EUROPEAN COMMISSION

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Long-term Performance of Engineered Barrier Systems

# Long-term Performance of Engineered Barrier Systems

# PEBS

## DELIVERABLE (D-N°:D6-2)

## Second Periodic Report

**Contract (grant agreement) number:** ***FP7 249681***

**Author(s):**

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| **Dissemination Level** | | |
| **PU** | Public | PU |
| **RE** | Restricted to a group specified by the partners of the PEBS project | |
| **CO** | Confidential, only for partners of the PEBS project | |

EURATOM

# PROJECT PERIODIC REPORT

| | |
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| **Grant Agreement number:** | **FP7 249681** |
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**Period covered:** **from 01.09.2011 to 28.02.2013**

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## DISTRIBUTION LIST

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[1] Usually the contact person of the coordinator as specified in Art. 8.1. of the Grant Agreement .

[2] The home page of the website should contain the generic European flag and the FP7 logo which are available in electronic format at the Europa website (logo of the European flag: http://europa.eu/abc/symbols/emblem/index_en.htm logo of the 7th FP: http://ec.europa.eu/research/fp7/index_en.cfm?pg=logos). The area of activity of the project should also be mentioned.

| | | |
|---|---|---|
| Christophe Davies | 1 digital file | no paper copy |

(D-N°: D6-2) – Project 2nd Period Report

Dissemination level : PU

Date of issue of this report : **27/05/13**

Declaration by the scientific representative of the project coordinator

I, as scientific representative of the coordinator of this project and in line with the obligations as stated in Article II.2.3 of the Grant Agreement declare that:

- The attached periodic report represents an accurate description of the work carried out in this project for this reporting period;
- The project (tick as appropriate) [3]:

  ☐ has fully achieved its objectives and technical goals for the period;

  ☐ **has achieved most of its objectives and technical goals for the period with relatively minor deviations.**
- The public website, if applicable

  ☐ **is up to date**

  ☐ is not up to date
- To my best knowledge, the financial statements which are being submitted as part of this report are in line with the actual work carried out and are consistent with the report on the resources used for the project (section 3.4) and if applicable with the certificate on financial statement.
- All beneficiaries, in particular non-profit public bodies, secondary and higher education establishments, research organisations and SMEs, have declared to have verified their legal status. Any changes have been reported under section 3.2.3 (Project Management) in accordance with Article II.3.f of the Grant Agreement.

Name of scientific representative of the Coordinator: .....Michael Mente.....

Date: .....27 / May / 2013

For most of the projects, the signature of this declaration could be done directly via the IT reporting tool through an adapted IT mechanism.

[3] If either of these boxes below is ticked, the report should reflect these and any remedial actions taken.

# Content

## 3.1 Publishable Summary

The PEBS project includes 7 work packages. Work packages 1 to 4 and the Chinese work package B are concentrating on research and technical work of RTD type. The work packages 5 and 6 focus on dissemination of results and the management of the action.

### 3.1.1 Work Package 1

The activities in WP1 are concluded and future work will be done with WP4. A final report describes current treatment of the early evolution of the EBS in safety assessments from a number of European national programs (state of the art). The description starts with an overview of the repository concepts. Both HLW and spent fuel repositories are covered as well as both clay and crystalline host rocks. This is followed by an overview of the assessment methodology used in the different programs. One important aspect of the methodology is the definition an application of safety functions. Basically, safety functions are a tool that is used for the evaluation of the performance as a function of time for individual repository components. The uncertainties in the early evolution in the EBS can generally evaluated with aid of the safety functions. The main part of the report describes the treatment of the THMC-process in the EBS and the potential impact on the safety functions. The significance of the identified uncertainties on the evaluation of the safety functions was also discussed. The report also summarizes the uncertainties and defines a number of cases or "scenarios" that will be assessed further within the PEBS project. Despite the differences in repository concepts, the safety functions defined for the engineered clay barriers are similar. The key processes occurring in the EBS in the early evolution of the repository that may affect the long the long-term performance are identical for all concepts on a fundamental level. However, the significance as well as the treatment of the processes in the safety assessment can differ between the concepts.

