Co-funded by the

European Commission

HORIZON 2020

CEBAMA

➢ **(Contract Number: 662147)**

Deliverable n° D3.01

Detailed WP3 description of scientific work: compilation of partner descriptions with update of Appendix B.

Editors: Andrés Idiart (Amphos 21)

Date of issue of this report: 01.09.2015

Report number of pages: 5 + 66 appendices

Start date of project: 01/06/2015 Duration: 48 Months

Project co-funded by the European Commission under the Euratom Research and Training Programme on Nuclear Energy within the Horizon 2020 Framework Programme		
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	
CO	Confidential, only for partners of the CEBAMA project	

ABSTRACT:

This report gives details of the planned work for each of the participants of Work Package 3, related to Modelling and Interpretation of the Experiments to be carried out in Work Package 1. Any deviations from the initial Project Plan are noted.

Keywords: modelling, work description

RESPONSIBLE:

Andrés Idiart (Amphos 21)

MAIN TEXT:

This deliverable aims to report the planned work for each of the participants of Work Package 3 (WP3) related to Modelling and Interpretation of the Experiments to be carried out within Work Package 1 (WP1). The Working Plan is based on Appendix B of the Proposal of the CEBAMA (Cement-based materials, properties, evolution, barrier functions) project. A detailed description of the work to be carried out by each participant can be found in that appendix and will therefore not be repeated here.

Each participant of Work Package 3 has explicitly stated that there are no changes that have been identified or foreseen to the Working Plan. Therefore, the contents of the Working Plan described in Appendix B are fully applicable for the duration of the project.

As an Appendix to the present Deliverable, two slides per participant are included, that were presented during the Kick-Off meeting of the project, describing the work to be carried out by each organization participating in WP3.

The goal of Work Package 3 is to model and interpret the experimental results obtained within Work Package 1. Some of the partners within WP3 have already established which experiments of Work Package 1 will be modelled. Other partners will establish these experiments during a dedicated meeting between WP1 and WP3 partners next fall, to be held the 2nd November in London. The table presented below gathers this information (TO BE CONFIRMED in the table corresponds to the cases where specific WP1 experiments to be modelled need to be defined during the Fall Meeting).

Work Package 3 activities have started right after the Kick-Off of the CEBAMA project. However, experimental results from WP1 will not be available until much later. Therefore, each WP3 participant needs to use an existing set of experimental data, which is relevant to their objectives within the project, in order to test and verify their modelling approaches. In the table below, an additional column is included in which these experiments are specified, when available.

Table 1. *List of experiments that will be modelled by each partner.*

ID	Partner	Existing experiments	WP 1 experiments
1	**KIT**	Existing data from long-term leaching experiments of full scale cemented wastes (Kienzler et al., 2004)	Diffusion experiments (HTO, Be, Cl, I) in low pH-cement-clay systems (experiments performed at KIT-INE).
2	**A21**	Accelerated leaching experiments on cement paste and concrete samples performed at Chalmers University, Sweden (Babaahmadi, 2015)	TO BE CONFIRMED
3	**BRGM**	-	Modelling the streaming potential and complex conductivity experiments on low-pH C-S-H samples. These experiments will be performed within WP1 by Juelich (complex conductivity measurements, see Okay et al., 2014) and KIT (streaming potential experiments, Li et al., 2015).
7	**JUELICH**	TO BE CONFIRMED	TO BE CONFIRMED
9	**NRG**	Existing cement degradation experiments (Sarkar et al., 2010; Sarkar et al., 2012)	Diffusion-reaction experiments of high-pH concrete-Boom clay interaction performed by TU Delft
10	**RWMC**	(1) Mechanical properties such as porosity-compaction relation have already been obtained (Ishii,2013 and Kobayashi 2015) (2) Ca-XAFS observation of long-term immersed C-B coupled samples and/or artificial analogue specimen (Negishi, 2013) (3) Measurement of dissolution ratio of montmorillonite by using PSI technique (Sato,2013)	(1) Ca-XAFS observation of long-term immersed C-B coupled samples (2) Measurement of generation and growth ratio of secondary zeolitic minerals by using Phase Shift Interferometry (3) Measurement of the change of swelling pressure of montmorillonite during the change from Na-type into Ca-type
11	**SCK·CEN**	TO BE CONFIRMED	Modelling of the experiments involving the characterization of transport properties in high-pH - Boom clay interface, performed by SCK·CEN and BRGM within WP1

14	**UDC**	Heating and hydration experiments performed by CIEMAT and UAM on columns containing concrete and compacted FEBEX bentonite having durations ranging from 0.5 to 11 years (e.g. Cuevas et al., 2012)	Dismantling and characterization of the experiments of heating and hydration performed by CIEMAT and UAM within WP1 on columns containing concrete and compacted FEBEX bentonite
16	**CVUT**	Laboratory diffusion experiments of non-reacting species on samples of bentonite non-altered by any contact with cement (performed in both CTU and UJV (CZ))	1. Laboratory diffusion experiments of non-reacting species on samples of bentonite altered after the contact with cement (performed in UJV). 2. TO BE CONFIRMED
18	**VTT**	Published results on Low-pH concrete from EU_DOPAS -project (Leivo et al., 2014; Holt et al., 2014)	VTT experiments from WP1 on Low-pH paste samples with various Ca/Si -ratios. Leached low-pH samples in distilled and groundwater. Low-pH concrete, mortar and injection grout samples.
20	**LML**	Normal compression tests, direct shear tests and hydromechanical test performed on concrete fractures (Yang, 2011); Uniaxial and triaxial tests on low pH concrete conducted by Andra	Four groups of laboratory tests: 1) Mechanical test under room temperature (20 °C) and saturated condition (relative humidity = 100 %), 2) Hydromechanical tests under 20°C and 100%RH, 3) Influences of temperature (40, 60, and 80 °C) and relative humidity (50, 70, and 90%), 4) Effects of chemical degradation (carbonation of low pH concrete)
23	**ANDRA**	-	TO BE CONFIRMED
24	**PSI**	Clogging experiments in a sand-tank (Poonosamy et al., 2015), diffusion experiments across fresh and altered clay/bentonite interfaces (Shafizadeh, 2015), alteration of cement/clay interfaces in the framework of the Mont Terri CI experiment (Jenni et al., 2014)	TO BE CONFIRMED

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APPENDICES:

No. 1. Slides from partners. This appendix includes 2 slides per partner, presented during the Kick-Off meeting, describing the Working Plan.

No. 2. Description of the work of WP3 partners, as presented in the Proposal to the EC (Appendix B).

Work description for each partner

WP3: Modelling and Interpretation

Task: Reactive transport modeling *(18 PM)*

Objective:

Interpretation of diffusion experiments (HTO, Be, Cl, I) in low pH-cement-clay systems (KIT-INE).

- Transport parameters i.e. diffusion coefficient, porosity + experimental setup, from WP1.
- Uptake mechanisms of radionuclides, i.e. Be from WP2.

Description of the tools:

Codes to be used and Thermodynamic database:

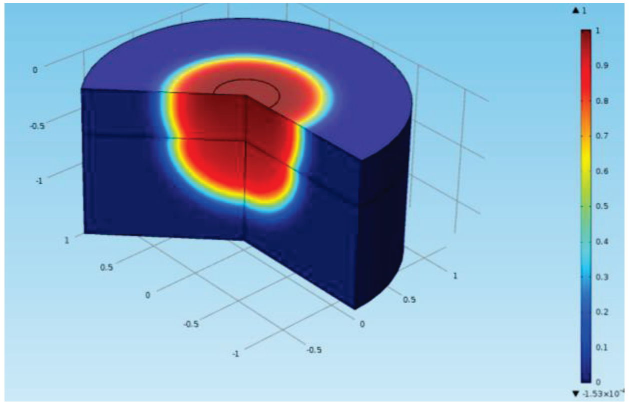
- PHREEQC 3.2
- Coupled Comsol v.5.0 – PHREEQC 3.2 (ICp)
- TDB: ThermoChimie v.9.0 + new data from Be obtained in WP2

WP3: Modelling and Interpretation

Task: Reactive transport modeling *(18 PM)*

Description of the work:

- Changes of the pore structure by dissolution of initial cement phases and the formation of alteration products by contact with clay porewater will be included in the model with feedback in the transport (diffusion).
- Mechanistic approach for the sorption of Be with the solid material will be also incorporated in the model.



Comsol v.5.0

Contribution of Amphos 21 to CEBAMA WP3

Brussels, 2nd July 2015

Overall objectives (38 PM)

1. Leading WP3 and the Modelling Task to integrate efforts within WP3 (micro-to-macro up-scaling)

2. Develop reactive transport models coupled with mechanics of concrete degradation with a description of microstructural processes in laboratory experiments. Experimental data to be used includes existing lab experiments and degradation experiments from WP1

3. Improve existing models and implement new couplings in the modelling frameworks to be used

4. Co-supervise PhD student with JUELICH: pore-scale reactive transport modelling using High Performance Computing (HPC)

5. Develop models to interpret the WP2 experiments on Mo sorption into cementitious matrices

Technical contribution to WP3

- Use of hydro-chemo-mechanical model to simulate cement and concrete degradation (calcium leaching) coupled to homogenization techniques for introducing microstructural information
- Modelling and interpretation of existing accelerated leaching lab and in-situ experiments (collaboration with SKB)
- Modelling and interpretation of experimental tests in WP1
- Co-supervision, together with JUELICH, of PhD student in pore-scale reactive transport modelling (Lattice Boltzmann + PhreeqC)
- Develop models to interpret WP2 experiments: batch Mo sorption with cement/pure solid samples and flow-through kinetic experiments during anionic substitution

AMPHOS 21
Scientific and Strategic Environmental Consulting

BRGM contribution to CEBAMA-WP3

Philippe Leroy

BRGM, Orléans, France

Geoscience for a sustainable Earth

brgm

BRGM contribution to CEBAMA-WP3 (8 PM)

Overall objectives:

The motivation of the work is to propose a monitoring tool to investigate, using geophysical non destructive methods, the mineralogy, porosity and pore network structure variations in concretes when the chemical composition of the pore water is modified. The electrical complex conductivity measurements performed in laboratory in WP1 will be inverted using a combined surface complexation and transport model to obtain the petrophysical properties of the samples.

