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# EURIDICE News

NR. 3





## IN MEMORIAM



We are sad to announce you that Paul Govaerts, member of the board of EIG EURIDICE and Managing Director of SCK•CEN, passed away in the beginning of this year. Paul Govaerts was one of the driving forces of the EIG. He co-signed the articles of associations in 1995 thus founding the EIG EURIDICE and delivered , as a member of the board, an important contribution to the further development of EIG EURIDICE. In the more than ten years that he participated in the management of the EIG the underground laboratory HADES was extended, different Research projects were launched and the mission of the EIG EURIDICE was extended. This evolution could certainly not have been possible without the positive attitude, the engagement and the collegiality of Paul Govaerts . We will remember him as a very human and intelligent person with whom it was nice working together.

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Dear Reader,

We are delighted to present you with the third issue of EURIDICE news. The only topic of this issue is the reporting of the results of the OPHELIE mock-up experiment.

The OPHELIE mock-up experiment was started in 1995 as a preparation for the PRACLAY in situ Experiment. The latter consisted at that time in operating a 30 metre long pilot gallery simulating the behaviour of a disposal gallery for vitrified HLW.

Before installing the PRACLAY Experiment, it was decided to first operate a large-scale surface test, called OPHELIE. The OPHELIE mock-up experiment consisted in the onsurface realization and operation of a 5 metre long mock-up of the PRACLAY Experiment .

Operation of the mock-up started in 1997 and the mock-up was dismantled in 2002.

The operation and the analysis of the OPHELIE mock-up experiment among others brought ONDRAF/NIRAS to change the HLW disposal design as well as the design and the objectives of the PRACLAY Experiment to be installed. Consequently, the PRACLAY Experiment was replaced by various other experiments, each contributing in its own way to the demonstration of the geological radioactive waste disposal feasibility.

The major challenge for the coming years is the realisation of the PRACLAY experiments. These experiments consist in onsurface and underground in situ experiments. The PRACLAY experiments should provide an important input for the Safety and Feasibility Cases (SFC) of ONDRAF/NIRAS. The first SFC aims at substantiating the feasibility, for a defined zone in Boom Clay, of the proposed disposal system as well as its capacity to ensure operational and passive long-term safety. As the publication of the first SFC by ONDRAF/NIRAS is planned around 2013, the first results of the PRACLAY experiments, which will be available around 2011, will be considered in the first SFC. The next edition of EURIDICE news will focus on the objectives and planning of the PRACLAY experiments.

We hope you will enjoy reading EURIDICE news

Jean-Paul Minon  
Chairman of the board

Marc Demarche  
Manager

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# THE OPHELIE MOCK-UP

## INTRODUCTION

Different repository designs have been studied in Belgium for more than a quarter of a century in the framework of the R&D programme for the deep disposal of radioactive waste in Boom Clay.

In the beginning of the 90s, the Belgian Agency for Radioactive Waste and Enriched Fissile Materials (ONDRAF/NIRAS) initiated the PRACLAY Project (PREliminary demonstration test for CLAY disposal) to demonstrate by direct experiment the technical and economical feasibility of the reference design for the disposal of vitrified high-level waste (HLW). This project involved the construction in situ of a fully instrumented 30 m long pilot gallery to simulate the behaviour of a disposal gallery, as defined in the reference design. Except the heat-emitting vitrified waste packages, replaced by electrical heaters, the materials and techniques used had to be as similar as possible to the real ones.

In 1995, in preparation for the construction of the pilot gallery, ONDRAF/NIRAS and EIG EURIDICE (named EIG PRACLAY at the time) decided to first operate a large-scale surface test, the OPHELIE mock-up (On-surface Preliminary Heating simulation Experimenting Later Instruments and Equipment – Figure 1).

The results obtained in the framework of the OPHELIE mock-up significantly influenced the decision to revise the reference design and to redefine the in situ PRACLAY Experiment.

The present Euridice-News gives an overview of the whole mock-up experiment, from the preliminary design studies to the lessons learnt.



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Figure 1: the OPHELIE mock-up during operation

## Reference design for vitrified HLW disposal in the 90s

The OPHÉLIE mock-up deals with the reference design valid in the middle of the 90s (Figure 2). In this design, the canisters are placed in a network of horizontal disposal galleries in Boom Clay at a depth of approximately 230 m. Each disposal gallery (2 m internal diameter and 200 or 400 m long), lined with concrete blocks, is first fitted with a metallic disposal tube of about 50 cm external diameter. The space between the disposal tube and the gallery liner is filled with a clay-based buffer material made up of pre-fabricated blocks installed on three concentric rings. Once the buffer material is saturated by water, the waste canisters are pushed into the disposal tube one by one. The heat transfer is thus optimized (good contact between the disposal tube and the buffer material, no remaining voids) when the canisters are installed and, because the buffer material is resaturated and the hydraulic pressure restored, only a limited volume of steam can develop around the disposal tube. Moreover the buffer installation is conducted in a safe environment without radiation and heat.

The long-term radiological safety of the disposal system is provided by multiple barriers with specific functions: the initial water tightness of the primary packaging and, mainly, of the disposal tube prevents water from the host formation to contact the glass matrix during the first 500 years (thermal phase). Between 500 and 10 000 years (isolation phase), once contacted by water, the physico-chemical stability of the glass matrix delays the release of radionuclides. After 10 000 years (geological phase), the Boom Clay host formation, the main barrier, and the material used to seal the galleries and the shafts retard and spread the transfer of radionuclides towards the aquifers.

The role of the buffer material is to protect both the natural and engineered barriers from negative impacts to maintain their integrity and their respective functions. The buffer material must have several properties:

- a thermal conductivity at least equal to that of Boom Clay ( $\lambda=1.7 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ) to prevent excessive temperatures in the engineered barriers. A good heat dissipation is indeed needed to minimise chemical processes detrimental to the corrosion of the metallic barriers and to enhance the durability of the glass matrix;
- a permeability lower or equal to that of Boom Clay ( $4 \cdot 10^{-12} \text{ m}\cdot\text{s}^{-1}$ ) to avoid hydraulic pathways around the disposal tube, and;
- a sufficient swelling capacity to fill the technological voids (space between outer rings of the buffer and the gallery lining, and joints between blocks) and an adequate swelling pressure to prevent the collapse of the disposal galleries and to minimise mechanical constraints on the disposal tube and on the host formation. A value of about 4 MPa is required.