### 3.1.2 Work Package 2

The three main tasks of WP2 have progressed in the period, in general, according to schedule:

***Experimentation on key HM processes and parameters***

Activities are related with the maintenance and supervision of the EB experiment at Mont Terri, the elaboration of the test plan and the sampling book of the dismantling operation. The execution of the dismantling operation was completed by the first week of February 2013.

The hydraulic conductivity of samples of the EB granulate material (GBM) taken during the drilling of the two pilot boreholes was measured. The values obtained were related to the dry density and in the order of the hydraulic conductivity expected for samples of FEBEX bentonite compacted to the same dry densities. An increase in the salinity of the pore water of the samples of GBM with respect to that of the reference FEBEX bentonite was observed, due to the fact that Pearson water was used during the artificial hydration of the EB buffer.

A report including the results of the three long-term infiltration tests performed with the pellets mixture used in the EB in situ experiment was issued by CIEMAT. In addition, the long-term permeability tests for evaluation of the evolution of permeability over time and of the influence of hydraulic gradient on hydraulic conductivity are still run-ning. Three of them performed with the FEBEX bentonite started years ago, whereas two others performed with the MX-80 b

***Experimentation on key THM processes and parameters***

In situ activities are related to the maintenance and supervision of the Febex mock-up test at the Grimsel Test Site and of the HE-E test at the Mont Terri Rock Laboratory.

Two TH tests in cells performed with FEBEX bentonite have being running for more than 11 years and providing information about the temperature and relative humidity in the bentonite. These tests have highlighted the influence of the thermal gradient on the hydration kinetics and the slowness of the saturation process. In addition, two TH tests in cells were designed to reproduce the conditions in the in situ HE-E test and are being carried out with a sand/bentonite mixture and with MX-80 bentonite pellets. The heater at the bottom of the cells was initially set at 100°C and finally at 140°C. After the hy-draulic equilibrium was reached (redistribution of water by the effect of the thermal gradi-ent), hydration with Pearson water started. The two materials show a completely different behaviour: hydraulic equilibrium for both heater temperatures was reached much earlier in the sand/bentonite mixture. This mixture reached a high degree of saturation shortly after the initiation of hydration, whereas the progression of hydration in the pellets cell is very slow and accompanied by a steady increase in swelling pressure. To support these TH tests, an infiltration test at laboratory temperature with measurement of the swelling pressure is being performed with the MX-80 pellets.

Related to the study on stress-strain behaviour three series of unconfined compression tests have been accomplished during the second period where attempts to study the following factors were made: enrichment of sodium sulphate, access to relatively large amount of de-ionized water before preparation and direction of compaction. Compared to reference specimens no large deviations were seen on specimens tested in the mentioned series. In addition, four series were accomplished during the first period and a final series of tests has started where impact of increased temperature is studied. In this final series specimens are exposed to increased temperature during 24 hours in a laboratory oven and after the exposure the swelling pressure and shear strength are determined at room temperature

***Experimentation on key THM-C processes and parameters***

Related to the THM-C mock-ups, the problems previously detected in relation to the sealing of the I/O ports in instrumentation rods during the hydration have reappeared, so hydration is made by pulses to prevent leakage. Most of the sensors were replaced but, in spite of the cautions taken, they became damaged again. In the mock-up simulating the granitic host rock concept only five sensors remain operative. In the mock-up simulating the clay host rock concept (saline water injected through concrete blocks) many sensors, particularly the closest ones to the hydration surface, were damaged during the water injection events, the rest became damaged by the saline water in a few weeks. The relative humidity (RH) values measured by transmitters before breakdown were always around or higher than 90% and more homogeneous in the mock-up simulating the granitic host rock concept. The experimental setup is operative and the mock-ups are running under the following conditions: heating at 40°C and hydration by low-pressure pulses. Before failure the transmitters indicated high RH values and the expected temperatures, consequently corrosion processes must be going on inside the structures.