Task 1:
Development of an electrostatic surface complexation model for the cement/water interface

- ✓ Development of a geochemical code in Phreeqc and Matlab for cement (in particular for low pH CSH) in contact with a saline water
- ✓ Development of a transport model to predict the zeta potential of cement particles
- ✓ Estimation of the parameters of the surface complexation and transport model using the results of Molecular Dynamics and Monte Carlo simulations, of surface charge and zeta potential measurements reported in the literature, and of some mineralogical and microstructural (D3E, BRGM), and electrokinetic experiments (streaming potential, streaming current, INE, KIT) performed in WP1

Task 2:
Development of an electrical low frequency complex conductivity model for cement

- ✓ Extension of the current electrochemical polarization models (membrane, grain polarization) to multivalents ions and mixed electrolytes in pore water
- ✓ Development of the geophysical code in Matlab and coupling of the surface complexation and complex conductivity model

Task 3:
Mineralogy, porosity, and pore network structure estimation

- ✓ Inversion procedure with a Matlab code. Estimation of the petrophysical parameters by decreasing the cost function between measured (WP1, in the mHz to kHz range, by IBG3, FZJ) and modelled complex conductivity data (BRGM)
- ✓ Comparison of the estimated petrophysical parameters to the measured ones in WP1

Nuclear Waste Management IEK-6
Institute of Energy and Climate Research

PhD studies on Pore-Scale Reactive Transport Modelling of Cementitious Materials using HPC

Collaboration between JUELICH and Amphos 21

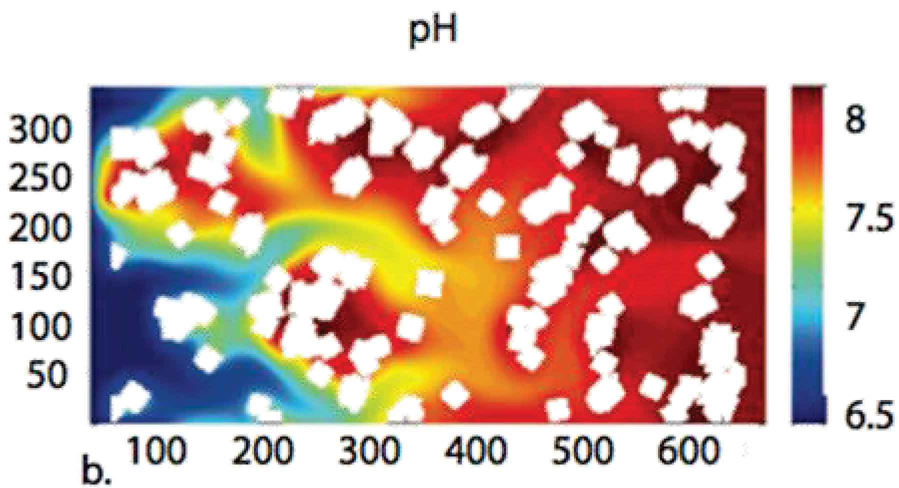
Motivation

1. Pore-scale physical and chemical heterogeneities exist in a range of length scales in cementitious systems
2. Pore structure evolution in concrete controls the continuum scale physical and chemical properties

Pore scale models are capable of capturing evolution of mechanical and transport properties of concrete through simulating various chemical degradation pathways in solid-liquid interface

Objective

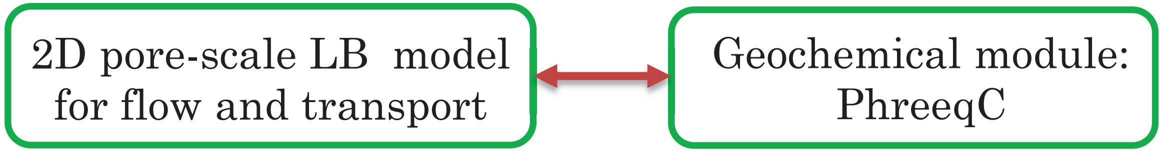
Develop and implement a reactive transport modelling tool at pore-scale using lattice-Boltzmann approach to study the impact of chemical degradation of cement paste on its physical properties (transport properties)



Pore-scale simulation of spatial pH distribution as a result of calcite dissolution at solid-liquid interface using lattice-Boltzmann method in a domain consisting of 350*700 nodes: Shafei et al. 2014

Methodology (39 PM)

1. Implement coupling between lattice-Boltzmann model (e.g. www.palabos.org) of flow (Navier-Stokes) and transport with chemical reactions (PhreeqC) for performing reactive transport simulations

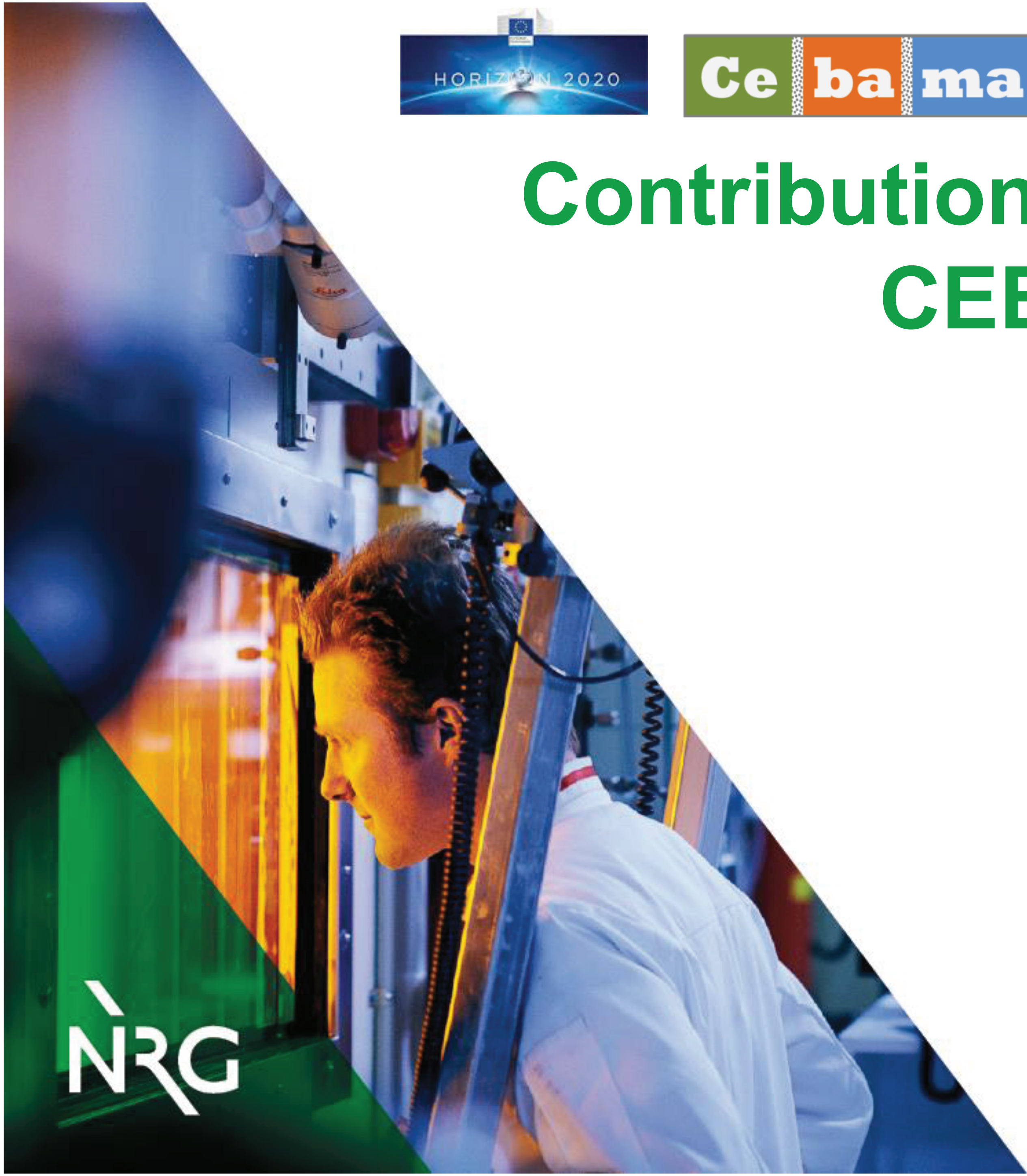


2. Simulation of concrete chemical degradation and radionuclide transport at the pore-scale using the High Performance Computing facilities available at JUELICH (JSC)

3. Upscaling to continuum-scale reactive transport: Darcy-scale constitutive relations for transport and geochemical properties to be used in iCP (interface COMSOL and PHREEQC)

Process modelling (connection to WP1/2)

Simulation of diffusion experiments for sorbing und non-sorbing radionuclides in cementitious systems using iCP for interpretation and evaluation of experiments performed within WP1



Contribution of NRG to CEBAMA WP3

Hans Meeussen

Brussels, 2 July 2015

Overall objectives (4 PM)

- To develop and test a state-of-the-art transport model that describes 1D reactive transport processes at the cement-clay interface.
- To design and interpret reactive transport experiments in a cement – clay system that will be carried out (within WP1) by TU-Delft.
- To make the experience in reactive transport modelling obtained within the US DOE funded Cement barriers project (www.cementbarriers.org) available to the CEBAMA project.

Main model characteristics

- **Reactive transport model description:**
  - Combines models for chemistry of concrete-clay, matrix components and radioactive elements within single system.
  - Includes state-of-the art chemical interactions in cement (CEMDATA07 thermodynamic database, www.empa.ch)
  - Combines chemical reaction, diffusion and radioactive decay processes.
  - Allows to take into account diffusion driven by electric potential
  - Incorporates feed-back between local chemical conditions and physical transport parameters (pore clogging, sulphate attack).
  - To be implemented within open source ORCHESTRA framework.