At the end of the 90s, the reference design was improved by providing the canisters with an “individual” stainless steel overpack (AISI 316L) to better ensure their isolation from water during the thermal phase. The overpack also provides a better radiological shielding during handling of the waste packages and contributes to decrease the linear thermal power of a gallery by spacing out the waste. This evolution of the reference design is also known as the “SAFIR 2” design.

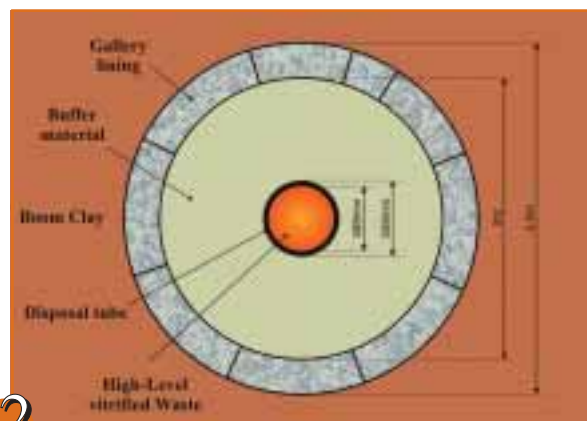


Figure 2: the reference design for heat-emitting vitrified HLW in the mid 90s (cross section of a disposal gallery)

## Objectives of the mock-up

In the framework of the R&D programme for the deep disposal of vitrified radioactive waste, ONDRAF/NIRAS and EIG EURIDICE decided in 1995 to construct and operate a large scale mock-up which simulated a section of a disposal gallery as far as the disposal tube and the buffer material are considered.

During the course of the experiment, the initial objectives of the mock-up were extended.

### Initial objectives

The main initial objective of the OPHELIE mock-up was to prepare the in situ PRACLAY Experiment. This implied the following:

- to develop a buffer material (composition, manufacturing) that met the design requirements and an artificial hydration system to simulate the hydration process encountered in a real repository;
- to test the procedures for the emplacement of this material and the instrumentation in a confined environment;
- to verify the robustness and the performances of the sensors in harsh conditions during more than four years;
- to evaluate the hydration time of the buffer material in order to determine when to start the heating phase of the in situ test which simulates the moment when the canisters are installed inside the disposal gallery.

Taking advantage of this large-scale infrastructure, it also served to monitor the thermo-hydro-mechanical (THM) behaviour of the buffer material.

A dismantling programme limited to direct observations and measurements of basic parameters of the exposed material (water content, density, thermal and hydraulic conductivity, and swelling capacity) was initially foreseen.

### Extension of the objectives

Some unexpected findings made during the operational stage of the experiment led to extend the dismantling programme. More specifically, it was completed with a more detailed characterisation of the buffer material to better understand the unexpected findings and to allow comparison between initial and exposed material. These unexpected findings mainly dealt with the following aspects:

- the lower than expected swelling pressure;
- the high apparent thermal conductivity (too high for a porous material), and;
- the presence of chemical species potentially harmful for the metallic barriers in the buffer material (enrichment in chlorides towards the dummy disposal tube) and in the hydration system.

## Design of the mock-up

The main components of the mock-up were the buffer material, the hydration system, the steel structure, the monitoring devices, and the thermal equipments.

### Specification of the buffer material and the hydration system

The development of the buffer material was the object of an extended programme carried out mainly by the Commissariat à l’Energie Atomique (CEA, France). Because of the horizontal configuration of the disposal galleries, precompacted blocks were considered as the most appropriate solution to install the buffer material. Soon, the geometry was fixed into a configuration of three concentric rings of blocks, with a technological annular space at the outside to accommodate for diameter variations of the gallery liner.

The main component of the buffer material was FoCa Clay, a bentonite coming from the Paris basin. Its major component (i.e., 80 wt % of the clay fraction) is an interstratified clay of 50 wt % Ca-beidellite and 50 wt % kaolinite. It has an important swelling potential. Sand was added to limit the swelling pressure. Addition of graphite enhanced the thermal conductivity. The final buffer block composition consisted of FoCa Clay at 60 wt %, sand (35 wt %) and graphite (5 wt %). Various production techniques were assessed at the manufacturing plant in Libos-Monsempron (France), resulting in an uniaxial compaction at 61 MPa. The average dry density of the blocks was  $2\,098\text{ kg}\cdot\text{m}^{-3}$  and the initial saturation degree about 79 %. The thermal conductivity ( $\lambda$ ) of the blocks after compaction was  $2.5\text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  and the saturated permeability about  $2 \times 10^{-14}\text{ m}\cdot\text{s}^{-1}$ .

In close relation with the study of different mixtures, CEA also designed the hydration system of the buffer. A hydration system from outside by means of a crown of 16 porous filter tubes installed at the periphery of the mock-up was chosen.

The whole system was tested in a pre-mock-up at real scale (Figure 3). The test demonstrated that the hydration of the buffer blocks caused a swelling in a few weeks. The most pronounced swelling was that of the external part of the outer ring.

Laboratory tests (suction and temperature controlled triaxial and oedometer tests) on the initial buffer material were carried out to enhance the knowledge of its THM behaviour and to calibrate/validate the coupled constitutive laws needed for the numerical simulations of the mock-up. Tests using environmental scanning electron (ESEM) photomicrographs and mercury intrusion porosimetry (MIP) techniques were also carried out to characterise the multiple porosity network of the material at different states.

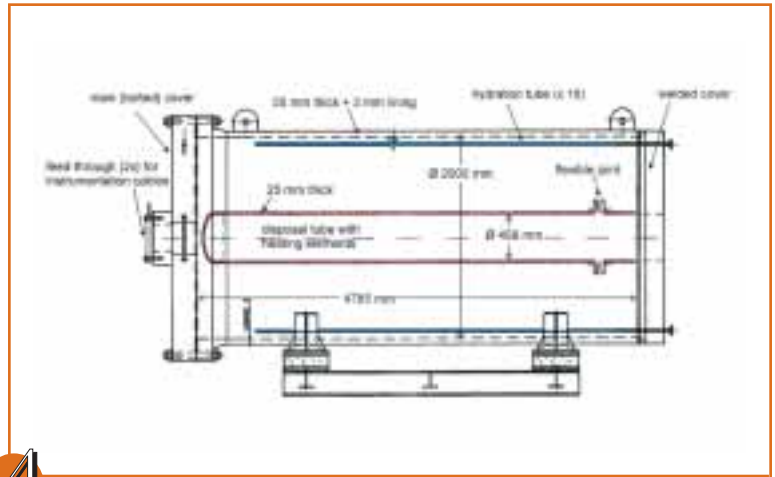


Figure 3: assembled CEA prototype mock-up before flooding (left), and the buffer material after flooding and removal of the steel structure (right)

## The steel structure of the mock-up

To limit the dimensions of the mock-up, a thick steel cylindrical jacket of 2 m inner diameter was used to simulate the confinement provided by the host formation and the concrete gallery liner.

