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# CEBAMA

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## Deliverable D1.02

# Agreement and documentation of systems to be studied (M06 - Nov 2015)

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| **Dissemination Level** | | |
| **PU** | Public | X |
| **PP** | Restricted to other programme participants (including the Commission Services) | |
| **RE** | Restricted to a group specified by the partners of the CEBAMA project | |
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**ABSTRACT:**

The deliverable report serves as a summary of the cement-bentonite interaction systems to be studied. It builds upon the original project plans and provides more concise details pertaining to the synergy between experimental work on various materials. Overview tables are provided, followed by Appendices summarizing the experimental material systems to be used by the 19 partners of WP1.


**RESPONSIBLE PERSON(S)**:

WP1 co-leaders: Erika Holt (VTT), Francis Claret (BRGM), Urs Mäder (UniBern)

**MAIN TEXT:**

Understanding the cement-bentonite interaction is experimentally studied on a variety of samples, with different degrees of ageing and environmental exposure. An overview of the systems to be studied is given in Table 1, categorized according to the level of leachate pH and the interface material. Some of the 19 partners involved in WP1 are studying multiple systems, thus partners may be listed in more than one category. A more elaborate classification of the systems to be studied is given in Table 2. The detailed description of these interface studies and materials can be found in the Appendices. Further details about the experimental methods and materials under investigation are developed during the first phase of the project, and will be reported in WP1 Deliverables D1.04 and D1.05, respectively.

*Table 1. Summary of concrete-bentonite systems under investigation.*

<table>
<tr><th>Cement material</th><th>Claystone</th><th>Bentonite</th><th>Other rock</th><th>Aged interface</th><th>Fresh interface</th><th>Interface to solution</th></tr>
<tr><td>OPC</td><td>SCK CEN</td><td>KIT, CIEMAT</td><td>CIEMAT</td><td rowspan="3">CIEMAT, RWMC, BGS, BRGM, SCK CEN, UJV, HZDR, LML, UAM, CSK, UNIBERN, IRSN</td><td rowspan="3">TU Delft, UJV, CTU, UAM, SCK CEN, ANDRA, IRSN</td><td rowspan="3">KIT, USFD, ULOUGH, VTT</td></tr>
<tr><td>OPC + LPH</td><td>BRGM, ULOUGH, HZDR, UNIBERN, IRSN, TU Delft</td><td>UJV, CTU, RWMC, HZDR, UAM, CSIC</td><td>CSIC, ULOUGH, USFD</td></tr>
<tr><td>LPH</td><td>LML, ANDRA</td><td>VTT, BGS</td><td>USFD, ULOUGH</td></tr>
</table>