- **The reactive transport model will be used to design diffusion experiments with macro and trace elements.**
- **By comparing model predictions with experimental results the current level of process understanding will be evaluated.**
- **The model description of the concrete matrix will be based on the experience obtained in this area from the cement barriers partnership [1, 3].**
- **The model description for the clay matrix and adsorption properties will be based on the multisurface model described by Dijkstra [4].**

RWMC's contribution to CEBAMA-WP3

Hitoshi OWADA
Radioactive Waste Management Funding and Research Center

RWMC's contribution to CEBAMA-WP3 (6 PM)

1. RWMC is interested in

- How to couple between geochemical–mass transport code and FEM clay – hydraulic-mechanical code.
- Confirmation of the effectiveness and necessity of such H-M-C coupling calculation.
- How to model the mass-transport at the Cement-Clay interface

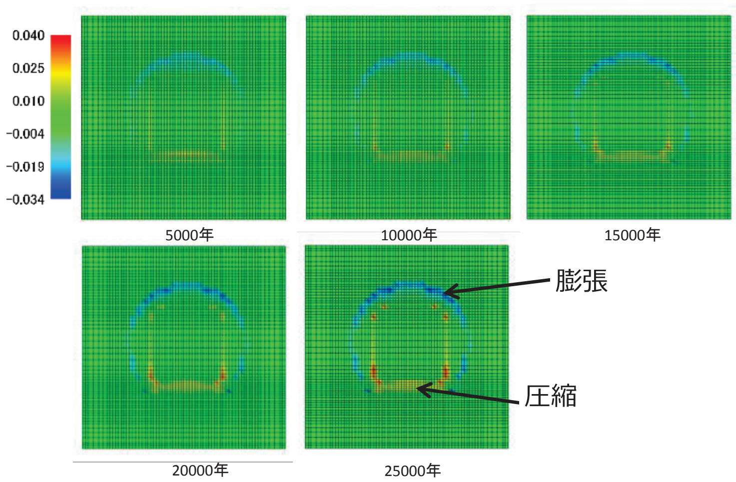
(In almost of the results of the H-C coupled calculation around the Cement-Clay interface, the altered zone are much wider than that of immersion experiment and natural analogues)

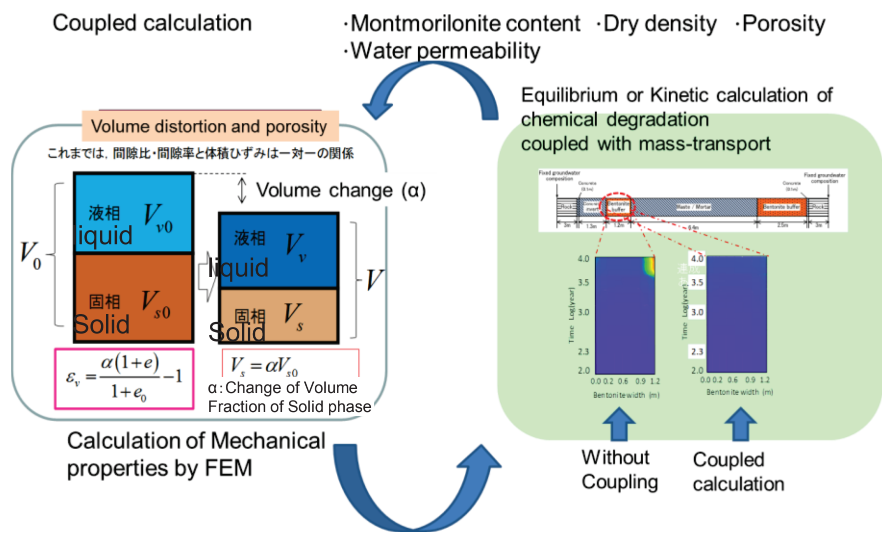
2. RWMC has already done (RWMC can share)

- Data of the expansion of altered zone around CC-bentonite interface of by 5 to 10 years immersed CC-bentonite coupled specimen.
- Modelling of mineralogy and chemical degradation of Fly ash and Slag mixed cements
- Modeling of the change of mechanical properties such as swelling pressure and hydraulic conductivity of bentonitic material due to chemical evolution
- Pseudo in-situ measurement of dissolution rate of montmorilonite and growth rate of some secondary minerals under compacted and alkaline condition

3. RWMC contributes

- Development of the Clay-Hydro coupling calculation FEM code "DACSAR" to couple with PHREEQC base mass-transport and chemical reaction coupling code.
- 2-D calculation with and without M-C coupling to confirm the effectiveness and necessity of such H-M-C coupling by using the concept and cross-section of Japanese TRU disposal.
- Reflect the fruits of WP1 year by year
- Reflect the fruits of Japanese CC-Clay project for TRU waste disposal





SCK•CENs contribution to WP3 of CEBAMA

Diederik Jacques

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SCK•CEN
STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

Objectives

Study interaction between high pH cementitous material and a clayey host rock (Boom Clay) focussing on chemical-phyiscal alterations within concrete:

- To validate if the modeling approach is able to predict macro-scale (continuum) transport properties (few hundreds of μm^3 to mm^3) using experimental information at the micro-scale (mainly spatial distribution of different concrete phase at µm scale).
- To apply a reactive coupled micro scale model for simulating the spatial-temporal evolution of geochemical changes
- To identify the most critical processes and parameters which affects the microstructural alterations and the macroscopic transport properties.

Work Program (8 PM)

- Task 1: Prediction of macroscopic variables from microstructural information
  - Starting from 3D geometries of concrete phases (input from FIB-SEM, WP1)
  - Derivation of 3D geometries fro 2D SEM images
  - Derivation of macroscopic properties using a lattice Boltzmann solver
- Task 2: Screening of important microstructural features affecting macroscopic transport properties
  - Based on virtual 3D geometries generated from statistical properties
- Task 3: Prediction of changes in properties resulting from geochemical alteration
  - Starting from 3D geometries of concrete phases (WP1)
  - Application of reactive-coupled lattice Boltzmann model

UDC contribution to the 1st CEBAMA meeting

Javier Samper

Civil Engineering School. Universidad de Coruña. Spain

UNIVERSIDADE DA CORUÑA

UDC contributions to CEBAMA (20 PM)

- **UDC** will improve reactive transport models and codes for modelling
  - The cement-clay interactions
  - The chemical, mechanical and hydrodynamic couplings with respect to the deformations,
  - The time evolution of porosity, permeability and diffusion coefficient,
  - The effects of low-crystal size mineral phases,
  - The retention of anionic species and carbonation processes.
- **UDC** will perform numerical models of heating and hydration laboratory experiments carried out by Ciemat on compacted bentonite (HB tests) to
  - Study the interactions of concrete and bentonite under repository conditions
  - Evaluate time-scale effects from concrete-bentonite test data measured after times ranging from 0.5 to 7 years

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UDC contributions to CEBAMA

- **UDC** will perform long-term reactive transport modeling of the concrete+bentonite barrier system of a high-level radioactive waste repository in a clay formation (performance assessment)
- **UDC** will evaluate model upscaling in time to contribute to the Modeling Task of WP3

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CTU contribution to the 1st CEBAMA meeting

Dušan Vopálka

Department of Nuclear Chemistry (DNC)
Czech Technical University in Prague (CTU), Czech Republic

WP3 – CTU-DNC-1 (6 PM)

Development of models

- improvement of methods of evaluation of diffusion experiments with non-sorbing species
- development of a geochemical model of interaction of cement and bentonite (PHREEQC environment)
- formulation of an advanced model of interaction of Sr and Ra with cementitious materials on the basis of data obtained in the experimental part of the study (WP2)
- development of methods that will contribute to an effective incorporation of the smart K_d concept into transport codes (for Ra retention and diffusion; GoldSim and/ or PHREEQC environments)

Cooperations

- CTU-DNC will closely cooperate with CTU-CEG and UJV Rez in evaluation of laboratory results and with SURAO by dissemination of results

WP3 – CTU-DNC-2

Use of developed models

- application of the improved model of diffusion experiment on the evaluation of diffusion experiments performed in WP1 by UJV
- use of the developed hydrogeochemical model of bentonite-cement interaction on the evaluation of data obtained by the experimental study performed (WP1 by UJV and CTU-CEG)
- sensitivity and probability study of developed interaction and transport models
- optimization of evaluation methods of results of different types of experiments performed in both our and cooperating laboratories (e.g. with help of UCODE connected with PHREEQC)

Cebama WP3
VTT contribution

Kick-off meeting, July 2 2015, Brussels

Tapio Vehmas, Aku Itälä

VTT objectives (7 PM)

- VTT objective is to develop well justified, credible and transparent modelling of simultaneous aging and leaching of low-pH concrete.
  - Comparison of various leaching scenarios to geochemical models.
    - Equilibrium compositions in various groundwater compositions.
    - Effect of polymerization degree.
    - Long –term behaviour of ternary mix designs.

Methods

- To reach these goals VTT will be using at least PhreeqC to model the batch experiments (if needed iCP, TOUGHREACT or Geochemist's workbench might be used).
- Laboratory paste samples are compared to Geochemical batch models in various groundwater compositions.
- Laboratory paste samples are accelerated in order to reach thermodynamic equilibrium.
- Leaching is also performed for equilibrium.
- Modelling is performed with PHREEQC.
- Kinetics are accounted by pozzolanic reaction degree and leaching volume.

LML contribution to CEBAMA-WP3

Jianfu SHAO

LML, University of Lille

LML contribution to CEBAMA-WP3 (30 PM)

Overall objectives:

Develop multi-scale modeling for hydromechanical behaviors of concrete and concrete/argillite interface by considering effects of mineralogical compositions, temperature, relative humidity and microstructure evolution due to carbonation.

Task 1: Development of a micro-macro model for concrete using nonlinear homogenization techniques

- ✓ Three scales considered: microscopic scale (CSH grains and pores), mesoscopic scale (small inclusions as sand), macroscopic scale (large size aggregates)
- ✓ Establish a close-form or semi close-form macroscopic failure criterion explicitly taking into account the effects of pores, small inclusions and large aggregates
- ✓ Including effects of temperature, relative humidity (or saturation) and chemical degradation (leaching and carbonation)
- ✓ Model's parameters identified from existing data on low pH concrete investigated by Andra and simulation of laboratory tests

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Task 2: Development of a hydromechanical model for concrete/Cox clay interface

- ✓ Taking into account of microstructural effects (asperity, tortuosity, chemical reactions) using a micro-macro approach
- ✓ Mechanical behavior of interface under both normal and shear stress
- ✓ Evolution of permeability
- ✓ Parameters identified from laboratory tests performed in WP1
- ✓ Numerical simulations of laboratory tests

Task 3: Numerical analysis of typical structures including concrete, Cox clay and interface

- ✓ Implementation of the proposed HM model into a computer code using an extended finite element method
- ✓ Numerical analysis of a simplified structure such as a representative section of gallery selling structure including expansive clay, concrete support massif, concrete lining and Cox argillite
- ✓ Study the key role of the interface in the long term stability of sealing structures in storage boreholes

Cebama WP3 - Interpretation and modelling

Andra's proposal

July 2nd, 2015

Context

Main objective: improving the description of the phenomenological evolution of an ILLW cell and a seal in order to support the PA analysis

- Through a process combining both
  - a change of scale (from experimental scale to host rock scale) and
  - a change in representation (from a phenomenological point of view to a performance assessment analysis)
- Set up a three-step approach divided into
  - Upscaling the data/model acquired at the laboratory scale to time and space scales representative of a deep underground repository
    - hydraulics and mechanics behaviour of concrete (WP1),
    - radionuclides migration (WP2)
  - Integration of the upscaled data/model in HMC modelling of ILLW cell and seal evolution (full chemical approach)
  - Large scale modelling through a PA approach (simplified chemistry)

Three-step approach (8 PM)

Processes scaling

- Based on experimental data (WP1 and WP2), the main objective is to extrapolate models for large scale components taking into account uncertainties (epistemic and stochastic variability of the acquired data).
- Focus will be made on the deterministic model of radionuclide uptake and redox evolution of interstitial water and the consequences on radionuclide retardation