Figure 4 shows the final design of the steel structure of the mock-up. It was dimensioned to withstand a maximum internal pressure of 5 MPa. The fixed extremity of the dummy disposal tube was welded at the backside of the mock-up to a corrugated joint allowing some displacements of the tube. The cover at the front side of the mock-up was bolted after installation of the buffer material and the other inner parts. The instrumentation cables passed through two bundles of press packing on the cover to ensure water tightness. All metallic parts in contact with the buffer were made of stainless steel: AISI 321 for the dummy disposal tube and AISI 316L for the hydration tubes. For the heaviest structures (main cylindrical jacket, backside and front covers), the carbon steel was protected against corrosion by means of a 3 mm thick AISI 304 stainless steel cladding.



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Figure 4: final design of the steel structure of the mock-up



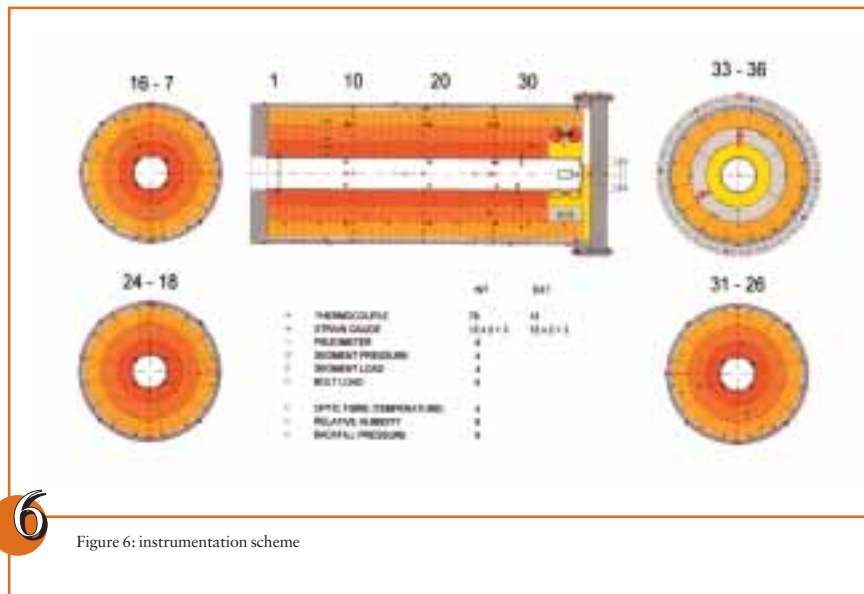
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Figure 5: the heater during assembly

## Thermal conditions

The challenge regarding thermal conditions inside the mock-up was how to simulate best the conditions of the in situ test.

A linear heat output of  $450 \text{ W}\cdot\text{m}^{-1}$  was imposed by the heater (Figure 5) placed inside the dummy disposal tube. This value is higher than the actual heat output of the waste forms (around  $350 \text{ W}\cdot\text{m}^{-1}$ ), to compensate for the limited duration of the experiment. A constant temperature was maintained at the extrados of the mock-up using heat tracing cables and an external thermal insulation. Scoping calculations performed during the preparation of the experiments indicated a maximum temperature of  $120 \text{ }^\circ\text{C}$  at the interface between the buffer and the intrados of the steel structure.



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Figure 6: instrumentation scheme

### Instrumentation

The instruments consisted mainly of temperature sensors and strain gauges, completed with pore pressure sensors, moisture sensors, and total pressure cells (Figure 6). Both the disposal tube and the outer steel liner were instrumented with 27 strain gauges each. The strain gauges on the outer liner transformed this structure into one large swelling pressure sensor, while the gauges on the disposal tube were intended to detect possible mechanical anomalies due, e.g., to a non-uniform swelling. The buffer material was instrumented with 78 thermocouples, 6 pore pressure sensors, 9 total pressure cells, and 9 relative humidity cells. These sensors were installed to follow the hydration, the swelling, and the heating of the buffer material.

## Assembly of the buffer material, hydration tubes, and instruments

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The installation of the internal parts of the mock-up (buffer blocks, hydration tubes and instruments) started in April 1997 and was completed four months later. The installation of the buffer blocks proceeded smoothly. Three concentric rings were placed around the disposal tube (Figure 7): the outer ring with 16 blocks, the middle ring with 12 blocks, and the central ring with 6 blocks. An annular space of 35 mm was kept between the outside ring and the steel jacket: this corresponds to the clearance needed for the buffer installation in a real gallery with always possible variations of the liner diameter, and it also provided space for the hydration tubes. A team of 3 persons needed less than one hour to install one buffer section (13 cm thick). Most of the time however was spent on installing the sensors on the disposal tube and in the buffer.

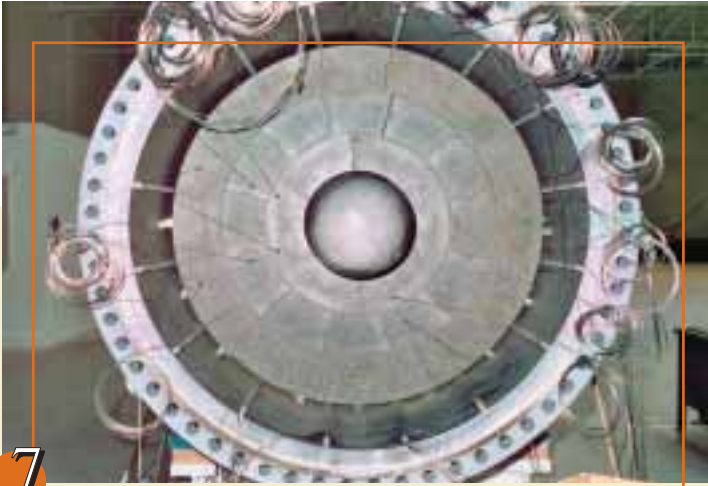
The final 0.50 m near the cover had a different structure (Figure 8): the middle ring was replaced by a ring of six concrete segments to test total pressure cells and load cells. The inside of the ring was filled with sand as inert material. To compensate the absence of swelling of concrete and sand, the outer ring was made up of clay-enriched blocks (85 wt % of FoCa Clay compared to 60 wt % in the other rings). Each of

the 149 instrumentation cables and tubes passed through two watertight covers (Figure 9).