*Table 2. Details about concrete-bentonite systems under investigation.*

| | | Type of experiment | Transport | Scale | Parameters | | | Analytical methods | | Coop within WP1 | Input for WP3 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | physical | mechanical | mineralogical | bulk | 2D | | |
| Partner | | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| 1 | KIT | diffusion in columns, interfae evolution in batch | diffusion | diffusion: mm - cm, porosity changes: nm - µm scale | porosity, geometry, diffusion coefficients | no | yes | XRD, SEM, Raman (?), AFM (if posible) | depending on SEM: µ-XRF, autoradiography | AMPHOS, BRGM, PSI, UJV | Diff. const = f(time), change of pore size, pore patterns, RN retention (Kd) relevant phases |
| 4 | BRGM | Examination of existing interfaces | | multi scale (cm to nm) | porosity, geometry | none | Multi techniques approach | XRD, IR, NMR, Raman, etc.. | SEM, EPMA, Autoradiography,... | SCK, ANDRA, UniBE | Pore network distribution latter used to support lattice Boltzmann simulation |
| 5 | BGS | 1. Aging of concrete batch testing; 2. transport testing shear apparatus | advective | multi scale (micron to cm) | Porosity, hydraulic conductivity, density, saturation, roughness of interface | Swelling properties of COx, shear properties (strength, friction etc) | yes | XRD | SEM, Optical, fluorescence | | |
| 6 | CIEMAT | Column experiments, FEBEX in situ experiment | adevective plus diffusive | µm, mm, cm | BET, porosity, permeability | none | yes (+ aqueous species mass balance) | IC, ICP-OES, CRDS, Optical Microscopy (+CL), FTIR, MIP, BET, XRD, RBS, uPIXE | SEM-EDX, TEM | CSIC, UAM | UDC (HB cells) |
| 7 | TUDelft | Electromigration | | µm, nm | porosity, geometry | none | yes | neutron activation analysis, XRD | gamma-counting, positron annihilation | NRG | reactive transport processes |
| 8 | FZJ | Granular column of plug samples | none | multi-scale (cm to nm) | porosity, geometry | none | none | Electrical impedance spectroscopy (EIS or SIP) | | BRGM (+VTT for NMR?) | Measurements of complex electrical conductivity |
| 12 | RWMC | (1)Long term immersion test and/or structure (2)1D compression test | | micro-m to cm | | (2)Dry density and compression stress relation, swelling pressure during Na-Ca exchange | | XRD, SEM-EDS and Phase Shift Interferometry | Ca-XAFS, Phase-shift interferometry and Atomic Force Microscopy | | (1)Dissolution and/or growth ratio of Zeolitic secondary minerals (2)Distributionnof altered,i.e. precipitation of secondary C-S-H and dissolutionof montmorilinite (3) change of mechanical properties due to chemical degradation, i.e. change of mineralogical conposition |
| 13 | SCK | diffusion, column, batch | diffusion, advection | multi scale (cm to nm) | hydr cond, diffusivity, porosity, surface, pore size distrib, cations, anions, pH | none | yes | XRD, TGA, N2 adsorption, MIP (+collaboration BRGM) | SEM-EDX (limited) (+ collaboration with BRGM) | BRGM, HZDR | transport properties (hydraulic conductivity, porosity, diffusivity), combined with µstructural analysis (BRGM) used to support lattice Boltzmann simulation |
| 15 | UJV | 1/ lab-aged (CTU) 2/ URL Josef 3/ diffusion study after interaction | diff | cm | porosity, anions, cations, pH, CEC, extent of alkaline front | Concrete: uniaxial strength | chemical analyses, mineralogical changes | XRD; LSC (tracer); gamma /AAS (Sr, Cs) | | CTU, VTT, SURAO | Diffusion parameters in relation with changes of material properties due mutual interaction ; material characterisation |

| | | | |
|---|---|---|---|
| diffusion / in situ / batch / electromigration | column / advection+diffusion | characterisation only | geomechanical tests |