Phenomenological modelling

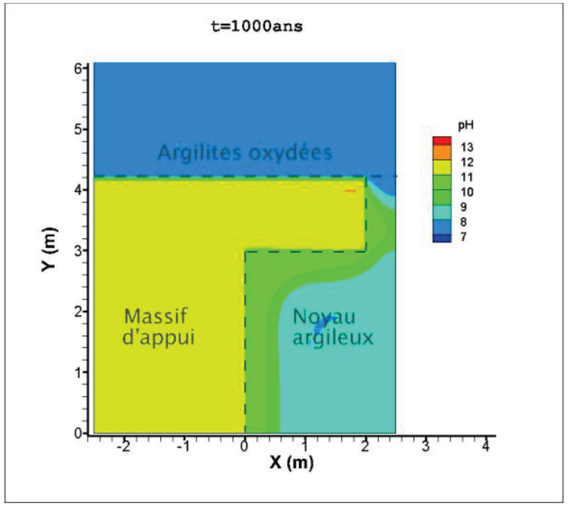
- H(M)CR modelling of a tunnel seal and a ILLW disposal tunnel over large time and space scales:
  - ILLW: focus on the chemical evolution of cementitious components in consistency with the related up-take processes of radionuclides and the hydraulic transient period
  - Seals: focus on interface between low pH concrete and clay materials (host rock, bentonite)

Performance assessment modelling

- Large scale modelling with a simplified representation of the HMCR processes

Time Schedule

- First and second years: Involvement in end users group meeting
- Third and fourth years: Modelling and reporting



Example of a reactive transport simulation at a seal/concrete interface in the French RWD concept

Paul Scherrer Institut

Georg Kosakowski

PSI contribution to CEBAMA WP3:
Modelling transport across reactive interfaces

PSI contribution to CEBAMA WP3 (40 PM)

Overall Objectives:

- Development of improved 2D and 3D conceptual and numerical models for couplings between chemical and transport processes taking into account the influence of charged mineral surfaces.
- Testing and comparison of different model approaches by analyzing experiments on interactions of concrete with clays or other materials.
- Application on long term geochemical evolution of repository near field.

Description of work to be performed:

- Implementation of electrically coupled multi-species transport in multi-porous media in open-source reactive transport codes OpenGeoSys-GEM and FENICS – GEM/Reaktoro.

PSI contribution to CEBAMA WP3

Description of work to be performed:

- Implementation of models that capture effects of charged surfaces in open source LMA and GEM chemical solvers (GEMS3K and Reaktoro).
- Analysis of experimental data (e.g. ongoing PhD projects at PSI on porosity evolution at material interfaces, Cement-Clay Interaction Project at the Mont Terri Underground Rock Laboratory, experiments at Uni Bern in WP1, and from other experimental projects in WP1).
- Application with 2D/3D cavern and/or repository scale models on long term geochemical evolution of a heterogeneous repository near field.

WP 3			KIT

Overall Objectives:

KIT quantifies the precipitation/dissolution processes in pores of cementitious materials, changes in pore size distributions and changes in permeability performs by experiments and modelling on interface reactions.

Coupled reactive transport modelling is applied for interpretation of the experimental results. Treatment of changes in transport parameters such as diffusion constants / permeability / etc. on the basis of dissolution / precipitation processes are included in the models. The models will be verified by comparisons with experimental data.

Objectives for different specific steps/periods (if applicable):

At the beginning of the project, no data will be available from WP1/WP2. For this reason, the conceptual model including reactions and data will be tested by application to existing data from other projects (i.e FP7-PEBS, -DOPA). Detailed objectives are:

- Calibration and testing of reactive transport models by comparison to already existing data from other projects: i.e long-term leaching experiments of full scale cemented wastes.
- Interpretation of diffusion processes leading to changes in diffusion properties (performed within WP1) and deduction of the reaction schema and data base for chemical/transport processes.
- Application of model and data base to real systems with respect to space and time (up-scaling activity).

Main expected outcome:

- Reactive transport model for diffusion of tracers (HTO, Be, Cl, I) in the interface of low pH cement - clay systems, using the information obtained in WP1 (transport parameters, kinetics) and WP2 (thermodynamic information on Be). Comparison between modeling and experiments.

Description of work to be performed:

- Reactive transport modeling approximations will be used to model diffusion over the interface of low pH cement - clay systems, using different codes:
    - PHREEQC
    - Coupled Comsol-PHREEQC (iCP, interface COMSOL-PHREEQC)
- and different thermodynamic databases: ThermoChimie /CemData. When needed, databases will be updated incorporating new data and reaction kinetics obtained in the project.

Cooperations

- -Beneficiaries: AMPHOS, BRGM, PSI, UJV (participation in the generic modelling task)
- -External participants: PEPS, DOPAS partners

Plans for dissemination:

- All results will be published in peer-reviewed journals, conference proceedings, etc.

WP3			Amphos 21

Overall Objectives:

Amphos 21 efforts on the modelling of the most relevant physico-chemical processes leading to concrete degradation and loss of barrier performance (in each deposition environment) will be focused on:

- Leading the modelling work package contributions from the different partners
- Description of a Modelling Task to integrate efforts within WP3, focusing on micro-to-macro up-scaling techniques
- Develop numerical thermo-hydro-mechanical-chemical (THMC) models with a level of interpretation that covers the gap between a description of microstructural processes and the laboratory experiments. These models will cover the degradation processes studied experimentally in WP1 and WP2, including (some of) the interfaces of concrete and bentonite/clays/granite. The experimental datasets to be used will also include existing field experiments and industrial analogues (see collaboration with SKB in description of work below) that concentrate on similar processes to those studied in WP1. Based on these models it is expected to obtain valuable results that can ultimately be used to derive simplified models for the behaviour at the macroscopic scale.
- Co-supervise a PhD student together with JUELICH on reactive transport modelling issues related to High Performance Computing (HPC) at the micro- and/or meso-scale

Objectives for different specific steps/periods (if applicable):

- Step 1: Contribute to the establishment of the details of the experimental program: to optimize the experimental program by establishing a close interaction between experimentalists and modellers
- Step 2: Selection and adaption of suitable software for development of a (thermo)-hydro-mechanical-chemical (THMC) model, also compatible with HPC environment at JUELICH
- Step 3: Quantify and interpret, with the use of coupled THMC numerical models at the micro- and meso-scale, the results generated within the framework of WP1 (Experimental) and also results from existing field experiments (see description of work below).
- Step 4: Up-scale the results to the macroscopic scale, where concrete can be treated as homogeneous.

Main expected outcome:

The expected outcome is a multi-scale *modelling framework* that incorporates the latest laboratory observations in micro-/meso-scale models that can be used to gain new insight in long-term degradation of cementitious systems under repository conditions

Description of work to be performed:

- *Step 1.* Active involvement from the inception of the experimental program in order to (a) contribute with a modeller perspective, and (b) account for potential modelling needs, parameterization and knowledge gaps, and model limitations.
- *Step 2.* Different modelling options and available software need to be critically evaluated in order to select the best approach for the interpretation of the experiments at the micro- and meso-scale, and to demonstrate the feasibility to implement such models in a HPC environment.

WP3			Amphos 21

- *Step 3.* The experiments and models should be capable of quantifying coupled physico-chemical processes (flow, transport, and mechanical properties, changes in microstructure, chemical reactions, etc.). The work will be focused on improving existing models and implementing new couplings in the modelling framework to be used. The experiments to be modelled include those performed within WP1 and WP2 (Amphos 21 experiments on retention of Mo) and existing experimental data relevant to the target physico-chemical processes in CEBAMA. In particular, results of concrete samples from SFR repository for LILW in Sweden (SKB) will be modelled. Experimental results from the project Concrete and Clay from Äspö URL (SKB) are also potentially of relevance to CEBAMA and could be the subject of further modelling efforts.
- *Step 4.* As a final step, the results from sophisticated micro-/meso-scale models developed within the project will be up-scaled to the macroscopic scale, where concrete is regarded as a homogeneous material. Simplifying assumptions will be considered so that the most relevant features can be correctly captured in an up-scaled model, e.g. using homogenization schemes.

Cooperation:

- Close cooperation with the following CEBAMA partners is foreseen:
- (1) KIT-INE: Amphos 21 will prioritize the modelling of experiments performed by KIT-INE within WP1 regarding clogging and diffusion-related issues in cementitious systems
- (2) JUELICH: Amphos 21 will co-supervise a PhD student in cooperation with JUELICH, which work will be focused on modelling coupled reactive transport processes in cementitious systems at micro-/meso-scale using HPC resources from Jülich
- CEBAMA external partners: cooperation with SKB is planned on the modelling of experimental data

Added value of Cebama:

This WP represents an opportunity to integrate the study of long-term concrete degradation processes with the out coming results from laboratory experiments. The development of numerical models at micro-/meso-scale will increase the overall understanding of the main processes governing the interfaces behaviour. It is the first European project that intends to study long-term cementitious materials under repository conditions involving experimentalists and modellers with different and complementary backgrounds. At the same time, the use of High Performance Computing resources will allow to go a step further in the use of advance models in this framework.

Planned training activities (if any):

- Participation in the annual project workshops,
- Participation in training events /schools in association with the annual project workshops

Plans for dissemination:

- Publish results at international conferences (NUWCEM, CONMOD, Euro-C, MRS, Migration, IGD-TP, etc.)
- Publish in peer reviewed journals and online resources (IGD-TP website, CEBAMA website)

WP 3			BRGM

Overall Objectives:

BRGM will model the physico-chemical interactions between cement materials and pore water, and their consequence on the evolution of the pore structure, size distribution, and resulting reactive transport properties of these materials. The mineralogical and microstructural experiments performed in WP1 will be used to calibrate the parameters of the electrostatic surface complexation model. This model will be developed at the microscopic scale and it will be combined with a complex electrical conductivity model established at the microscopic and macroscopic (laboratory) scale. This model will predict the evolution of the porosity, and of the grain shape and size distribution as a function of the pore water chemistry using low frequency complex conductivity measurements. This work will be conducted in close collaboration with IBG team of Forschungszentrum Jülich, which will provide complex conductivity measurements.