The hydration tubes were connected to the water injection system. This latter allowed to control the water pressure up to 1 MPa and to measure the water volume injected.

No sensors were damaged during the installation. Most important was a good protection of the cable ends; as the mineral insulated cables were not finished during the installation (a connector could only be attached after having passed through the instrumentation cover with feed-through), the bare wire ends proved to be fragile, and some required a new termination. It was also important to test the water tightness of the sensors and cables assemblies systematically before the sensors became inaccessible.

After the emplacement of the bolted cover, the hermetic sealing of the sensor cables, the installation of the central heater and the external tracing cables (from September to November 1997), the mock-up was ready for the operational stage.



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Figure 7: assembly of the mock-up with the three buffer rings, sensor cables, and hydration tubes



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Figure 8: view of the concrete segment and clay-enriched blocks at the extremity of the buffer set-up near the front cover



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Figure 9: hermetic sealing of the sensor cables

## The operational stage

The operational stage of the mock-up ran from December 1997 (start of hydration) to October 2002 (dismantling), as illustrated on Figure 10. It consisted of the same three phases as those foreseen for the in situ PRACLAY Experiment: hydration at ambient temperature, heating, and cooling.

### Hydration

The hydration with a simplified formula of synthetic Boom Clay water ( $1.17 \text{ g}\cdot\text{dm}^{-3} \text{ NaHCO}_3$ ) began early December 1997. First, all technological voids in the mock-up (about  $1.6 \text{ m}^3$ ) were filled in less than one hour. Then the water pressure was gradually increased to reach 1 MPa after three weeks. Different techniques were used to monitor the progress of hydration: direct measurement of the water inflow, relative humidity sensors installed in the non-saturated buffer, follow-up of the swelling pressure, and measurement of the pore water pressure.

After about two months, a hydraulic test was performed to check for possible connections between the different hydration tubes. This test indicated that the outer part of the buffer material had filled the free annular space between the tubes.

### Heating

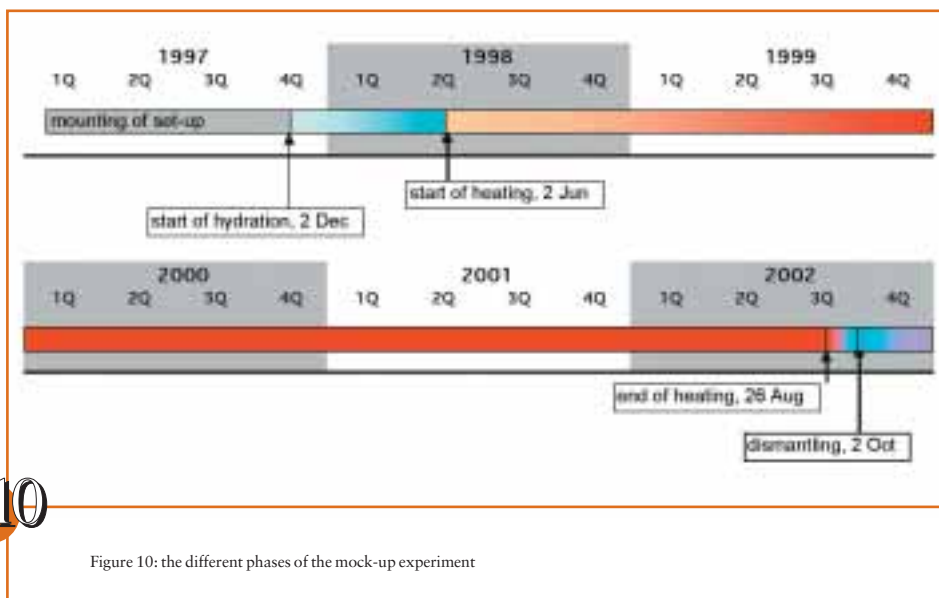
Six months after the beginning of hydration, the heating elements were switched on. After two months of heating, a first maximum of temperature was reached, with values at the disposal tube reaching  $105^\circ\text{C}$ .

In November 1998, the external heating was switched on to increase the overall temperature level. Gradually, the temperatures increased to reach, in June 2000,  $117^\circ\text{C}$  on the outside of the buffer and about  $140^\circ\text{C}$  near the disposal tube.

On two occasions, the hydration system was disconnected to test the thermo-hydraulic interaction. This resulted in pressure peaks up to respectively 2.5 and 4 MPa due to expansion of the water.

### Cooling

It was decided to proceed to a fast cooling in order to avoid major spatial changes in, e.g., chemical composition. In other words, the actual mock-up state would be “frozen” by a quick cooling. The heating systems were switched off at the end of August 2002, and the thermal insulation was removed after a few days to accelerate the cooling of the mock-up. To keep the hydration pressure at the prescribed levels, more than 100 litres of oxygen-free water had to be injected to compensate for the shrinkage of the water and other components.



## Dismantling

The dismantling of the mock-up was an essential step of the experiment. It gave an excellent opportunity to investigate the processes that could have influenced the performance of the different mock-up components. A particular attention was thus paid to its preparation, for the practical aspects (sampling, labelling, packaging, ...) as well as for the establishment of the associated scientific programme (objectives, observations and measurements during dismantling, post-dismantling analyses, ...). Adapted procedures of Quality Assurance were defined and applied.

### Dismantling operations

The dismantling activities began on 10th October 2002 and finished 10 days later. They were performed by two teams of nine persons, working in 12 hours shifts. The dismantling allowed a direct access to the engineered barriers (Figures 11), making it possible to observe the evolution of the different components after more than four years of conditions representative of those prevailing in situ. The study of the joints, the spatial variability of the buffer material, and the corrosion of metallic parts were of primary importance.

About 500 samples were taken after precise positioning by means of a theodolite. They came from both the buffer material and the various metallic parts (including the sensors). For the buffer, the most representative zone was the axial central zone, which had been subjected to the highest temperature and the lowest axial gradients.