*(continues)*

*(Table 2, continued)*

| | | Type of experiment | Transport | Scale | Parameters | | | Analytical methods | | Coop within WP1 | Input for WP3 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | physical | mechanical | mineralogical | bulk | 2D | | |
| Partner | | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| 17 | ULOUGH | Diffusion | Diffusion | nm - cm | porosity, permeability | none | yes | XRD, NMR, Raman, EXAFS, XANES | SEM-EDX, dig. Autorad. | USFD, Amphos | Data for speciation-solubility & chemical transport modelling |
| 19 | CTU | 1/ ageing - interaction in tests 2/ samples from Josef "cartridges" | adv | cm | | Bentonite: swelling P, hydr cond, retention, liquid limit; Concrete: strengths | no | no | no | UJV, VTT | Together with UJV - data sets of diffusion parameters and mechanical properties changes due to mutual interaction |
| 20 | USFD | Batch | Diffusion | nm,µm, mm | porosity, permeability, tortuosity | none | Support Ulough chemical characterisation | MIP, gas adsorption, oxygen permeametry, NMR | µXAS, neutron radiography, SEM/EDX... | ULOUGH, (?) HZDR | Diffusive transport model as a function of cement, curing time & groundwater comp. |
| 21 | VTT | Batch | | nm,µm, mm | C-S-H,anions, cations, pH | Pozzolanic reaction degree (strength) | | ICP, NMR, XRD | SEM-EDX, TEM | Posiva, Andra, UJV/CTU, CSIC (?) | Verification of geochemical simulations of C-S-H behaviour in low pH concrete |
| 22 | HZDR | confined column | adv + diff, one phase | sentitive to ~10k atoms in 1mm³ | changes of and total eff. porosity, permeability, in 3D | like SCK and like UniBE | like SCK and like UniBE | like SCK and like UniBE | Autoradiography | SCK, UniBe, PSI | Quantiative 4D tracer distribution in clay, cement and their interfaces as benchmark data set |
| 23 | LML | direct shear in triaxial cell | water flow | cm | porosity, rugosity | elastic, strength | carbonation effect | | | ANDRA | Hydromechanical data |
| 24 | UAM | *Core-interface<br>*mm cell infiltration interface | adv+diff | nm,µm, mm | BET, porosity (CIEMAT or CSIC collaboration) | none | C(A)SH, MSH, carbonates | Small size samples (mg) XRD, GI-XRD, FTIR | SEM-EDX, SEM-CL, TEM | CSIC, AEG-CIEMAT, Uni-Be, | UDC (HB cells) |
| 25 | CSIC | percolation test | Permeability and diffusion | µm, mm | Chemical/mineralogical changes, porosity | | yes | Bulk methods: XRD, TGA, IR ... | Mapping methods: SEM-EDX, | CIEMAT-AEG/UAM, VTT | Data from long-term HB6 cell from concrete OPC aging, CSH evolution (UDC) |
| 28 | ANDRA | in situ | diff | multi scale (µm to dm) | shrinkage vs hydration, porosity | mechanical strength | composition vs hydration | vibrating gauges, TDR | SEM, XRD | BRGM, LML | HM behaviour of low pH concrete, atm. chemical evolution, chemical evolution at COx/low pH interface |
| 30 | UNIBERN | confined column | adv + diff | µm, mm | porosity, geometry | none | yes | XRD, WC | SEM-EDX, radiography | HZDR, BRGM | |
| 31 | IRSN | Tournemire 70°C (CEMTEX) - 1 D experiments | Diffusion | nano, µm, mm | porosity | as option | Multi-technic | XRD, TEM, BET, TGA,... | SEM-EDX | UniBe?, BRGM? | |
| | | | | | | diffusion / in situ / batch / electromigration | column / advection+diffusion | characterisation only | geomechanical tests | | |

**APPENDIX: Detailed Answers per Partner, regarding their systems to be studied**

**1. KIT/Bernhard Kienzler, Vanessa Montoya**

- The systems investigated by KIT include hydrated OPC (CEM I) samples of 28 days setting (as reference). Priority is given to low pH concretes produced according SKB R 12-02. These samples have been stored for ~one year at room temperature under saturated vapour.
- The solution used for equilibration of cement/concrete samples before starting the diffusion experiments will be a bentonite water according to Fernández, A. M.; Baeyens, B.; Bradbury, M.; Rivas, P., Analysis of the porewater chemical composition of a Spanish compacted bentonite used in an engineered barrier. Physics and Chemistry of the Earth, Parts A/B/C 2004, 29, (1), 105-118.
- Batch interaction experiments for determination of equilibrium conditions.
- The cement/concrete samples are dressed to the appropriate size, fixed in epoxy resin, cut to appropriate length, surfaces polished. The embedded samples will be closed by lids equipped by tubing for the diffusion experiments. The samples will brought in contact with excess Opalinus pore water. HTO and anionic tracers will be used to test the diffusion properties.

After reaching constant pH, the cement/concrete samples will be cut parallel to the direction of the diffusion and investigated by different methods.

**3. BRGM/F. Claret**

Two systems will be studied in close collaboration with SCK-CEN:

- A Boom Clay/concrete interfaces will be sampled in the HADES underground research facility (URF) (Mol, Belgium). Samples will be taken from the Connecting Gallery, in which low-permeability high-pH concrete wedge blocks are in contact with Boom Clay for 12 years. A technical meeting is planned the 2$^{nd}$ of December 2015 to plan the sampling
- Percolation type experiments. Study of the interface Boom Clay/cement by means of lab experiments with low-strength high-porosity high-pH cement mixture representative for the backfill composition in the Belgian disposal concept (NRVB-type). In these experiments, the chemical conditions can be rigorously controlled. The conditions in the HADES URF will be mimicked (advective flow regime from the clay to the cement). A set of experiments will be set up which can be stopped as function of interaction time. For each interaction time, 2 experiments are performed in parallel, one will be used for measuring the diffusive parameters (by using radiotracers or dissolved gas – to be decided), the other one is foreseen to conduct chemical and µ-structural analysis.