Related to the laboratory studies on interfaces, the two medium cells FB3 (iron/bentonite) and HB4 (concrete/bentonite) were dismantled after 4.5 years of operation and both cells were unsaturated when dismantled. In general, it has to be considered that mineral phases produced during the long-term alteration at the concrete-bentonite interface as well as the bentonite thickness affected by the alteration process are relevant in reaction and transport modeling validation. Results have revealed a <2 mm thickness of mineralogical alteration of FEBEX bentonite. However, the chemistry of the porewater and the exchangeable cations changes affected larger thickness than the mineralogical changes. Chloride was detected in the corrosion products from the iron-bentonite cell. Initial precipitation of chloride seems to play a relevant role in the first stages of the corrosion process, as it helps to initiate it. In this test, a deep blue iron oxide was found together with goethite and hematite. Its characterization confirmed the formation of Fe(II) and mixed Fe(II)/Fe(III) phases (ferrous hydrox-

ide and magnetite). However, no alteration of the bentonite was observed. The distribution of exchangeable cations varied along the bentonite column: exchangeable sodium increased close to the hydration surface and dropped near the interface, whereas magnesium decreased in saturated zones and increased near the heater. The tests FB4 (iron/bentonite) and HB5 (concrete/bentonite) will be dismantled on April 2013, after almost 7 years of operation.

The experiments performed in small cells proposed to simulate repository times ranging from 1000 to 3000 years were dismantled in September 2012. The experiments ran isothermal (60°C) during 18 months and when they were dismantled the bentonite was fully saturated. After dismantling and analysis it was found a back-diffusion of chloride from the cells towards the hydration bottle; a remarkable increase of chloride content was analysed on the hydration solution after dismantling the experiments. Diffusive transport of magnesium from the hydration solution towards the cells took place. In addition, high concentrations of chloride and sulphate were found both in mortar and in magnetite. The cells with mortar exhibited a 1-3 mm rim of mineralogical and physical (BET, porosity) alteration, which is similar to that found in medium cells (PEBS, NF-PRO) or in other experiments conducted in ECOCLAY. However, no dissolution features were observed in magnetite and the iron content did not increase in the vicinity of the magnetite/clay interface. No iron-oxide precipitation or iron-rich newly-formed phases at the magnetite/clay interface were observed.

### 3.1.3 Work Package 3

The five main tasks of WP3 are all well advanced and, in general, according to schedule. Some delays in the preparation of deliverables do not affect the overall project schedule.

- HM modeling of the EB experiment

  CIMNE performed coupled hydromechanical calculations using an enhanced double structure formulation in which it is considered that the micro water is immobile and only exchanges with the water in the macropores take place. Potential variation of water density in the microstructure is included in this model. It is found that the increase in microporosity during hydration is significant and has modeling implications, for instance in the simulation of the injected flow rate.

- THM modeling of the planned heater test HE-E

  Design and predictive modeling of the HE-E experiment were finalized and published in Deliverable D3.2-1. Development of new or improved constitutive models and identification of parameters for the engineered barrier materials of the HE-E test were continued. In particular, CIMNE has performed extensive modeling of the HE-E test using a coupled THM formulation and a revised dataset for parameters taking advantage of the new information becoming available as the project progressed. A variety of geometric models has been used and both NAGRA and GRS buffer materials have been specifically considered. Respective documents are in preparation. The provisional dataset used for the design calculations was revised and new data obtained in WP2 were integrated.

  A full interpretation of the HE-E dataset based on the monitoring data and the outcomes of the modeling after 15 months of monitoring was performed and summarized in a paper submitted to the Geological Society Special Publication which resulted from the Clay Conference in Montpellier in 2012.

- THM modeling of bentonite buffer

  In deliverable D3.3-2, an overview of the tasks in the Swedish safety assessment modeling report that included bentonite buffer was given. Descriptions, main results, and discussions about uncertainties/limitations/inadequacies in material representations were given for tasks considered most relevant for the PEBS framework. Several of the issues discussed in this deliverable relate to a lack of generality and couplings in the used models. It was also mentioned that for the systems we consider, good agreement has commonly been obtained between theories based on thermodynamical/chemical considerations and experimental findings. Studies of more general theories for mixtures, which use thermomechanics as a basis when formulating the material representations, is therefore considered interesting. Consequently, Clay Technology has been working on an overview of "thermomechanically" based mixture theory and a relevant material model within this theory that is applicable for systems consisting of bentonite (montmorillonite), water and air. This will be documeneted in a PEBS report.