Objectives for different specific steps/periods (if applicable):

- The objective of BRGM team is to describe accurately the physico-chemical reactions occurring at the mineral/water interface, in the bulk pore water, and their influence on the reactive transport properties of concrete through the description of the evolution of the porosity and pore network structure. It is a multi-scale modeling approach with the coupling of an electrostatic surface complexation model (microscopic scale) with a complex conductivity model (microscopic and laboratory scale). The complex conductivity model is influenced by conduction and polarization current densities occurring at the surface of the charged particles and in the bulk pore water. The parameters of the two models will be calibrated using mineralogical, microstructural, and complex conductivity experiments performed in WP1. The development of the complex conductivity or spectral induced polarization (SIP) model will be done mainly during years 2 to 4, because complex conductivity data will be recorded during years 1 and 2 (there are no complex conductivity measurements available for low pH CSH and cement concrete in the literature). Nevertheless, the electrostatic surface complexation model of low pH CSH and concretes can be developed earlier using potentiometric titration and electrophoretic mobility measurements reported in the literature (during year 1 and year 2). In addition, the theory of the SIP model can also be developed during year 1 and year 2.
- The first step (work to be done during year 1 and year 2) is to develop and calibrate the parameters of the surface complexation model describing the concrete/pore water interface. The measured surface charge densities and electrophoretic mobilities (zeta potentials) of well characterized (mineralogical composition, microstructure) low pH CSH and concretes will be described by the surface complexation model. The electrochemical properties of low pH CSH and concretes will be firstly modeled using potentiometric titration and electrophoretic mobilities measurements reported in the literature. The surface complexation model will also use information gained from molecular dynamics simulations studies reported in the literature. The surface complexation model will calculate the surface site densities of adsorbed ions in the electrical double layer around the charged particles.
- The second step (work to be done during years 2 to 4) is to use the computed surface site densities of adsorbed ions as input parameters of the complex conductivity model. This model will also use the results of the mineralogical and microstructural experiments performed in WP1 to describe the evolution of the pore structure and size distribution (PSD) associated with the clogging or opening of pores (due to change of the concrete mineralogy because of Ca-leaching and carbonation). The predictions of the complex conductivity model will be compared to the low frequency complex conductivity

WP 3			BRGM

measurements, in order to non-intrusively estimate the evolution of the porosity, grain shape and size distribution (these experimental data will be recorded during year 1 and year 2).

Main expected outcome:

- A robust and accurate electrostatic surface complexation model to describe the concrete/pore water interface.
- Low frequency complex conductivity measurements on concrete materials.
- A mechanistic complex conductivity model to non-intrusively estimate the change of the pore structure and size distribution as a function of the evolution of the pore water chemistry.

Description of the work to be performed:

The evolution of the mineralogy and microstructure of the concrete as a function of the chemical composition of the pore water has strong implication on the reactive transport properties of these materials. The change of the porosity, pore structure and size distribution (clogging or opening of pores associated with Ca-leaching and carbonation) will be described using our multi-scale modeling approach (mineral/water interface, concrete sample at the laboratory).

We propose the following methodology:

- Development of a triple layer model for the low pH concrete/water interface.
  - A bibliographic synthesis will be firstly realized to choose the model. The members of the BRGM team have already published many papers on the modeling of the surface electrochemical properties of clays and silica immersed in pore waters of different chemical compositions (for instance [1-3]). A special attention will be dedicated for the description of the sorption of Ca^{2+} ions in the Stern layer.
  - The surface complexation model will be applied to CSH (Calcium Silicate Hydrates) with a low Ca/Si stoichiometric ratio (for instance, 0.66), and immersed in a salt solution (NaCl, NaI, KCl, LiCl, CsCl). The mineralogical and microstructural analyses performed in WP1 will help to develop a realistic surface complexation model. The calculations will be performed using the geochemical software Phreeqc coupled with the parameter estimation software Pest. In addition, the Matlab code developed by researchers of the team (to describe the surface electrochemical properties of clays and silica) will also be developed and used because it can be easily modified and may give better parameter estimations (compared to the use of Phreeqc with Pest). In addition, the complex conductivity model is written in Matlab, and it may be better to have only one code simulating simultaneously the electrochemical properties of the mineral/water interface and the complex conductivity of the sample. The predicted surface charge densities and electrophoretic mobilities will be compared to the measured ones.
- Development of a mechanistic complex conductivity model for water saturated low pH concretes.
  - The Stern layer electrochemical polarization model developed by P. Leroy (BRGM team, [4-5]) will be firstly extended to multivalent and multi-ionic species by extending the analytical equations to the different species. The Matlab code will be therefore implemented. In addition, the finite element software Comsol Multiphysics will also be used to solve numerically the ion transport equations (associated with ions

WP 3			BRGM

electromigration and diffusion) around the Stern and diffuse layers of a charged particle with a given shape and size. The Comsol Multiphysics code will be coupled with the Matlab code in order to compute the complex electrical conductivity of the sample made of concrete particles and bulk pore water.

- Coupling of the surface complexation and complex conductivity models.
    - The two models will be directly combined through the calculation of the surface charge densities, zeta potentials, and surface site densities of adsorbed ions in the Stern and diffuse layers. The predictions of the complex conductivity model will be compared to the low frequency [0.1 mHz- 45 kHz] complex conductivity measurements performed on low pH concretes in contact with pore waters of known chemical composition. Evolution of the porosity, grain shape, and size distribution will be estimated by matching the predicted to the measured data. It will also be compared with microstructural measurements performed in WP1.

Low frequency complex conductivity measurements will be carried out by the IBG team of Forschungszentrum Jülich. Comparison of the predictions of the complex conductivity model with the experimental data will be performed in close collaboration with the IBG research team.

[1] Tournassat C., Chapron Y., Leroy P., Bizi M., Boulahya F. (2009). Comparison of molecular dynamics simulations with triple layer and modified Gouy-Chapman models in a 0.1 M NaCl-montmorillonite system. Journal of Colloid and Interface Science, 339, 2, 533-541.

[2] Leroy P., Devau N., Revil A., Bizi M. (2013). Influence of surface conductivity on the apparent zeta potential of amorphous silica nanoparticles. Journal of Colloid and Interface Science, 410, 81-93.

[3] Tournassat C., Grangeon S., Leroy P., Giffaut E. (2013). Modeling specific pH dependent sorption of divalent metals on montmorillonite surfaces. A review of pitfalls, recent achievements and current challenges. American Journal of Science, 313, 395-451.

[4] Leroy P., Revil A. (2009). Spectral induced polarization of clays and clay-rocks. Journal of Geophysical Research, 114, B10202, doi:10.1029/2008JB006114.

[5] Okay G., Leroy P., Ghorbani A., Cosenza P., Camerlynck C., Cabrera J., Florsch N., Revil A. (2014). Spectral induced polarization of clay-sand mixtures. Experiments and modelling, submitted to Geophysics.

Cooperations:

- *IBG team of Forschungszentrum Jülich will provide very accurate low frequency complex conductivity measurements of low pH concretes in contact with pore waters of known chemical composition.*
- *Cebama partners.*

Added value of Cebama: multi-disciplinary cooperation on international level between specialized research teams will result in an efficient increase in understanding of phenomena playing on clay/concrete interface. In our modeling approach, we will combine the expertise of IBG team of Forschungszentrum Jülich regarding the recording of low frequency complex conductivity measurements with the expertise of BRGM regarding the mineralogical and textural analyses, and

WP 3			BRGM
the geochemical and complex conductivity modeling.			
Plans for dissemination: • Joint papers, posters/presentations at conferences, open research reports …			

WP 3			JUELICH
Overall Objectives: Application of high-performance computing (HPC) to reactive transport modelling of multi-scale processes in cementitious repository environments.			
Objectives for different specific steps/periods (if applicable): - Selection of appropriate simulation software and modelling tools (e.g. COMSOL coupled to PhreeqC via the iCP interface) and adaptation to the HPC environment - Application of the simulation software to quantify and interpret experimental data derived from Cebama WP 1/2 as well as existing data from laboratory experiments and natural/technical analogues (e.g. data used in the Ecoclay II studies, Tournemire analogue, and/or data from the long-term cement studies (LCS) project) - Up-scaling to the macroscopic level and evaluation of application to safety assessments			
Main expected outcome: - Improvement of modelling approaches and tools for the simulation of multi-scale processes and transport phenomena affecting the RN transport in cementitious systems and through interfaces with the host rock to underpin safety assessments.			
Description of work to be performed: - Reactive transport modelling of experiments performed within WP 1 as well as existing experimental data and analogue studies for analyses, interpretation and quantification of transport processes. - Evaluation of options how to represent/include the uptake mechanisms of radionuclides elucidated in WP 2 within the modelling tools. - Up-scaling and simulation of multi-scale coupled reactive transport processes and modelling of RN transport through (aged) concrete materials and concrete/host rock interfaces in the repository environment.			
Cooperations - JUELICH collaborates with Amphos 21 – based on a shared PhD student – regarding the application of HPC to the modelling of coupled reactive transport processes. Exchange with the Uni Sheffield will provide additional information on transport properties of cementitious materials. Cooperation with BRGM regarding SIP measurements of transport properties of cements. - External cooperation on reactive transport, e.g. with ANDRA, SKB, and POSIVA (as well as Amphos 21) within the iMaGe initiative as well as with industry partners such as Brenk Systemplanung, Aachen, Germany, in common research activities.			
Plans for dissemination: - Publications in peer reviewed journals - Presentations at international conferences (e.g. Goldschmidt, Migration, IGD-TP)			

9 / NRG

WP 3			NRG

Overall Objectives:

- To develop and test a state-of-the-art numerical model describing 1D reactive transport processes at the cement-clay interface in collaboration with TU Delft (see proposal TU Delft).
- To design and interpret reactive transport experiments that will be carried out within WP1 by TU-Delft (see proposal TU Delft).
- To make the experience in reactive transport modelling obtained within the US DOE funded Cement barriers project (www.cementbarriers.org) available to the Cebama project.

Objectives for different specific steps/periods (if applicable):

- *Task 3.1: Setting up initial model to define boundary conditions for experimental work carried out by TU-Delft*
- *Task 3.2: Evaluate and update model to interpret experimental results obtained by experiments TU-Delft*
- *Task 3.3: Use developed cement-clay interface model to evaluate significance of processes over longer time scales*

Main expected outcome:

- *Increased understanding of processes occurring at the cement – clay interface*
- *Experimental evaluation of the level of understanding of processes at interfaces*
- *Open source numerical reactive transport model, based on ORCHESTRA framework*
- *Contribution to training of students (at TU-Delft)*
- *Scientific papers on this topic*
- *PhD thesis*

Description of work to be performed:

- Setup a reactive transport model for the experimental system present at TU Delft describing matrix components in cement and clay. Initially this model will be used to design the exact experimental conditions, and at a later stage to interpret the experimental results. The initial version of the model will make use of previously available data on thermodynamic modelling of cement systems available from literature and from our earlier work.
- Main characteristics of model description:
    a. Combines models for chemistry of concrete-clay, matrix components and radioactive elements within single system.
    b. Includes state-of-the art chemical interactions in cement (CEMDATA07 thermodynamic database, www.empa.ch)
    c. Combines chemical reaction, diffusion and radioactive decay processes.