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Figures 11: dismantling activities. The movement of sand (white zone) could be observed after removal of the cover (above). Dismantling tasks (below)

## Post-dismantling characterisation programme

The main objective of the post-dismantling characterisation programme was to better understand the phenomena occurring during the hydration and the heating phases of the experiment, as well as their causes and consequences on the performance of the different components. The post-dismantling analysis focused mainly on the examination of the THM behaviour of the buffer material, the possible changes of its mineralogical composition, the corrosion of the metallic parts, and the sensors performances.

The post-dismantling analyses on the THM characteristics of the buffer aimed at the following:

- analysing the hydration and swelling processes in the mock-up: determination of the spatial distribution of the density and water content;
- studying the THM properties: measurements of the thermal conductivity, the saturated permeability, the water retention, as well as the swelling pressure of the exposed material.

Oedometer tests and triaxial tests were equally performed on the exposed material to study whether the mock-up conditions had modified its mechanical properties.

A mineralogical characterisation of the exposed buffer materials was performed to verify possible changes which could explain the evolution of the THM properties and to support the chemical and corrosion studies.

The programme to assess the corrosion of the metallic structures included amongst others the following aspects:

- the surface analyses of corroded parts from the stainless steel liner, the hydration tubes, the disposal tube and the sensors, to determine the types and the degree of attacks;
- the characterisation of the elemental composition of the corrosion products;
- the chemical analyses of the pore water and solid phases to identify the chemical conditions prevailing in the mock-up, and;
- the microbial analyses to detect bacterial activity which could give rise to localised corrosion problems.

The dismantling and following analyses gave also the opportunity to obtain a better picture of the sensors performance. Several sensors had indeed failed during the operational stage. To find convincing answers to the various failure modes, an extensive sensor observation programme was planned, including visual inspection and corrosion analyses as explained above.

## Results

This section synthesises the results obtained during the operational stage, the dismantling and the post-dismantling activities.

### THM behaviour of the buffer material

The global analysis of the THM behaviour of the buffer material was based on the measurements obtained during the operational stage and the observations made during the dismantling, completed with the knowledge gained from the laboratory THM characterisation tests. Numerical modelling using the parameters derived from the laboratory tests results was also performed to help with the interpretation of the complex THM processes (evolution of the temperature, the pore water pressure, as well as the total stresses) experienced by the material.

### Swelling performance

The observations made during the dismantling showed that the buffer material had filled by swelling all the technological voids, in particular the technological annular space initially present at the periphery of the buffer near the hydration tubes. Such swelling is favourable to limit the microbial activity and the corrosion of the disposal tube. Though all three concentric rings swelled, generating a radial displacement of the bentonite blocks, the swelling mainly occurred at the external ring because of the presence of the large initial annular space. The swelling process was thus non homogeneous. It was also still possible to identify the blocks and the joints between them (Figures 12). With some extra precautions during the dismantling operations, most of the bentonite blocks of the inner and the medium rings could even have been recovered nearly intact.



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Figures 12: the buffer material filled all technical voids but the joints between blocks were still visible

## Swelling pressure

The swelling pressure generated in the mock-up was lower than expected, and decreased with time after reaching a maximum value. The swelling pressure values were comprised between 1.5 and 2 MPa, without considering the peaks due to artificial undrained heating events, while a value of 4 MPa was expected.

The evolution of the swelling pressure cannot be explained only by the average density of the buffer in the mock-up (for the development of the buffer material, the initial technological voids were averaged over the whole mock-up). The generation of the swelling pressure in the mock-up depended not only on the THM boundary conditions of the mock-up, but also highly on the THM behaviour of the buffer itself. One of the most important findings of the extended laboratory tests is indeed that the material presents a much lower “plasticity yielding” stress upon hydration, heating and loading. Another aspect is that the material presents a small potential of “collapse” (reorganisation of the micro-structure which may induce a decrease of the stress) upon hydration when approaching saturation. Numerical simulations using the parameters derived from the extended laboratory THM characterisation programme reproduced fairly well the observed evolution of the swelling pressure (Figure 13).

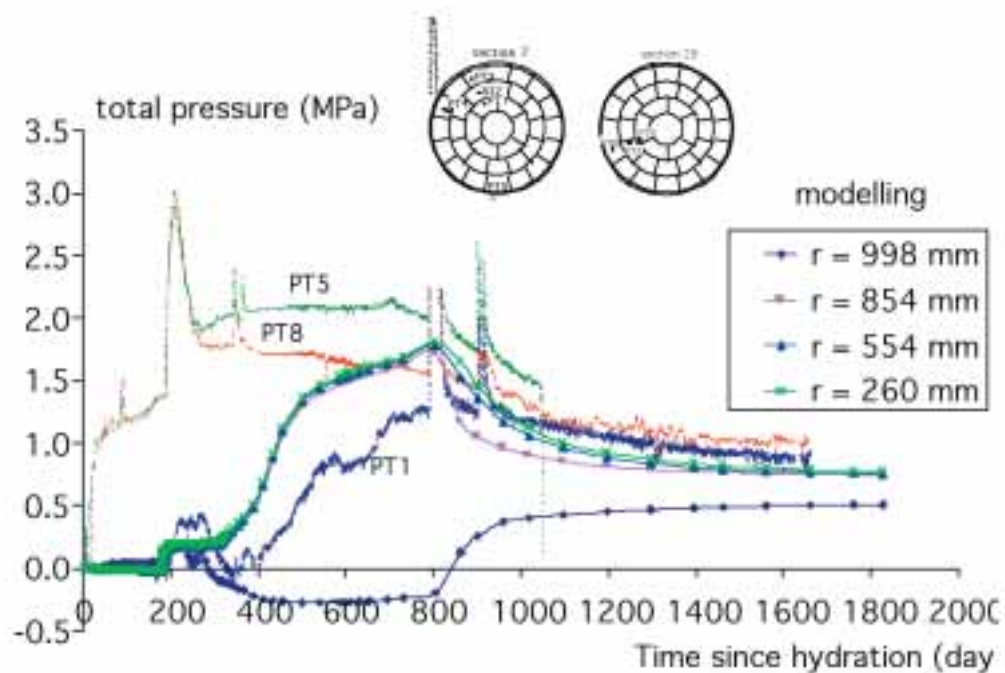


Figure 13: evolution of the total pressure: comparison between experimental data and simulation results (bolted lines)

