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Basis for application to Safety Case and Performance Assessment

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1 INTRODUCTION

This report series aims at interpreting how the results of the research conducted within CEBAMA can be applied and contribute to the Safety Case. This is the first of two reports, which will be updated at the end of the project. Given the complexity of the experiments and the models developed within CEBAMA, it is in general too early at PM18 for the partners to produce results directly applicable to the Safety Case. Nevertheless, we present here some briefing on the impact aimed at within the project and highlight the value of some of the work undertaken to date.

Cement-based materials are key materials used in repositories of low, intermediate and high level waste in crystalline or clay rock host-rock concept. These materials are used as waste forms, as liners, as seals as well as structural components. The relevance that the performance of these materials has for the Safety Case is obvious, and this is why CEBAMA focuses on the study of cementitious materials. Having been used for so long time and for multipurpose functions, not only in the radioactive waste community but in many others, quite large amount of data on these materials exist. Cebama addresses key issues of relevance for *long term safety* and key scientific questions related to the use of cement-based materials in nuclear waste disposal applications. The scientific quality and impact of the project builds on joining the best expertise available to tackle these problems and emphasizing how the knowledge can be applied in Performance Assessment and the Safety Case**.**

According to IAEA (2012)[1] the Safety Case is the collection of scientific, technical, administrative and managerial arguments and evidences in support of the safety of a disposal facility, covering the suitability of the site and the design, construction and operation of the facility, the quantitative assessment of short and long term risks as well as more qualitative safety indicators and assurance of the adequacy and quality of all of the safety related work associated with the disposal facility. Safety Assessment, an integral part of the Safety Case, is driven by a systematic assessment of radiation hazards.

Among other objectives, CEBAMA aims at improving the knowledge base for the Safety Case via the following specific items:

- Safety impact of microstructural and porosity changes of cementitious materials
- Safety impact of cement degradation

[1] IAEA (2012) The Safety Case and Safety Assessment for the Disposal of Radioactive Waste. IAEA Safety Standard Series, Specific Safety Guide. No. SSG--23. International Atomic Energy Agency Vienna, 2012

- Creation of long-term models
- Decrease uncertainties in radionuclide retention processes
- Development of modelling expertise and methodologies
- Upscaling modelling in time and space

To ensure that the project is directed towards application, the End User's Group represent the view of the users of the research. The organisations taking part of the EUG of CEBAMA are:

- NAGRA (CH)
- ANDRA (FR)
- POSIVA (FI)
- SKB (SE)
- COVRA (NL)
- ONDRAF/NIRAS (BE)
- RMW (UK)
- ENRESA (ES)
- SURAO (CZ)

The former organisations come from countries in very different stages of implementation, so that the implication that the results of CEBAMA have on their interests may vary from case to case. A brief description of the relevance that cementitious materials have in each of the concepts represented by the EUG has been detailed in Deliverable D1.03 and will not be repeated here. What is clear is that there are many aspects of cement-based materials to tackle with and some have been investigated within particular national and international projects. Open issues are many and although CEBAMA will not be able to address them all, it will contribute to decrease uncertainties, filling in knowledge gaps that impact the Safety Case and the associated Safety/Performance Assessment. From the discussions held along the project and from the description of each one of the concepts, we can delimit some the aspects of interest to investigate and that are specifically planned within the CEBAMA research activities, which are:

- How do cement-based materials affect the isolation properties of the host rock and the clay backfill? Experimental studies are performed to understand the interface processes between cement-based materials and the host rocks (crystalline rock, Boom Clay, Opalinus Clay (OPA), Callovo-Oxfordian (COX)) or bentonite backfill and assess the impact on physical (transport) properties are addressed by WP1
- How do specific radionuclides of interest behave in the presence of cement-based materials, or in media altered by the presence of these materials? WP2 addresses the behaviour of radionuclides which have high priority from the scientific and applied perspective in cement-driven environments: Be, C, Cl, Ca, Se, Mo, I, Ra
- How well are we able to predict changes in transport properties of cement-Improve validity of numerical models to predict changes in transport? Work is aiming to support advanced data interpretation and process modelling, covering mainly issues responsible for the changes in transport properties and extrapolate the models for safety case application.