  CIMNE has performed extensive analyses of the long term THM cell tests performed by CIEMAT incorporating a variety of new processes, such as threshold hydraulic gradient, thermo-osmosis, and a double structure formulation. In addition, analyses combining double structure and thermo-osmosis have been undertaken. A significant improvement of the predictions is obtained with these enhanced models, although it is complex to identify the most relevant features on the basis of numerical analyses alone. However, analyses combining more than one new process appear especially promising. Similar analyses of the mock-up test have been initiated. CIMNE has also performed coupled THM analyses of CIEMAT column tests carried out using the buffer materials (NAGRA and GRS) employed in the HE-E tests. Two different stages have been modelled: i) a purely thermal one followed by ii) hydration. The first stage has allowed to validate the vapour transfer formulation with satisfactory results. The comparisons with the results of the hydration phase are ongoing but first results are very encouraging.

  Inverse modeling of the FEBEX in situ experiment using iTOUGH2 was started by TK Consult within the reporting period. The aim is to apply "simple" TH-model(s) to simulate the long term FEBEXe in situ test (instead of "expensive" THM) and evaluate if with this simplified TH approach to represent THM processes the main characteristics of the data can be captured. For that task an inverse modeling framework was developed that allows for automatic sensitivity analysis and parameter estimation based on Maximum Likelihood estimation theory. This was subsequently applied to the FEBEX data base of 15 years in situ measurements. Once the optimal parameters are identified an extrapolation by simulating long term behaviour (including uncertainty analysis) is foreseen.

- Modeling of THM-C experiments on bentonite buffer

  Improved THMC models have been tested with data from several types of heating and hydration experiments performed by Ciemat, including different tests on the canister/bentonite and the concrete/bentonite interfaces.

  Regarding the corrosion experiments, the THm model was calibrated with water content and dry density data measured at the end of the experiment. The kinetic parameters of magnetite precipitation were calibrated by using Fe weight content data which are representative of the precipitation of Fe(OH)2(s). For the most part, simulations agree well with experimental data. Model results indicate that: 1) The main properties of the bentonite remain unaltered; 2) There is a sequence of corrosion products, Fe(OH)2(s) and magnetite being the end members; 3) Fe2+ is sorbed by surface complexation; 4) Fe2+ cation exchange is less relevant than Fe2+ sorption; and 5) Corrosion products penetrate a few mm into the bentonite.

For the concrete experiments, the THM model was calibrated with the water content and dry density data measured at the end of the experiment and temperature and relative humidity measured during the test. The parameters of the reactive transport model were calibrated with aqueous extract data measured at the end of the test. For the most part, simulations agree well with experimental data.

- Extrapolation to repository long-term evolution

  In order to provide input to WP4, several long-term simulations are planned. Modeling cases and case variants for the long-term extrapolation exercises were agreed upon during several meetings. The finalized case table task comprises the following simulation tasks:

  1. Isothermal buffer evolution: Long-term extrapolation of the EB experiment configuration to investigate seal performance
  2. Thermo-hydro-mechanical evolution of the buffer at temperatures up to 100 °C: This involves long-term extrapolation of the FEBEX in-situ test with two variants – one using a constant heater temperature of 100 °C and the other using a heat decay curve representative for real HLW
  3. Thermo-hydro-mechanical evolution of the buffer (T > 100 °C): Long-term extrapolation of the HE-E using a realistic source term: Three modeling teams will be involved (CIMNE, NAGRA/TKConsult, GRS)
  4. Geochemical evolution, especially at interfaces: Involves studies of the canister/bentonite interface, the concrete/bentonite interface, and the long-term evolution in granite

  The actual simulations are currently prepared. The models required for long-term extrapolation have been developed and tried, although they also continuously improved to incorporate the progress in understanding and the new information generated by the project and elsewhere.