**4. NERC-BGS/Rob Cuss**

Examining interface properties from fresh to aged, investigating how transport properties vary on a yearly interval during the life of the Cebama project. Test specimens are prepared at the start of the experimental program. Samples of Callovo-Oxfordian claystone will be re-hydrated and then sheared to create rough fracture surfaces; this will give information on rehydration and the shear properties of intact COx. Ten samples will be prepared with a rough surface, with an additional ten samples prepared with a planed, smooth surface. Two types of COx will be used; clay-rich COx from repository depth and higher carbonate content COx from shallower depth. Cement will be prepared and cast directly on the COx surface. Samples will be stored under stress in a solution to aid aging of the cement and interface until testing. Each experiment will examine the transport properties of the interface during shearing (until failure), giving information on both the changes in flow and strength as the interface ages. Post-test petrology will define the degree of chemical aging.

## 5. CIEMAT/María Jesús Turrero/Elena Torres

CIEMAT investigations include the following samples/systems:

A. **<u>Aged interfaces</u> between cementitious materials (OPC) and engineered barrier components, specifically bentonite.** The aim of this work is to evaluate the up-scaling effect, specially focused on the transient state of a repository, by means of the study of the concrete-bentonite interface at different scales: laboratory tests and the real-scale FEBEX experiment. For this purpose, geochemical processes and specific transport properties will be investigated.
   - **Concrete/bentonite coming from a full-scale engineered barrier system** (EBS), the FEBEX experiment at the Grimsel Test Site (GTS). The experiment is based on the Spanish reference concept in crystalline rock in which the canisters are placed horizontally in drifts and surrounded by a clay barrier constructed of highly compacted bentonite blocks. Heating started in 1997 and since then a constant temperature of 100°C has been maintained while the bentonite buffer has been slowly hydrating in a natural way (ENRESA 2000). After five years of uninterrupted heating, the heater closer to the gallery entrance was switched off. In the following months the concrete plug, heater and all the bentonite and instruments preceding and surrounding it were extracted. The remaining part was sealed with a fib-reinforced shotcrete (CEM II). New sensors were installed in the buffer, and a second operational phase started and was running (ENRESA 2006) until dismantling along 2015 (FEBEX-DP, http://www.grimsel.com/gts-phase-vi/febex-dp) after 18 years of continuous heating and natural re-saturation. **The new shotcrete/bentonite interface has been operating for 13 years** and will be studied in the context of CEBAMA**.**
   - **Concrete/bentonite coming from the dismantling of a small scale laboratory experiment (HB6)** planned to simulate as close as possible repository conditions. A 30-mm of OPC based concrete slab (CEM I) was placed over a 70-mm of compacted FEBEX bentonite cylinder in a hermetic cylindrical cell. The materials were subjected to the thermal and hydraulic gradients similar to the ones expected in the repository. The experiment started operating in 2006 during NF-PRO European Project and will be dismantled in 2016 after **10 years of continuous operation.**

B. Laboratory experiments with **<u>fresh high pH concrete</u> (CEM I / CEMV)** to study the porosity changes in OPC associated to chemical perturbations, with special focus on their possible effects on RN transport. The impact on solid porosity of groundwater changes in pH, redox, sulphates, carbonates and calcium will be analysed.

**6. TUDelft/Denis Bykov**

The object of study is the interface between low and high pH cementitious materials (CEM-III/B 42.5 LH HS (concrete); CEM-I (foamed concrete); CEM-III/B 42.5 LH HS (foamed concrete)) and the Dutch Boom clay. The materials chosen for investigation are essentially relevant to the Dutch disposal concept. In addition, a comparison between concrete made with blast furnace slag and Portland cement is expected to stimulate the information exchange with other partners within Cebama. The interface will be fresh (months or maybe years). In addition to Boom clay, bentonite can be also investigated if the experimental time is left. The benefit of examining bentonite is that the organic matter in bentonite is insignificant compared to Boom Clay. The organic matter can have a large buffer capacity to alkaline fluids. Thus, for conceptual understanding of the impact of organic matter it can be worthwhile to include bentonite as well.