## Heat transfer (temperature evolution)

During the operational stage, an unexpected observation was made: the temperature profiles revealed an apparently enhanced heat transfer mechanism. A value as high as  $4.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  for the thermal conductivity was derived on the basis of the temperature gradient between the dummy disposal tube and the external jacket and considering a heat power of  $450 \text{ W}\cdot\text{m}^{-1}$  and only conduction transfer mechanism. To explain the apparent high value of the thermal conductivity, a series of one-dimensional numerical simulations based on the THM properties obtained from the laboratory tests were carried out. Numerical results showed that, for such a material, the heat transfer is dominated by conduction. Other modes of heat transfer are insignificant: there are no evident effects on the radial temperature gradient. A detailed analysis of the temperature measurements at different components of the mock-up suggested that the high apparent thermal conductivity resulted mainly from the “loss of heat” through different components in different directions. Temperature measurements revealed that the hydration tubes and the two covers of the mock-up constituted the main sources of the heat loss, despite their isolation. A loss of as much as 15 to 18 % of heat was assessed. Moreover, the fact that the thermal conductivity increases with saturation played an important role in the observed temperature profiles in spite of its intrinsically high value. Taking into account the above-mentioned factors, the temperature evolution in the mock-up can be reproduced reasonably well by the simulations (Figure 14).

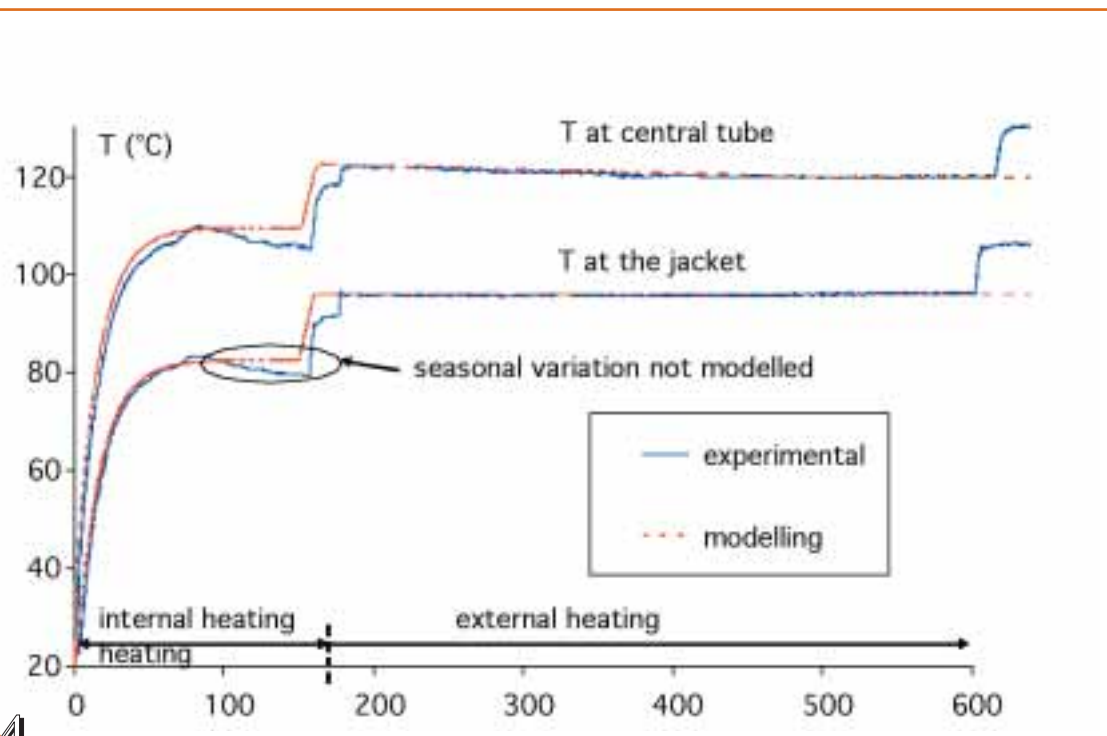


Figure 14: evolutions of the temperature close to the external liner and to the disposal tube: comparison between experimental data and simulation results. Only the evolution during the first 600 days - where the main heat transfer mechanism appeared - is simulated here

## Saturation

Direct measurement of the water content and dry density of the exposed material performed during the dismantling indicated that the buffer material did not reach saturation (95 % close to the disposal tube and 100 % close to the external liner).

However, numerical simulations suggested that the mock-up had reached saturation about 900 days after the beginning of hydration. In addition, the mass balance calculation based on the quantity of water injected and the total porosity of the mock-up also indicated that the mock-up was likely fully saturated.

This discrepancy between simulations and experimental results still needs to be clarified. Following uncertainties require further investigation:

- from the point of view of saturation measurement, the inevitable expansion of the samples after their deconfinement from the mock-up, which depends further on the “in situ” density, loss of water by evaporation before the on-field moisture measurements led often to underestimate the saturation degree;
- from the point of view of numerical simulation, considering hydraulic transfer at microstructure scale would be helpful to better understand the hydration mechanism of the buffer material.

Nevertheless, it is worth noting that the hydraulic conductivity of the exposed buffer material remained very low: between  $10^{-13}$  and  $5 \times 10^{-13} \text{ m}\cdot\text{s}^{-1}$ . The difference with that of the initial material (about one order of magnitude) can be explained by the variability of the blocks density and by microstructural changes (pore size distribution) induced by wetting, and enhanced by heating.

## Mineralogy

The mineralogical changes observed on the exposed buffer material were very limited. They cannot explain by themselves the modifications of the THM properties of the buffer material as described above.

The main, but limited, modifications concerned the presence of neoformed calcium sulphate crystals ( $\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$ : gypsum) at the contact of the clay buffer with the disposal tube, and at the interface with the stainless steel liner at the periphery. The ubiquitous presence, mainly in the joints, of gypsum crystals also indicated that the redox conditions were globally still oxidizing in the centre of the mock-up and inside the bentonite blocks.

No important iron sulphide recrystallisations have been observed inside the buffer blocks, nor in the joints between them. No clear redox-gradient could be evidenced in the mass of the buffer materials. Reducing conditions, related to sulphate-reducing bacteria, were only observed in the water of the hydration circuit. Some crystals of iron sulphide were also only observed by scanning electron microscopy (SEM) in a sandy lens extending below the buffer material and in some corrosion products. So, a very steep redox gradient would likely be observable, but only in a very narrow zone in the immediate vicinity of the hydration tubes.