WP 3			NRG

d. *Allows to take into account diffusion driven by electric potential*

e. *Incorporates feed-back between local chemical conditions and physical transport parameters (pore clogging, sulphate attack)[3,5].*

f. *To be implemented within open source ORCHESTRA framework [2]*

- *The reactive transport model will be used to design diffusion experiments with macro and trace elements.*
- *By comparing model predictions with experimental results the current level of process understanding will be evaluated.*
- *The model description of the concrete matrix will be based on the experience obtained in this area from the cement barriers partnership [1, 3].*
- *The model description for the clay matrix and adsorption properties will be based on the multisurface model described by Dijkstra [4].*

References:

1. *Cement barriers project (www.cementbarriers.org)*
2. *Meeussen, J.C.L., 2003, ORCHESTRA: An object-oriented framework for implementing chemical equilibrium models, Environmental science & technology 2003, 37 (6), 1175-1182.*
3. *Sarkar, S., Mahadevan, JCL Meeussen, H Van der Sloot, DS Kosson, Numerical simulation of cementitious materials degradation under external sulfate attack, Cement and Concrete Composites, 2010, 32 (3), 241-252.*
4. *Dijkstra, J.J, JCL Meeussen, RNJ Comans, Evaluation of a generic multisurface sorption model for inorganic soil contaminants, Environmental science & technology, 2009, 43 (16), 6196-6201.*
5. *Benchmark for reactive transport codes in the context of complex cement-clay interactions, Nicolas C.M. Marty, Philippe Blanc, Olivier Bildstein, Francis Claret, Benoit Cochepin, Su Danyang, Eric C. Gaucher, Diederik Jacques, Jean-Eric Lartigue, K. Ulrich Mayer, Johannes C.L Meeussen, Isabelle Munier, Ingmar Pointeau, Liu Sanheng and Carl Steefel, 2014, Submitted to Computational Geoscience*

Cooperations:

- *Within Cebama close collaboration with TU-Delft (WP1)*
- *External collaboration within this subject area with Vanderbilt University (existing collaboration on modelling long term behavior and radionuclide migration through cement barriers.)*

Added value of Cebama:

- *Provide means for European integration of knowledge on reactive transport modelling in clay-cement systems*
- *Share experience of NRG in work on Cement Barriers (www.cementbarriers.org) with European partners*
- *Make state of the art knowledge on cement processes available to PA scale modelling as carried out within the Dutch national research programme (OPERA) on geological disposal of radioactive waste.*

WP 3			NRG
Planned training activities (if any): • *Training students in using reactive transport model as part of TU-Delft curriculum* • *ORCHESTRA geochemical modelling and transport course*			
Plans for dissemination: • *Scientific papers, on reactive transport behavior of nuclides and* • *PhD thesis* • *Open source numerical reactive transport model, based on ORCHESTRA framework* • *Contribute to training of students (TU Delft)*			

WP 3			RWMC

Overall Objectives:

1. To develop the method of H-M-C coupled calculation for mainly bentonitic material. The main point of RWMC's work is the reciprocal influences between change of (1) mechanical and hydraulic properties due to chemical evolution and (2) changes of the chemical alteration due to the change of hydraulic properties. Those changes due to Cement-Bentonite interaction.

Reference bentonitic material is Kunigel V1 and cementitious materials are OPC and Low alkalinity cement in Japan, but other cementitious materials, i.e. flyash mixed cement , are also in consideration.

Objectives for different specific steps/periods (if applicable):

The results of WP1 will be used as input for this work. The outcome of this work could be useful for evaluating the long term performance of barrier system including cement-clay systems.

RWMC has already done the modeling of the alteration of various cementitious materials, i.e. OPC and mixed cement, and also has been carrying out many experiments of cement-bentonite coupling. RWMC has also natural analogue project of pH~11 alkaline alteration of bentonite formation in Philippines. Those results will be able to share in CEBAMA project, and RWMC will reflect the fruits of those experiments and studies, including the dissolution rate of montmorillonite under compacted condition, as the input for the long term mineralogical changes. The changes of mechanical and hydrological properties have been modeled as a function of "effective montmorillonite density" in FY2000 report and 2nd progress report for TRU waste disposal in Japan. RWMC uses that relationship for the M-C coupled calculation.

Main expected outcome:

- Method of the coupling between geochemical –mass transport code and FEM mechanical calculation code including developed FEM code "DACSAR" which can calculate the mechanical properties with the change of mineralogical change, e.g. mass and density change, of the solid phase in each element due to mass transport and chemical degradation.

Description of work to be performed:

1. Development of FEM code "DACSAR" for the calculation of the change of mechanical properties and density of bentonitic clay and cementitious material. 2D geochemical-mass transport calculation code will also be developed. With the fruits of WP1 and other experiments of RWMC will be reflected for that development.

Cooperations:

- RWMC has already done the modeling of the change of water permeability of bentonitic material due to chemical alteration. RWMC will be able to share that.

Added value of Cebama:

Planned training activities (if any):

DACSAR had been developed by Tokyo institute of technology and Kobe university*. Modification of DACSAR regarding M-C coupling calculation is a collaborative work between Kobe Univ. (Prof. Iizuka) and RWMC.

A part of geochemical calculations will be performed by Hokkaido Univ. (Prof. Sato) under the contract with RWMC.

WP 3			RWMC
* Iizuka and Ohta : A determination procedure of input parameters in elasto-viscoplastic finite element analysis, Soils and Foundations, Vol.27, No.3, pp.71-87,(1987).			
Plans for dissemination: • Results of RWMC's project must be published as annual report to METI, and will be submitted to scientific journals.			

WP 3			SCK•CEN

Overall Objectives:
The overall objectives are (i) to validate the prediction of macroscopic variables using experimental information on the microstructure, and (ii) to apply a coupled reactive transport model for simulating the relations between chemical degradation, microstructural changes and macroscopic transport properties at the micro-scale, i.e. explicitly taken into account the spatial arrangement of different phases (e.g. liquid, portlandite, calcium-silicate-hydrates (C-S-H), clinkers), at the interface between concrete and the host rock. The reactive transport model, within a lattice-Boltzmann framework, uses micro scale information from WP1 (e.g. chemical composition, spatial-temporal information of phases) as an input for the model and is validated against micro-scale experimental data and macro-scale (continuum scale) measurements of transport properties (porosity, tortuosity, permeability).

Objectives for different specific steps/periods (if applicable):

The objective of SCK•CEN is to study the interaction between a high pH cementitious material and a clayey host rock (Boom Clay), with a focus on the chemical-physical alterations within concrete. Specific objectives are:

- To validate if the modeling approach is able to predict macro-scale (continuum) transport properties (few hundreds of μm³ to mm³) using experimental information at the micro-scale (mainly spatial distribution of different concrete phase at μm scale).
- To apply a reactive coupled micro scale model for simulating the spatial-temporal evolution of geochemical changes.

To identify the most critical processes and parameters which affects the microstructural alterations and the macroscopic transport properties.

Main expected outcome:

- Validated micro scale flow and transport model for predicting macroscopic transport properties used on microstructural information from experiments on the interactions at the interface between a cement-based material and a host rock
- Developed conceptual model for simulating geochemical, microstructural and macroscopic transport properties within cement-based materials at the interface with a host rock
- List of sensitive parameters and processes affecting the micro scale alterations and macroscopic transport properties during the interaction of a cement-based material and a host rock
- Guidelines for continuum scale modeling of the interface processes

Description of work to be performed:
The work package will be performed with a three-dimensional coupled reactive transport model based on a lattice Boltzmann approach to solve the water flow and solute transport equations and iPHREEQC as the geochemical solver. The lattice Boltzmann approach allows incorporating the spatial distribution of different relevant phases in a discrete manner enabling to simulate alterations of micro scale properties and features and the macroscopic transport variables. This allows simulating the interface processes starting from basic principles. SCK•CEN has developed this three-dimensional code and demonstrated its versatility for cementitious materials (e.g., Patel et al., 2014) including explicitly C-S-H transport properties

Task 1 – Prediction of macroscopic variables from microstructural information
The first task provides a validation of the approach to predict macroscopic physical properties using a given microstructure (unaltered or altered) by comparison with measured macroscopic properties. The spatial distribution of concrete phases forms the input to the water flow and

WP 3			SCK•CEN

solute transport solvers in the lattice Boltzmann framework for calculating selected macroscopic transport properties. The main challenge is to link the estimated transport properties to the measurements between which a scale-discrepancy exists. This may result in the use of additional upscaling techniques. A second challenge is the expected anisotropy in degraded samples as opposed to isotropy in well hydrated initial samples.
Two approaches are followed to render the three-dimensional geometries: (i) The multi-scale investigation methodology described in WP1 of SCK•CEN and BRGM (FIB-SEM) gives a direct experimental measurement of the 3D geometry; and (ii) derivation of 3D geometries from 2D SEM images using the available information on the spatial arrangement of the different phases (e.g. by multi-point statistical algorithms starting from 2D SEM images which serves as training images. The main challenges are to extend and apply the algorithm to multiphase systems and to anisotropic system. Part of the validation exists in comparison of statistical parameters characterizing the spatial structure between the 3D geometries from (i) and (ii).

Task 2 – Screening of important microstructural features affecting the macroscopic transport properties
Within this task, the sensitivity of basic concrete properties (e.g., type of cement, w/c ratio, aggregate content, etc.) on parameters characterizing the microstructure (e.g., pore size distribution, connectivity, geostatistical parameters describing the spatial correlation of the different phases) and macroscopic transport properties during different stages of degradation. Because a full sensitivity analysis is not possible within the time available within this project, an exploratory sensitivity study will be performed.
This could be based on the available SEM and FIB-SEM data from WP1 in four consecutive steps creating a kind of series of virtual porous media: (i) characterizing statistical parameters characterizing the microstructure from the SEM images, (ii) obtaining statistically generated training images by perturbing the statistical parameters, (iii) generating 3D geometries with the multi-point statistical algorithm, and (iv) performing lattice Boltzmann calculations.
More ideally and preferable (but depending on the available experimental data), experimentally obtained SEM images from other experiments in WP1 in this project could be used directly to generate 3D geometries (thus taking step (iii) and (iv) only as outlined above). This is of particular interest for experimental set-ups (e.g. natural analogues) in which direct measurements of macroscopic properties are not possible. In addition, this will give information on real porous media instead of 'virtual' ones (statistically derived).