**10. RWMC/Hitoshi Owada**

RWMC researches on the chemical reaction around interfaces between bentonite and OPC or Fly-ash mixed cement. Kunigel-V1 and MX80 are selected as the bentonite material. 5 to 15 years immersion tests have been carrying out by using cement-bentonite coupled specimen for the observation of the expansion of altered zone around C-B interface by using Ca-XAFS and other analyses. Observation of coupled specimen of OPC/FAC concrete and bentonite-sand mixture obtained from former GMT test facility of Grimsel Test Site will also be carried out.

Additional information for the calculation in WP3: Dissolution rate of montmorillonite under the compacted and alkaline condition had been obtained by using Vertical Shift Interferometory (VSI) as a function of Temperature, pH and Pressure. (VSI is a kind of phase shift interferometer). Growth ratio of some zeolitic minerals will also be obtained.

**11. SCK CEN/Norbert Maes; Quoc Tri Phung (with input from F. Claret, BRGM)**

Studied systems: OPC, Boom clay, fresh + aged interface (concrete – Boom clay) → as described in Table 1. Two systems will be studied in close collaboration with BRGM:

- A Boom Clay/concrete interfaces will be sampled in the HADES underground research facility (URF) (Mol, Belgium). Samples will be taken from the Connecting Gallery, in which low-permeability high-pH concrete wedge blocks are in contact with Boom Clay for 13 years. A technical meeting is planned the 2nd of December 2015 to plan the sampling
- Percolation type experiments. Study of the interface Boom Clay/cement by means of lab experiments with low-strength high-porosity high-pH cement mixture representative for the backfill composition in the Belgian disposal concept (NRVB-type). In these experiments, the chemical conditions can be rigorously controlled. The conditions in the HADES URF will be mimicked (advective flow regime from the clay to the cement). A set of experiments will be set up which can be stopped as function of interaction time. For each interaction time, 2 experiments are performed in parallel, one will be used for measuring the diffusive parameters (by using radiotracers or dissolved gas – to be decided), the other one is foreseen to conduct chemical and µ-structural analysis.

**13. UJV/Petr Večerník, Radek Červinka, Václava Havlová**

**Materials**

**Bentonite** Czech bentonite (Ca-Mg type); commercial product of Keramost, plc. company, laboratory labell "B75" is used.

**Cement** - two cement types are used: ordinary and low pH (cooperation with VTT, Finland) cements.

**Cement-bentonite systems**

Two types of experimental systems are used: laboratory and in-situ.

**"In-situ experimental system"**

In-situ experiments were designed in the following way: patrons (perforated tubes), filled with a combination of bentonite samples (cylinders, bulk density of 1.65 $g/cm^3$) and cement material (cylinders, bulk density of 1.8 $g/cm^3$) were placed into prepared boreholes in the URL Josef in 2010 The aged samples will be obtained from this experimental system,.

**"Laboratory experimental system"**

Laboratory experiments were designed in the following way: cement samples were placed and surrounded by bentonite suspension in the pressure vessels (experimental reactors). CEG CTU is responsible for sample preparation has responsibility, ÚJV is responsible for bentonite suspension preparation.

New samples will be obtained from this experimental system.

Chemical, physical and mineralogical changes will be studied on materials interfaces.

**15. ULough/Matthew Isaacs, Monica Felipe-Sotelo, David Read**

The interaction of cements with synthetic groundwaters will be studied using radioactive tracers ($^{3}H$, $^{14}C$, $^{35}S$, $^{36}Cl$). The cement samples will be prepared and provided by the University of Sheffield (USFD) and will be identical to those used by our partner. Three groundwaters are to be investigated, representing a saline system, a crystalline, meta-sedimentary facies and a clay groundwater (Corallian-Oxfordian). The experiments will provide insight into the chemistry occurring at the interface of the cements with the respective groundwaters, in particular, the kinetics of precipitation – dissolution reactions and the effect of the groundwater solutions on the composition of mineral solids in the cements.