  UDC has performed numerical modeling for the long-term hydrochemical evolution of the bentonite barrier of a spent-fuel, carbon-steel canister repository in granite. The model accounts for canister corrosion, the chemical interactions of corrosion products and bentonite, mineral dissolution/precipitation, Fe2+ and H+ surface complexation reactions on three types of sorption sites and cation exchange reactions. The model considers the generation of $H_2$(aq) which is allowed to diffuse through the bentonite. Model results indicate that canister corrosion causes a marked increase in pH and the concentration of dissolved Fe2+ and a decrease in Eh. Most of the released Fe2+ diffuses from the canister into the bentonite where it precipitates mainly as magnetite and to a lesser extent as siderite. Fe2+ sorbs by surface complexation on weak sorption sites and undergoes cation exchange. Sorption plays a relevant role in the geochemical evolution of bentonite. The competition of Fe2+ and H+ for the sorption sites near the canister/bentonite interface causes several sorption fronts. Model results lead to significantly high $H_2$(g) pressures. The reduction of bentonite porosity due to mineral precipitation near the canister/bentonite interface could be large and result in the clogging of the bentonite pores.

### 3.1.4 Work Package 4

As results in the project gradually become available, in the second reporting period the basis was developed for reaching the objectives of WP4 with as a main purpose summarizing and integrating the results and relate the results and uncertainties to the long-term safety functions of the repository components and to the overall long-term performance of the repository,

During the second 18 month reporting period the main objective was to initiate the process for reviewing the findings of the WP2 and WP3 experiments and models in order to develop a more complete qualitative process-related description of the early evolution phase of the repository (the first several hundred years) and the residual uncertainties in the evolution. The 4 cases identified as a result of WP1-4 interactions namely: Case 1, uncertainty in water uptake in buffer (T< 100°C), Case 2, uncertainty in T evolution in buffer (T >100°C), Case 3, uncertainty in HM evolution of buffer and Case 4, Uncertainties in chemical evolution, served as a basis for that. Based on the defined Cases, in close interaction with WP2 and WP3, a detailed questionnaire addressing the process understanding and uncertainties therein was developed to guide the integration of the project results. This questionnaire will allow to structure the outcomes as the results of experiments and models and their significance in relation to long-term safety functions of the buffer, canister and host rock (clay and crystalline) are planned to be discussed in a quantitative (where possible) fashion, also including the importance of residual uncertainties.

The planning for an integration workshop covering WP2-3-4 activities, which will take place in September, and identify the progress made in the project beyond the current state- of-the- art, started within this reporting period.

### 3.1.5 Work Package B

- The mock-up operations so far are running very well and better than proposed. The complete sensor system was initially proposed for a reduced liability, but it runs very fine.

- So BRIUG decides that the China-Mock-up will not be dismantled within the PEBS project period.

- This implies that two of the period's reports (DB-3 and DB-4) and one report of the final period (DB-5) cannot be established within project implementation. The experts of CNNC and BRIUG discussed this issue in detail. The China-Mock-up is the first large-scale THMC coupled experiment in China, and its successful operation will play an important role in investigating the long-term behaviors of GMZ-bentonite. Hence, the decision was made that BRIUG will continue to perform the experiment.

### 3.1.6 Work Package 5

The base for all dissemination efforts was compiled in the COMMUNICATION ACTION PLAN what gives the frame for the PEBS NEWSLETTER and the PEBS WEBSITE.

BGR prepared 3 additional Newsletters to inform about actual results and progress.
Following additional requests a PEBS POSTER will be prepared. The preparation of the BENTONITE WORKSHOP and TRAINING is advanced but has to be postponed for one year because of an unforeseen accident. The REGULATORY WORKSHOP was implemented with experts from 10 regulators.

NAGRA presented PEBS at the International Conference on Environmental Remediation and Radioactive Waste Management ICEM 2011 in Reims. NAGRA presented PEBS at the ANDRA conference in Montpellier, October 24 and 25, 2012. BGR and SKB presented PEBS at the EURAWASTE conference in Montpellier at October 26, 2012. BGR presented PEBS at the Waste Management Symposium 2013, February 24 to 28, 2013, at Phoenix, Arizona. The PEBS Final Conference, February 6 and 7, 2014, is in preparation. A first flyer is distributed at several conferences and workshops in Europe and USA.

CIEMAT and UDC prepared 3 publications for scientific magazines.

BGR maintains the PEBS website. All approved public reports were posted for downloading. Now 48 Deliverables were prepared. 43 are unclassified.