Task 3 – Prediction of changes in geochemical, microstructural and macroscopic transport properties
The methodology of task 1 is extended for simulating geochemical alterations due to chemical gradients between the host rock (Boom Clay) and the cement-based material (high pH concrete). Again, experimental data from WP1 (SCK•CEN and BRGM) are used to generate the initial 3D geometry of the concrete phases. The reactive lattice Boltzmann model provides numerical data on the change in spatial distribution of solid phases, geochemical composition, and microstructure. At regular times (cfr. times in the experiments), macroscopic transport properties are calculated. The geochemical models for the cement-based material and for the Boom clay (percolation tests or samples for HADES) are based on previous work on concrete degradation (e.g., Jacques et al., 2010, Jacques et al., 2014) and experience within SCK•CEN. Comparison of the model with experimental data is done in a statistical way by comparing different variables (e.g. element concentration and ratios at micro scale, phase fractions, ~µm³), as function of the interface distance.

Cooperations

WP 3			SCK•CEN
Strong link with the experimental program of the SCK•CEN – BRGM proposal in WP1			
Plans for dissemination: Joint papers, posters/presentations at conferences, open research reports			
Deliverables and milestones proposed (the information will be integrated for the respective workpackages): **Milestone 1:** Validated modelling framework to calculate macroscopic transport properties from experimental micro scale information on spatial distribution of different phases. **Deliverable 1:** Prediction of macroscopic transport properties using (FIB)SEM images: methodology and screening analysis (report). **Deliverable 2:** Modelling and interpretation of selected experiments on alterations at the interface between a high pH concrete and Boom Clay (report). This work will be integrated in the 'Modelling Task' within WP2. SCK-CEN will contribute to this integration activity.			

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WP 3			UDC

Overall Objectives:

1) Improve the reactive transport models and codes for modelling the cement-clay interactions and the chemical, mechanical and hydrodynamic couplings. Improvements include: a) Accounting for the deformation with a coupled THC-M model, b) Considering the time evolution of porosity, permeability and diffusion coefficient, c) Identifying the low-crystal size mineral phases such as CSH, MSH and CASH; d) Account for the retention of anionic species (C, Cl and sulphate species) and e) Accounting for carbonation processes. These model developments will be tested with already existing lab test data.
2) Testing model improvements with experimental data from heating and hydration tests carried out by Ciemat on compacted bentonite (HB tests) to study the interactions of concrete and bentonite under repository conditions and analyze how such interactions affect the concrete and bentonite properties. The results of these tests are already available and will also be used to evaluate time-scale effects because these tests were dismantled after times ranging from 0.5 to 7 years.
3) Long-term reactive transport modeling of the concrete and bentonite barrier system of a high-level radioactive waste repository in a clay formation (performance assessment)
4) Evaluate model time-scale effects from HB test data

Main expected outcome:

- Improved reactive transport models and codes for modelling the cement-clay interactions and their chemical, mechanical and hydrodynamic couplings. Model testing and validation with laboratory experiments. Evaluate time-scale effects. Extrapolation to the long-term performance of the repository.

Description of work to be performed:

- Task 1: Improvements of reactive transport models and codes for modelling the cement-clay interactions and their chemical, mechanical and hydrodynamic couplings. The reactive transport code of UDC will be improved and extended to cope with: a) The deformation with a coupled THC-M model, b) The time evolution of porosity, permeability and diffusion coefficient, c) The identification of the low-crystal size mineral phases such as CSH, MSH and CASH; d) The retention of anionic species (C, Cl and sulphate species) and e) The carbonation processes. These model developments will be tested against already existing data.
- Task 2: Numerical model of the heating and hydration laboratory experiments performed by Ciemat on compacted bentonite (HB tests) to study the interactions of concrete and bentonite under repository conditions. The model will include these features: a) Non-isothermal single and two phase flow, b) Thermodynamic & kinetic models, c) By accounting for the deformation with a coupled THC-M model, d) By considering the time evolution of porosity, permeability and diffusion coefficient, and e) Carbonation processes.
- Task 3: Long-term reactive transport modeling of the concrete and bentonite barrier system of a high-level radioactive waste repository in a clay formation (performance assessment)
- Task 4: Evaluate model upscaling in time. Contribute to the Modeling Task of WP 3.

WP 3			UDC

Cooperations

1) *With the rest of the partners of WP3: on the Modeling Task*
2) *With CIEMAT-AEG/UAM/CSIC who will provide the data for modeling the HB laboratory experiments*
3) *With CEBAMA external partners: ENRESA (Spain)*

Planned training activities (if any):

- *UDC will contribute to training courses on geochemical reactive transport modeling focused on cement-clay interactions.*

Plans for dissemination:

- *1) Present results at national, European and international conferences and meetings;*
- *2) Publications of technical reports; and*
- *3) Publications of papers in ISI journals.*

WP 3			CTU

Overall Objectives:
Czech Technical University in Prague (CTU), Department of Nuclear Chemistry (DNS) will contribute to the development and use of interaction models and transport codes that will help to evaluate experiments performed in WP1 and WP2 in UJV and CTU (description of bentonite-cement contact and interaction of selected radionuclides with cementitious materials). A previous experience with the modelling geochemistry and diffusion transport in the PHREEQC3 environment, in which an advanced description of interaction of studied species with the rock surface (ion exchange and/or surface complexation) was utilized, will be further developed. An original approach for evaluation of in-diffusion experiments respecting a broader set of conditions of experiment, which we realized with use of GoldSim, will be extended also to systems that should be described by the smart Kd approach.

Objectives for different specific steps/periods (if applicable):

- Improvement of methods of evaluation of diffusion experiments with non-sorbing species.
- Development of geochemical model of interaction of cement and bentonite and application them of the evaluation of experiments performed in WP1 by UJV and CTU-CEG.
- Formulation of an advanced model of interaction of Ra with selected cementitious materials used in the experimental part of the study.
- Development of methods that will contribute to an effective incorporation of the smart Kd concept into transport codes (for Ra retention and diffusion).

Main expected outcome:

- Evaluation of a set of diffusion experiments of different non-sorbing species in the cement blocks performed in UJV. Participation on the interpretation of obtained results.
- Proposals for effective incorporation of non-linear sorption models of Ra into a transport code used in the laboratory, simulations performed with this code and evaluation own diffusion experiments (WP2).

Description of work to be performed:

- Use of characterization of cementitious materials obtained in WP1 and WP2 in CTU and UJV to formulation of geochemical model of interaction of cement and bentonite.
- Improvement of diffusion model prepared in GoldSim that will respect the approach of smart Kd.
- Formulation of model of Ra interaction with cement and application of it in a diffusion model realized in PHREEQC.
- Evaluation and interpretation of experiments performed in laboratories of UJV and DNS.

Cooperations
CTU-DNS will closely cooperate with CTU-CEG and UJV Rez in laboratory experiments and with SURAO by dissemination of results.

Planned training activities (if any): involvement of 1-2 Ph.D. students and 1-2 pre-graduates

Plans for dissemination: Transfer of data and experience with formulation of models and simulation within the project, to students and to scientific community.

Deliverables and milestones proposed (the information will be integrated for the respective workpackages):

M1 – Construction of models describing (i) diffusion of non-sorbing species in samples of geometry relevant for WP1 and cement-bentonite interactions (for experiments prepared by UJV and CTU-CEG, WP1) and (ii) uptake of sorbing species on cementititous materials (based on own preliminary study with Sr^{2+}). Periodic report – after 2nd year.

M2 – Optimization of evaluation methods of experimental results obtained in WP1 and WP2 realized in GoldSim and UCODE+PHREEQC. Periodic report – after 3rd year.

WP 3			CTU
M3 – Final report: Interpretation of experimental results obtained in WP1 and WP2 by co-operating Czech laboratories.			

WP 3			VTT

Overall Objectives:

On the basis of WP1 results, existing modeling methods and predictions[2],[3] will be evaluated using computer programs such as RHREEQC or Geochemist's Workbench. Quantitative microstructural models of CSH degradation in varying environments will be improved based on the iterative process with improved results obtained in WP1. Results will either validate the modelling or give an estimation of the modelling reliability.

Objectives for different specific steps/periods (if applicable):

- Validate geochemical simulations applicability to various C-S-H degradation and ion replacement mechanisms. Models of batch reactions.
- Develop time dependent model of pozzolanic materials, clay/groundwater leaching and groundwater compositions, focused on low-pH concrete.

Main expected outcome:

- Improved understanding of geochemical modelling of calcium-silicate-hydrates in nuclear waste repository environment (report).
- Justified, transparent and credible numerical model of alkaline plume development caused by low-pH concrete degradation in different groundwater/clay environment.

Description of work to be performed:

VTT will develop numerical model to predict time depended effects of low-pH concrete/ groundwater/ clay interaction on formation of alkaline plume. Evolution of solid composition and external solution will be modelled. Modeling is divided to two tasks, first one modelling equilibrium compositions and C-S-H degradation in batch reactions with geochemical computer simulations (the actual samples are studied in WP1, enabling experimental verification). Second task is development of the time dependent model.

Task 3.x.1: Simulation of batch reactions will be performed with PHREEQC (or Geochemist's Workbench) –computer program. Simulation will give the estimation of equilibrium compositions and concentrations in various leaching volumes. Also the uncertainties in C-S-H degradation and ion replacement will be dealt individually, enabling further corrections in the time depended numerical model.

Task 3.x.2: Time depend effects of low-pH concrete/ groundwater/ clay will be modelled with the same programs. Using existing databases, such as Thermochimie, Thermoddem etc. Extreme care will be taken to model the solid solution behavior of C-S-H. Time dependency of pozzolanic reaction and groundwater leaching will be included in the model. Final corrections to the model will be made on the basis of information gained from batch reactions from sub-task 3.x.1.

The VTT work effort of 5 PM corresponds to 53 k€ of work effort, as well as travel costs associated with a researcher exchange visit to another partner institute (i.e. potentially to Amphos21).

Cooperations:

Complimentary modeling methodologies will be employed by the different RTD providers to optimise a spectrum of results. Close cooperation is hoped with Amphos21.

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[2] Soler, Joseph. Reactive transport modeling of grout-water interaction in a fracture at the ONKALO site, Posiva 2011-83 (2011)

[3] Koskinen, Kari. Effects of Cementitious Leachates in the EBS, Posiva 2013-04 (2014), 55p.

WP 3			VTT

Close cooperation and iteration with WP1will be established throughout the work, so that modeling is developed simultaneously to experimental studies (cooperation with CTU, Andra/LML).

Close cooperation with POSIVA, SKB, ANDRA and other nuclear waste management organisations will be arranged by establishing work package steering group where special technical questions concerning this work package will be addressed.