## 16. CTU/Lucie Hausmannova

CTU will study effect of interaction of cements and bentonite on mechanical properties. Selected samples will be heated. Several combinations will be studied (see Table 1).

The following materials will be used:

- Czech Ca-Mg bentonite (brand name: Bentonite 75). Industrial product of Keramost a.s. Keramost a.s. mines are located in north-east part of Czech Republic,
- Portland cement - CEM II 42,5 R (same cement was used in previous experiment, it is described in **Error! Reference source not found.**)
- Low-pH cementitious binder (cement+flyash+silica fume) from VTT and
- Underground water from Josef gallery (environment during interaction).

CTU-CEG will test strength properties of samples made by cement paste and hydraulic conductivity, swelling pressure, retention curve, water absorption ability of bentonite.

*Table 1 – Combination of elements studied in defined loading procedures*

| | Ageing procedures | Bentonite 75 | Josef water | Portland cement | LpH cement | 95°C |
|---|---|---|---|---|---|---|
| new procedures: loading 9/18/27 months | suspension + OPC (LPH) 95°C | x | x | x | x | x |
| | suspension + OPC (LPH) | x | x | x | x | |
| | water + OPC (LPH?) 95°C | | x | x | x | x |
| | water + OPC (LPH?) | | x | x | x | |
| | suspension | x | x | | | |
| | OPC (LPH) | | | x | x | |
| aged samples 60 months | patrony | x | x | | | |
| | patrony + OPC | x | x | x | | |

**17. USFD/Claire Corkhill**

We will study 3 key systems, which comprise different cement compositions (see Q1.3), of different ages in contact with the following groundwater compositions:

- Granitic groundwater (e.g. Allard groundwater)
- Clay groundwater (representative of COx groundwater)
- High ionic strength, salt solution (representative of seawater)

Exact compositions will be agreed with ULough at the CEBAMA WP1 meeting.

Cements cured for 28 days, 1, 2 and 3 years will be investigated in these solutions. Cement curing will take place under the following conditions:

- Ambient, constant humidity
- Controlled $CO_2$ conditions
- Controlled anoxic conditions

## 18. VTT/Tapio Vehmas

VTT’s experimental work is focused on low-pH materials interacting with bentonite and the interface solutions.

Our system is low-pH concrete, low-pH bolt mortar and low-pH injection grout in finish nuclear waste repository, ONKALO environment. ONKALO environment is more precisely KBS-3V engineered barrier system, with bentonite backfill in crystalline rock deep ground repository. In the current project, the system is defined in the perspective of cementitious materials. In the studied environment, cementitious materials endure moist to wet environment during the operational life-time of the repository and then beyond during the closure phase. Factors affecting the cementitious materials are assumed to take place through aqueous medium. Composition of the contacting water can alter from glacier- and salty groundwater to bentonite saturated water. The bentonite type is MX80 for buffer and Friedland clay for tunnel backfill. The composition of bentonite saturated water in not yet defined, but is based on deep ONKALO water doped with bentonite. The reference concrete recipe is based on the DOPAS project (FP7) for concrete tunnel plugs, having both silica fume and fly ash as cementious supplementary materials.

The most important parameter that will be defined on the cementitious materials interface is the pH leachate and the evolution of pH leachate levels throughout the repository lifetime. Sufficiently low pH of the cementitious materials will ensure the stability of KBS-3V -engineered barrier system. Planned experimental methods either simulate various environments in ONKALO or provide more detailed understanding in the factors that define the pH-value in the cementitious materials interface. The main identified factors affecting the interface pH is the reaction degree of pozzolanic reaction, solution composition and leachate volume. However, these have internal dependencies and demand simultaneous testing.

**19. HZDR/Johannes Kulenkampff**

Our experiments will be conducted in close collaboration with SCK CEN on samples from the Boom clay – concrete interface, and with UNIBE on samples from the cement – Opalinus clay and aged cement – bentonite interfaces. The samples will be prepared by the partners. The system is characterized as columns or cylindrical samples with dimensions on the decimetre scale, which allows considering heterogeneity effects.