The former investigations will help the Waste Management Organisations to:

- optimize geological site selection processes based on knowledge regarding groundwater and host rock impacts to engineered barrier system safety,
- modify the design basis (such as density and dimension requirements) for engineered barrier systems, including concrete and clay components;

- adjust material characteristics specifying required parameters (such as montmorillonite content of clay, percentage allowance of trace chemicals);
- react to deviations in material parameters, which may arise during quality control or construction,
- reduce redundancy in designs needed for safety margins, due to increased confidence in the engineer barrier system design tolerances and bounding uncertainties,
- provide scientific and transparent justifications to safety regulatory authorities, related to engineered barrier designs and specifications, thus accelerating the implementation toward repository operation.

The complexity of the investigations within CEBAMA produces that at an early stage of the project, the direct impact on the Safety Case cannot be readily appreciated. In the following sections we have made an effort to briefly highlight the value of the investigations already undertaken in the Safety Case (SC) and the Safety/Performance Assessment (SA, PA).

2 Early contributions of WP1 to the Safety Case and Performance Assessment

WP1 has the largest work effort in the project and is directly focused on the study of cementitious materials and their impact on the clay-buffer material present in the repository either as barrier or as host-rock. The stability of clay materials in engineered barrier systems is potentially endangered by the high pH leachates from cement–based materials. According to Safety Assessment, the pH of the cementitious leachates should mainly remain under pH 10. The developed methods in Cebama aim at generating a reliable procedure to assess the pH of the cementitious leachate considering a diversity of compositions both of the material and of the interacting groundwater.

The objectives of WP1 for the first period are as follows:

- preparation of concrete samples casted for verification studies,
- determine the pH of various low-pH mix designs,
- studying the effect of the leaching of cementitious materials on clay materials of engineered barrier system.

There has been a tremendous work developed by WP1 during this initial stage of the project in selecting and preparing samples for analyses, besides initial experimental set ups.

It is interesting to note that there are already direct inputs to the NWMO of the work developed in WP1, helping in the licensing process of POSIVA, by contributing to give answers raised by the regulatory authority (STUK).

A brief summary of the contribution of some of the works already undertaken for the Safety Case is given below.

- Characterization of old (13 years) Boom Clay-Concrete interfaces from the HADES laboratory, contributing to process understanding.
- Studies of Callovo Oxfordian clay in contact with low pH concrete have started.
- Characterization of FEBEX bentonite-concrete interface samples.
- Preliminary transport tests with CEMV columns indicate an increase of the porosity with decrease in the retardation coefficient of 36Cl, of interest for transport prediction purposes.

- Assessment of pH development in leachate after interaction of cement with different groundwater compositions, including saline granitic groundwater and saline bentonite groundwater, has been conducted what provides basic information needed for simulations supporting SA.
- Visualization techniques have been tested, that will be applicable during operation and closure of repositories.
- Alteration of bentonite in contact with shotcrete from FEBEX experiments (13 years contact) has shown carbonate concentration and precipitation of Mg-silicates as main alteration processes. These affect a depth below 5 mm of concrete from the bentonite interface. A disappearance of Portlandite and incorporation of Al and SO_3, as well as formation of Ettringite has been observed at 1cm from the clay interface. Cl has migrated 5cm in concrete. These results confirm basic information for process understanding and input to long-term models of concrete-bentonite interfaces supporting SA.
- Comparison of the behavior of high and low pH concrete is underway and will provide results in the second period of the project, very valuable for design and selection of materials to be used in the repository construction.

3 Early contributions of WP2 to the Safety Case and Performance Assessment

WP2 focuses on the behaviour of some radionuclides which have high priority from the scientific and safety case perspective in cement-driven environments, which were selected to be: Be, Ra, C, Cl, Tc, Se, Mo and I.