**Corrosion behaviour of the metallic parts**

In November 1998, during the heating phase, water was seen seeping from cables of strain gauges installed on the disposal tube. A corrosion study indicated that these leaks were caused by pitting corrosion of the stainless steel protection of the cables.

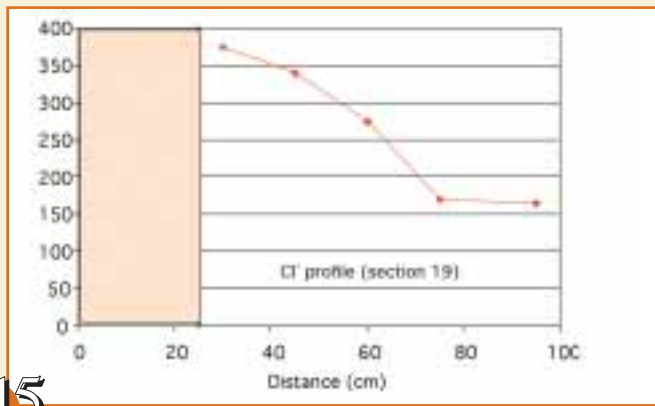
Chemical analyses of water from the leaks revealed a relatively high concentration of chloride ( $1 \text{ g}\cdot\text{dm}^{-3}$  instead of  $0.1 \text{ g}\cdot\text{dm}^{-3}$  expected for FoCa Clay water) originating from the buffer material itself. The concentration of other species (e.g., dissolved organic carbon, dissolved silica, or nitrates) was also higher than expected. Chemical analyses of major elements in water squeezed out from the exposed buffer material sampled during the dismantling later confirmed a process of enrichment of some solutes towards the disposal tube (Figure 15). At least two mechanisms could explain such enrichment:

- an advective transport of salts by a water front migrating through the unsaturated buffer material during the hydration phase, and;
- the Ludwig-Soret effect (thermo-diffusion): diffusion of a solute in a temperature gradient (non-diagonal coupled transport, Onsager’s reciprocal relation).

Another observation made during the heating phase, and relevant for the metallic corrosion, dealt with the presence, in the water from the hydration system, of dissolved sulphides at concentrations up to about  $6 \times 10^{-4} \text{ mol}\cdot\text{dm}^{-3}$ . Thiosulphates produced by the partial oxidation of sulphides during sampling operations were also detected ( $10^{-5} \text{ mol}\cdot\text{dm}^{-3}$ ) in some water samples. The presence of sulphides was also confirmed by the reducing conditions with very negative redox values ( $E_h$  between  $-250 \text{ mV}$  and  $-400 \text{ mV}$ ) that were measured on-line in the water injection system by means of a flow-through cell.

Microbially-mediated sulphate reduction was the most plausible mechanism to explain the presence of dissolved sulphides in the hydration system of the mock-up. It likely occurred at room temperature inside the mock-up during the hydration phase, or in the “cold” parts of the water injection system later during the heating phase. For this reason, microbial analyses of the water and of the (initial and exposed) buffer material have been performed.

The microbial analyses of water from the injection system have revealed the presence of high concentrations of different bacteria. More than 150 000 cells per ml of



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Figure 15: enrichment of chlorides towards the disposal tube

water were detected by classical culture and numeration for sulphate-reducing bacteria (SRB), thiosulphate-reducing bacteria (TSRB), and methanogenic bacteria.

The results of the microbial analyses performed by two different laboratories on the initial and exposed solid samples were less clear and sometimes contradictory, probably due to the difference in the detection techniques used, and to the location of the samples. However, when microbial activity could be detected in the solid, it was always low. The most striking feature was probably the detection in the exposed material of thermophilic bacteria in enrichment cultures at 80 °C.

Another unexpected observation made during the operational stage was the very low pH values. A pH of 5 was measured in the hydration system and a value as low as 3.5 was even observed in the water seeping from the defective strain gauges instead of 8.5 for the fresh injected NaHCO<sub>3</sub> water. Simultaneously, a high content of dissolved CO<sub>2</sub> was measured in the water. The source of CO<sub>2</sub>, or conversely the reason of acidification, remains however unclear. Part of the CO<sub>2</sub> could have been produced by the microbial activity in the hydration system. An alternative mechanism could be the production of CO<sub>2</sub> by the decarboxylation of the natural organic matter present in the FoCa Clay submitted to a moderate thermal stress. This process was already observed for kerogen isolated from Boom Clay and exposed to the same range of temperature.

Large amounts of dissolved organic carbon (> 750 mg C dm<sup>-3</sup>) were measured in the water. This organic matter could have contributed to fuel the

observed microbial activity. Its source still remains unclear: organic matter naturally present in the FoCa Clay or a contamination by hydrocarbons (mixture of gasoline and oil) released by defective Glötzl cells used for total pressure measurements (Figures 16). These cells appeared to have been severely damaged by a corrosion problem caused by an inappropriate choice of the metal of their envelopes.

Despite the unfavourable chemical conditions observed in the mock-up, the corrosion of the stainless steel liner (AISI 304) and of the disposal tube (AISI 321) in contact with the buffer material appeared to be very limited. Corrosion spots were only detected on the intrados of the liner. They were caused by technological artefacts, such as welding points used to fix cables of instruments and sensors. The central heating tube withstood even better, and no localised corrosion points could be detected, excepted in a small zone near the cover of the mock-up where the disposal tube was in contact with sand, and where a dissolved oxygen concentration gradient could have occurred in the beginning of the hydration phase.

The most striking observation dealing with corrosion in the mock-up was certainly the general corrosion of hydration tube # 07 exhibiting a green crust containing Cr<sup>3+</sup> oxides (Figure 17). A failed relative humidity sensor was discovered not far from this tube and it is very probable that the electrolysis induced by a direct current applied to this sensor during 5 years caused this very spectacular corrosion.



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Figures 16: severely corroded Glötzl cells responsible for the release of hydrocarbons in the pore water of the buffer material



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Figures 17: galvanic or electrolytic corrosion on the total length of hydration tube # 07

## Sensor performance

The OPHELIE mock-up has enabled the critical success factors for monitoring to be identified (Table 1). They are related to the sensor design (including cabling), to the installation, and to the sensor environment (considering also chemical conditions in addition to the thermal, hydraulic and mechanical strains). Detrimental effects such as water intrusion, galvanic corrosion, or thermal breakdown have to be avoided through an adapted design, validated by rigorous testing, and installed according to specific procedures.