**20. LML/Jianfu Shao**

The system studied is a plane interface between Cox claystone and low pH concrete. The interface is composed by two half cylinders, the first one is the Cox claystone, and the second one is the low pH concrete which is directly fabricated in a cylinder mould. The parameter defining the aged interface is the hydration time of concrete. The estimated duration is 3 to 6 months. The diameter of the cylinder is 37mm and the height is about 40 mm.

## 21. UAM/Raúl Fernández Martín

UAM focuses the study on the geochemical reactivity, which characterizes the concrete/bentonite interface, mainly in a small scale (mm to nm) thickness regarding the chemical and mineralogical changes in both materials. Of course, the knowledge of the bulk scale processes (cm to m?) affecting concrete and bentonite has to be considered through the input of other participants. Time frame for geochemical reactions is also of prominent significance. This is why we consider small-scale new experiments involving similar materials to be studied in long-term experimental cells and in situ scenario. These generic objectives are going to be studied by means of the following systems:

**EXPERIMENTAL SYSTEMS AND OBJECTIVES**

| #System | Cement material **(C)** | Bentonite **(B)** | Aqueous phase | Bulk scale (thickness) ºC | Focus scale | Time scale | Objective |
|---|---|---|---|---|---|---|---|
| #1 Interface | CEM-**I** (OPC)/quartz mortar | FEBEX 0.5mm grain size | Grimsel type groundwater | ~5-10 mm **C/** ~5-10 mm **B** **25 ºC** | µm/nm | 1 week-several months | First mineral phases formation **where/when** |
| #2 Interface | CEM-**II** (AL type*) and LPH mortars | FEBEX 0.5mm grain size | Grimsel type groundwater | ~5-10 mm **C/** ~5-10 mm **B** **40ºC** | µm/nm | 1 week-several months | First mineral phases formation **where/when** |
| #3 Bulk interface cell experiment | CEM-**I**-SR-45 (OPC)/quartzite concrete | FEBEX 5 mm grain size | RAF clay formation saline water ** | 30 mm **C** 70 mm **B** **40 ºC** | mm/µm | 10 years | Mineral phases in a cell constrained experiment **Is there any relationship to #1 short-term?: ageing?** |
| #4 In Situ GTS shotcrete/ bentonite interface | CEM-**II** AL 32,5 R, with nanosilica, steel and polypropylene fibres | FEBEX 5 mm grain size | **AGED** Grimsel type groundwater | 1000 mm **C** / > 1000 mm **B** | cm/µm | 13 years | Mineral phases in unconstrained conditions **Is there any relationship to #2 short-term?: ageing?** |

*: to be defined similar to FEBEX in situ shotcrete plug

**: Concrete-bentonite system was expected to be significant in a clay formation context in Spain. It is a Na-Mg sulfate water and can be useful to consider sulfate and Mg interactions in the concrete system affected by the bentonite.

## 22. CSIS/Maria Cruz Alonso/Jose Luis Garcia Calvo

CSIC focuses its studies on the concrete part of different systems. CSIC evaluates the modifications promoted in concrete by its interaction with bentonite and/or granite and the corresponding groundwaters. In this sense, CSIC considers the next systems in CEBAMA project:

Long-term interactions (Existing aged concretes):

1. Febex shotcrete plug: aged concrete cores extracted from a full-scale experiment in the natural conditions of the Grimsel test site in contact with Febex bentonite re-saturated with granitic groundwaters. Operation time 13 years.
2. HB6 concrete from a small-scale laboratory cell experiment from CIEMAT: aged concrete in contact with FEBEX bentonite re-saturated with clayey groundwater. Operation time 10 years.

Early age interactions (New concretes):

1. High pH concrete (type of cement: CEM I): interaction with simulated granitic groundwater borehole and/or simulated Febex bentonite waters.
2. Low pH concrete in contact with simulated granitic groundwaters borehole and/or simulated Febex bentonite waters.

Different analysis will be carried out to evaluate the aging of the concrete samples. The analysis will be performed in the concretes zones in contact with the bentonite or the granite waters, and in parts less influenced by this interfacial zone. The tests will be focused on the determination of the hydrates alteration (C-S-H gel dissolution processes and chemical or mineralogical modifications as ettringite formation other salts interaction with cement paste (Cl, Mg, alkalies etc) and interaction with aggregate interfaces, affecting on porosity and permeability of the concrete matrix and components' interfaces.