Added value of Cebama:

With close cooperation between Cebama partners modeling work can be performed with high efficiency. Different partners have high knowledge on different modeling subjects and methods. By interchanging information and experience fast progress is expected. VTT will bring experience on cementitious materials reaction chemistry, kinetic and morphology and experience on utilizing it in geochemical environment. Ability for multiple countries to apply similar modeling techniques and validation, using local material and environmental parameters.

Planned training activities (if any):

Expect to have researcher (PhD student) exchange with other partners, especially to gain competences and exchange of information to Spain.

Plans for dissemination (note: similar to WP1 text):

Two PhD dissertations will use results from the Cebama project in their work. Both works are expected to be finalized during the project duration. One of the works is concentrated on WP1 and second more on WP3.

At least 3 scientific journal publications will be generated (i.e. *Journal of Cement and Concrete Research*, *Journal of Applied Materials*) along with 3 conference and/or trade journal presentations (such as and IGD-TP events, Posiva Oy and VTT organization's public magazines). VTT will contribute to the Cebama web page and periodic newsletters.

VTT as a non-profit research organisation will disseminate the results of this research in the future research and development projects at national and EU level. Through the good connections with their local stakeholders, VTT can spread and promote the technologies developed in this research, thus helping nuclear waste management organisations in their assignment. VTT's own know-how and competence in cement based materials - groundwater - bentonite interaction related subjects will strengthen through this project, and cooperation at EU level will be reinforced.

WP 3			LML

Overall Objectives:

The LML will develop multi-scale modeling for hydromechanical behaviors of concrete and concrete/argillite interface by considering effects of mineralogical compositions, temperature, relative humidity and microstructure evolution due to carbonation.

Objectives for different specific steps/periods (if applicable):

- Task 1: development of a micro-macro model for concrete using nonlinear homogenization techniques
- Task 2: development of a hydromechanical model for concrete/Cox clay interface
- Task 3: numerical analysis of typical structures including concrete, Cox clay and interface

Main expected outcome:

- Development of original multi-scale models for concrete and concrete/argillite interface taking into account effects of microstructure evolution and environmental conditions;
- Development of an efficient tool for numerical modeling of multi-physical coupling problems including concrete/argillite interface

Description of work to be performed:

The work proposed in WP3 will be essentially based on the state-of-the-art and experimental obtained in WP1. In previous studies, the LML developed a high expertise on thermo-hydromechanical modelling of cement-based materials and clayey rocks as well as interfaces. In particular, the LML takes a leader positions on multi-scale approaches of heterogeneous geomaterials. Three tasks are proposed.

In the task 1, it is proposed to develop a micro-macro constitutive model for concrete using nonlinear homogenization techniques. Three scales will be considered. At the microscopic scale, the mechanical behaviour of solid CSH grains and the effect of pores between such grains will be taken into account. At the mesoscopic scale, the effect of small size inclusions (sand grains for instance) embedded in the porous cement paste will be considered. At the macroscopic scale, the effect of large size aggregates will be finally investigated. Using rigorous nonlinear homogenization techniques, we will establish a close-form or semi close-form macroscopic failure criterion explicitly taking into account the effects of pores, small inclusions and large aggregates. The advantage of the multi-scale approach is that various microstructural parameters (pores, inclusions etc.) are explicitly included in the macroscopic criterion. Therefore, it is possible to explicitly account for the effect of chemical degradation on the macroscopic properties. For instance, the decrease of porosity and the production of calcite due to carbonation can be explicitly considered by the micro-macro model. Finally, including the local effects of temperature and relative humidity (or saturation) on the mechanical behaviours of cement paste, it is also possible to properly investigate the thermo-mechanical and hydric-mechanical coupling by the micro-macro model. The model’s parameters will be identified from representative data on low pH concretes previously investigated by Andra.

In the task 2, we propose to develop a hydromechanical model for concrete-argillite interface. Again a micro-macro approach will be employed to capture the effects of microstructural parameters such as local asperity and chemical reactions. This model will be able to describe the mechanical behaviour of interface under combined normal and shear stress, the evolution of

WP 3			LML

permeability, the modification of interface microstructure (asperity degradation, carbonation). The parameters of the interface model will be identified from the laboratory tests performed in the WP1 by the LML. The last step of this task is devoted to the numerical simulations of laboratory tests using the proposed hydromechanical model. For this purpose, we propose to simulate not only the laboratory tests performed in WP1 but also the previous tests conducted by the LML in the framework of Andra's programs.

The task 3 is devoted to the numerical analysis of typical structures including concrete/argillite interfaces. For this purpose, the proposed constitutive models for concrete and interface will be implemented in an extended finite element (XFEM) code. The emphasis will be put on the hydromechanical behaviour of the interface between concrete and argillite (Cox clay). The XFEM method is particularly suitable for modelling this kind of imperfect interfaces with displacement discontinuity. The LML has developed a XFEM code which is able to deal with THM-C (thermo-hydromechanical and chemical) coupling problems in both saturated and partially saturated conditions. A simplified structure will be designed including concrete, Cox clay and interface. For instance, we can consider a representative section of gallery selling structure including expansive clay, concrete support massif, concrete lining and Cox argillite. A series of numerical calculations will be realized in order to investigate the THM responses in such a structure and in particular in the interface. The objective is to study the key role of the interface in the long term stability of sealing structures in storage boreholes.

Cooperations:

- *The LML will take advantage of Andra's expertise for the design of the simplified structure to be modeled in Task 3.*
- *No cooperation expected with external partners*

Added value of Cebama: Multi-disciplinary and complementary approaches will be a key factor for the success of such a complex project

Plans for dissemination:

- Publication of papers in peer-reviewed journals
- Communications in international conferences

Deliverables and milestones proposed (the information will be integrated for the respective workpackages):

Formulation of original multi-scale models for the hydromechanical behavior of concrete and concrete/argillite interface

Development of a new numerical code for multi-physical modeling of typical structures including concrete/argillite interface

WP 3			ANDRA

Overall Objectives:

Integration of information in order to improve multi-physical representation of disposal structures behaviour

Main expected outcome:

Thanks to the up-scaling of the data obtained from other work packages at small scale, this WP3 aims at:

- Set up an integrated model of the phenomenological evolution of ILLW disposal cells considering the radionuclide retention on concrete in saturated condition and in chemically evaluated concrete
- Modelling (hydro)chemical evolution of seal and ILLW disposal
- Performance Assessment at global scale of ILLW disposal including radionuclide transfer (quantification of performance indicators related to RN release and migration)

Description of work to be performed:

- First task is part of the “Modelling Task”. It will consist of developing conceptualisation of the model in order to represent the chemical and radionuclide (CR) evolution of seal and the ILL tunnel at large scale. This work is done by setting up a methodology of up-scaling the data/models obtained in experiments developed in the frame of the R&D program of Andra. According to the progress of mechanical processes model coming from the *in-situ* experiment developed in the WP1, the mechanical processes will be also conceptualised. Thus, part of extrapolation will be based on experiments modelling, taking into account uncertainties (epistemic and stochastic variability of the acquired data). Some focus will be made on specific phenomenology such as the determinist model of radionuclide uptake and redox evolution of interstitial water and consequences on radionuclide retardation.

- Second task will consist in modelling tasks divided into two parts:
    - (1) (HM)CR modelling of the seal and the ILL tunnel over a large scale of time and space. Concerning the ILL tunnels, improvement should be provided by taking into account more continuously in time and space the chemical evolution of the OPC materials in consistency with the related up-take processes of radionuclides. Concerning the seal, the modelling will focus on interface between low pH concrete and clay materials (host rock, bentonite). Levels of coupling (M and C) will depend on results from of the R&D program of Andra and numerical capabilities of the tools;
    - (2) modelling at large scale in Performance Assessment perspective, taking into account results from WP3. At this scale, hydraulic, chemical and mechanical evolution will be taken in a simplified way, by modifying/adjusting hydro-dispersive and retention input data of material and their interface in space and time. Thus, hydraulic and radionuclide fluxes will be provided through different surfaces, which would increase the knowledge of radionuclide transfer in disposal cell up to long time scales. At the ILLW disposal scale, performance indicators will be proposed based on radionuclides release. A special focus will concern the seal where low pH concretes are in contact with clay host rock and bentonite. Because of the role of seals in hydraulic flow distribution at disposal scale, performance indicators based on hydromechanics evolution of the closure system will be proposed.

WP 3			ANDRA
Cooperations *The conceptualisation step is based on data/model acquisition.*			
Plans for dissemination: • WP3 Technical meeting presentations and general assembly			

WP 3			PSI

Overall Objectives:

Development of improved 2D and 3D conceptual and numerical models for couplings between chemical and transport processes. Testing and comparison of different model approaches by analyzing experiments on interactions of concrete with clays or other materials. Application on long term geochemical evolution of repository near field.

Main expected outcome:

Enhanced understanding of how to abstract feedback between transport of solutes, chemical reactions and porosity clogging in numerical models on different scales.

Description of work to be performed:

- Implementation of electrically coupled multi-species transport in multi-porous media in open-source reactive transport code OpenGeoSys6-GEM in combination with compatible models that capture effects of charged surfaces implemented in open source LMA and GEM chemical solvers (GEMS3K and Reaktoro). We plan to use a Donnan approach for effects of charged mineral surfaces/interlayers on pore water composition; a solid-solution approach for cation exchange, surface complexation and/or interlayer hydration of clay minerals.
- Testing the applicability of new implementation by analysis of multi-tracer experiments periodically conducted at material interfaces that are prone to change of porosity. Experimental data from two ongoing PhD projects at PSI on porosity evolution at material interfaces, from Cement-Clay Interaction Project at the Mont Terri Underground Rock Laboratory (see project Uni Bern) and from other experimental projects in WP1 (SPECT/CT at PSI in cooperation with Uni Bern, PET in cooperation with HZRD, …).
- Application with 2D/3D cavern and/or repository scale models on long term geochemical evolution of a heterogeneous repository near field.

Cooperations

- *Close collaboration with Uni Bern on evaluation/planning of transport experiments (e.g. SPECT/CT) and sample characterization*
- *Collaboration with experimental projects in WP1 that invest transport over material interfaces*
- *Cebama external partners UFZ Leipzig (OpenGeoSys-GEM development)*

Added value of Cebama:

Empa/PSI will develop a thermodynamic model of Se and I uptake by cementitious materials, considering S(-II) competition under reducing conditions, which can be implemented in reactive transport models.

Planned training activities (if any):

PhD student should receive training in thermodynamic modelling, reactive transport modelling, and modern software development with C++.
Courses on thermodynamic modeling with GEMS-PSI (in cooperation with e.g. EMPA) and on reactive transport modeling with OpenGeoSys-GEM could be organized in the framework of the project.

Plans for dissemination:

Presentation at international conferences, publication of results in peer-reviewed scientific journals, publication of code (open-source)