Table 1: performances of the different sensor types used in the mock-up.

Sensor type	Installed	Failed	Comments
Thermocouples	100	3	Corrosion due to nearby relative humidity sensor
Internal strain gauges	27	27	Water ingress due to corrosion of soldered connections Drift effects due to water ingress, temperature variations and long term
External strain gauges	27	0	Good performance for short-term phenomena – Interpretation at long term difficult due to long-term and thermal drift Some possibility of thermal drift compensation by dummy gauges
Piezometers (vibrating wire)	6	5	Failed due to high pressure peak (4 x rated pressure) Connection between sensor body and cable not hermetically sealed
Total pressure sensors (buffer)	9	6	Good short-term performance, but poor sealing of sensor body at long term with water ingress. Connection between sensor body and cable not hermetically sealed
Relative humidity sensors	9	9	Irreversibly damaged in water-saturated conditions, with galvanic corrosion (stainless steel and copper)
Concrete segment sensors	8	8	Fragile due to hydraulic tubing Not suited for elevated temperatures, with rapid galvanic corrosion due to water-saturated environment

### **Thermocouples**

Most thermocouples – simple, robust, and stable devices – performed very well. Only three failed on a total of 100.

### **Strain gauges**

In general, the strain gauges that we applied (resistive type) performed well for short-term effects (a few weeks), but thermal and long-term drift made them less suited for long-term monitoring in the field, especially due to their sensitivity to the omnipresent water at high pressure and temperature.

The internal strain gauges placed on the disposal tube were not adapted to their environment. Although specified for the temperatures and water pressures that were present in the mock-up, they were attacked by the pore water (enriched by  $\text{Cl}^-$  during hydration). In particular, the solder at most sensor-cable joints had disappeared, with the stainless steel parts still intact. The thermal effects further interfered largely with the mechanically induced output signal. Because the expected mechanical strains were very low (due to the increased thickness of this tube), the final interpretation of the sensor output became too complicated.

By contrast, all the external gauges functioned well and showed the initial pressure increase during hydration according to calculations. In the long term however, the quantitative interpretation became too difficult due to drift effects, and were only able to show trends such as the decreasing total pressure.

### **Piezometers**

From the six piezometers, most failed when the water pressure reached values up to four times the specified range due to the pressure peaks. Apart from this aspect, the sensor design had a good potential to function in water-saturated media at elevated temperatures. It is therefore a good candidate to complement our conventional piezometers, which are based on a separate filter connected to an accessible pressure sensor.

### **Total pressure sensors (buffer)**

The total pressure sensors provided good measurement data during the initial phase of the experiment. With time, some sensors failed. Three were still functioning at the end. The insulation and continuity tests of the sensors' electrical circuits indicated that their wiring was probably affected by water ingress. This sensor type is well suited to measure the buffer swelling (as is the case in, e.g., the RESEAL experiment) provided the design and the electrical cables are adapted.

### **Relative humidity sensors**

The relative humidity sensors showed valid data right after the beginning of the hydration. After the start of the heating, however, most sensors showed an invalid output: capacitive sensing cells do not function in the presence of liquid water.

### **Concrete segment sensors**

Finally, the sensors in the concrete ring performed well initially, before failing because of the high temperature and corrosion problems. Their rather complex design made them prone to failure.

## Conclusions and lessons learnt

The OPHELIE mock-up simulated a section of a waste disposal gallery, in order to prepare the in situ PRACLAY Experiment and to review several technical aspects of its design. The mock-up focused on the engineered barriers of the disposal system: the buffer material, a mixture of FoCa Clay (60 wt %), sand (35 wt %) and graphite (5 wt %), the metallic disposal tube and the hydration system. The mock-up also allowed a large-scale investigation of the THM behaviour of the buffer material as well as of its interactions with the other barriers.

It was possible to demonstrate the feasibility to manufacture and to install the buffer in representative conditions and to better understand problems related to it. However, the manufacturing of the bentonite blocks remains an expensive process.

Globally, regarding its thermo-hydro-mechanical properties, the buffer material fulfilled its role: after four years of hydration and heating, it kept a low permeability and a high thermal conductivity. Although it swelled and filled all the technological voids, the swelling pressure remained low and the swelling process was not homogeneous.

It is difficult to conclude about the saturation degree reached by the buffer material. According to numerical simulations, a full saturation should have been achieved. However, direct measurements of the saturation degree indicated incomplete saturation (95 to 100 %). Nevertheless, the high and constant value of the thermal conductivity associated with the rapid swelling of the material would allow the waste to be installed before complete saturation.

Despite unfavourable chemical conditions (high chloride concentration, presence of free sulphides associated with sulphato-reducing bacteria), no significant signs of corrosion could be detected on the AISI 321 disposal tube in contact with bentonite. The resistance of the AISI 304 was barely lower. It exhibited a better corrosion resistance than expected: corrosion spots were only detected at the place of technological artefacts like welding points. These problems could be avoided in the future by using appropriate techniques.

However, the favourable behaviour of these two stainless steel alloys has to be confirmed over a longer period of time.

The lessons learnt from the instrumentation programme will be taken into account in the framework of the PRACLAY Experiment. Besides the loss of data, the failure of sensors can also jeopardise the performances of the engineered barriers (e.g., galvanic/electrolytic corrosion, or contamination by hydrocarbons, as observed in the mock-up). So, an extended preliminary selection of the sensors followed by a detailed test programme including corrosion aspects is necessary to guarantee the long-term reliability of sensors in harsh conditions with elevated temperature and pressure.

The OPHELIE mock-up highlighted the complexity to determine and to understand the main processes (THM, chemical, corrosion, ...) controlling the behaviour of the engineered barriers in a saturated environment at temperatures exceeding 100 °C.

Associated with the presence of chlorides observed in the buffer, these observations were at the origin of the decision taken by ONDRAF/NIRAS to re-examine in-depth the Belgian reference design for the disposal of vitrified HLW and to develop the concept of a supercontainer. A more homogeneous cement-based material offers, amongst others, the advantage to create long-term chemical conditions favourable for the lifetime of the metallic barriers. Moreover, cement-based materials are well known because of the large number of applications and existing studies. The new reference design also considers a lower linear thermal power resulting in temperatures below 100 °C in the engineered barriers.

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