**23. ANDRA/Xavier Bourbon**

« system » means the full association of all materials and conditions. In our case this means "*concrete/host rock interface in operating conditions*"

- Cementitious materials: low pH concrete, both formulations presented during our meeting in London: TL (ternary blend CEM I/SF/BFS) and TCV (ternary blend CEM I/SF/FA) (see my presentation slide#10);

- Host rock: Callovo-Oxfordian argillite;

- Conditions: the walls have been poured in a gallery, one side on the host rock (physically cleaned before pouring) and one side in atmospheric conditions in the gallery without any protection.

The "samples" prepared last year (2014), will be studied in different ways, with different goals. The main purpose is to validate our hydration model for low pH cement (at a representative scale…1m$^3$) to describe the evolution of physical properties. Some are monitored (to follow the evolution of the physical properties) and some others are not (to be sampled).

We plan to drill some samples from the BBP experiment (poured in our URL in bure, the 1$^{st}$ April 2014). The agenda has to be defined but we will probably drill the first samples next year (cylinder 8 cm diameter). The aim is to assess the "initial" behaviour of our two walls made with TL and TCV (mainly the setting period and the physical behaviour after setting, in operating conditions). The walls are 20 cm thick. We will drill the full thickness and part of the host rock, to keep "safe" the cement/clay interface and to have some chemically undisturbed clay too.

We will then have four kinds of samples:

1-gallery side: low pH TL and TCV carbonated (atm. cond.) and dried materials. In our URL the temperature is 21±1 °C, RH between 45 and 75% and the CO2 partial pressure the same as the atmosphere;

2-sound: "heart" of the wall: sound concretes as reference materials (probably the main part of the total length);

3-interface with Callovo-Oxfordian argilite. Both materials in contact: disturbed (?) cements and clay;

4-undisturbed clay host rock. The host rock has been "cleaned" before pouring the concretes; part of the argillite has been removed just before the experiment. The argillite was dry but not chemically disturbed (but still in the EDZ).

This will be available sample if any information is needed. Obviously we plan to analyze these samples. Some part could be saved for other experiments and shared with other labs.

For the moment (within the next few months), we do not plan to prepare any other samples in this context. But as we plan to start creeping experiments, we will have to prepare these two concretes (not before the middle of 2016).

**25. UNIBERN/Urs Mäder**

UNIBERN studies solely aged interfaces between Opalinus Clay and OPC/LPH.

System: interface region (altered material) and adjacent unaltered regions (mortar/paste/claystone), and pore water content

Aged interface: 3-years of interaction at in-situ conditions (CI Experiment, Mont Terri URL) produced complex, zoned mineral alteration on both sides of interface

Pore water content: preserved pore water at time of sampling and its evolution during ageing; subsequent changes during advective-diffusive column experiments

**26. IRSN/Alexandre Dauzeres**

As part of the current options retained by Andra in France for the high level waste cell, a plug of concrete should be emplaced to close definitively the cell in order to confine the bentonite swelling when the water will come back in the system. This plug (probably low-pH concrete) would then be in contact with bentonite at one side, its periphery being in contact with the host rock, a claystone (if the steel insert used for the deposit of the waste is removed). In these conditions, the cementitious material will be exposed to a long period of thermal transient (until 70 °C, based on modelling) and to progressive resaturation by the water coming from the host rock (claystone = CLS).

Literature shows clearly a lack of data concerning the evolution of cementitious materials in representative conditions of interfaces, in this range of temperature.

The CEMTEX (CEMent Temperature EXperiment) project launched in 2012 aims to study the physical-chemical evolution of cementitious materials (low-pH and OPC) exposed to these conditions (full saturation, 70 °C) in order to improve the numerical simulations.

Currently, we are studying interfaces between OPC or low pH hardened cement pastes with the Toarcian Claystone from the Tournemire URL at 70 °C (detailed in part 1.2).

The first sampling and characterization phase has shown that the contact between the cement paste and the CLS are in agreement with the foreseen objectives.