behavior of natural isotopes of the U and Th decay series in the boom clay - A natural analogue study", Proc. ENS TOPSEAL '99 Conf., Antwerp, 10-14 Oct. 1999 (Vol. 2), pp. 214-218, (1999).
15. I. WEMAERE, J. MARIVOET, H. PITSCH, C. BEAUCAIRE, C. MARLIN, J.L. MICHELOT, A.C. PHILIPPOT, M. HASSANIZADEH, and F. VAN WEERT, The PHYMOL Project. Proc. NEA Workshop on "Use of hydrogeochemical information testing groundwater flow", Borgholm, Sweden, 1-3 September 1997. NEA-OECD, Paris, pp. 347-353, (1999).

WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

16. L. HARDY, I. WEMAERE, I. VAN KEER, J. MARIVOET, and X. SILLEN, “Transport modeling of environmental isotopes within performance assessment of a radioactive waste repository in clay”, Proc. Int. Conf. on Tracers and Modeling in Hydrogeology, Liège, 23-26 May 2000. IAHS (proc. in preparation)
17. E. VALCKE, P. VAN ISEGHEM, N. GODON, and N. JOCKWER, “The development of an integrated in situ corrosion test on radioactive HLW glass (CORALUS)”, EUR 18323, pp 79-92, (1998).
18. B. DEREPPER, and G. VOLCKAERT, “Large scale in situ demonstration test for repository sealing in an argillaceous host rock – The RESEAL Project”, EURADWASTE'99, Luxembourg, 15-18 Sept. 1999, (To be published).