The approach of WP2 is bottom-up, generating basic data with relatively simplified systems to incorporate in more complex systems and models. A description of the systems and models to study that allow this bottom-up line was given in deliverable D2.02, and is summarised in Table 3-1 below to show how this WP builds from simple to more complex systems.

Table 3-1. Summary of work plan of WP2, indicating the type of systems studied for each radionuclide and the partners undertaking the work. Sol. Indicates whether solubility experiments will be conducted. S: sorption experiments. C: solid Characterization. D: Diffusion tests.

RN	Partner	Sol.	high pH cement-based materials								individual phases			
			CEM I		CEM II		CEM V		Mortar		CSH	AFm	AFt	LDH
			Fresh	Age.	Fresh	Age.	Fresh	Age.	Fresh	Age.				
Ra	KIT-INE		S								S			
	JUELICH		S+C	S+C			S+C	S+C			S+C	S+C	S+C	S+C
	RATEN		S+D+C				S+D+C							
	CTU	Yes			S+D	S			S+D	S				
	SURREY	Yes	S+C								S+C	S+C	S+C	S+C
Be	KIT-INE	Yes	S								S	S		
Mo	A21	Yes					S+C				S+C	(S)		
	BRGM											S		
	JUELICH		S+C	S+C			S+C	S+C			S+C	S+C	S+C	S+C
Se	BRGM											S		
	JUELICH		S+C	S+C			S+C	S+C			S+C	S+C	S+C	S+C
	PSI/EMPA											S+C		
	SURREY	Yes	S+C								S+C	S+C	S+C	S+C
I	JUELICH		S+C	S+C			S+C	S+C			S+C	S+C	S+C	S+C
	SURREY	Yes	S+C								S+C	S+C	S+C	S+C
	PSI/EMPA											S+C		
C-14	RATEN		S+D+C				S+D+C							
	ARMINES						D							
Cl, Tc	SURREY	Yes	S+C								S+C	S+C	S+C	S+C
Sr	CTU	Yes			S+D	S			S+D	S				

As can be appreciated in the previous table, the experiments will provide parameters related with sorption, diffusion and solubility which are directly applicable in safety assessment. The detailed study of the processes underlying the parameters is facilitated by the bottom-up approach, and is a requisite to produce robust arguments supporting the Safety Case. Gaining and showing process understanding is a cornerstone in any Safety Assessment.

It is worth mentioning the State-of-the-art report, which was submitted as deliverable D2.03 and contained a very detailed review of previous work of interest for the objectives of this WP.

Despite the early state of the project, some of the works undertaken already have produced relevant data and/or observations to include in PA exercises. Some of them are briefly commented below.

- The studies undertaken on Ra indicated that this element was rapidly and strongly retained in CSH but not on AFm, what must be recognized in the models developed to predict the behavior of Ra in the long-term evolution of cementitious materials.
- Selenium, as molybdenum, form oxo-anions under cementitious conditions, normally presenting lower degree of retention. However, the large quantities of concrete used may still provide for important retention of these ions. The understanding of the processes responsible for the retention of these radionuclides and its quantification is therefore very relevant for the SC and PA studies.
- The solid powellite ($CaMoO_4$) was predicted as solubility controlling phase of molybdenum under the cementitious conditions prevailing in ILLW repositories. Preliminary results on the study of the solubility of this solid phase are in agreement with the calculated solubilities. This represents an increase in the reliability of concentration limits used in many of the solubility assessments undertaken by the NWMO and applied as concentration limits for the

Safety and Performance Assessment Exercises. Powellite may not only limit the release of toxic Mo and of radiotoxic 41Ca, it may also act as host phase for trivalent actinides.

- The experimental findings indicated that the stability of Se(VI)-containing AFm was higher than that of Se(IV)-bearing AFm. This result may indicate higher retention of Se(VI) than Se(IV), with implications for PA calculations. Experiments are underway to determine the results and will be ready in due time.
- The study of the dissolution of AFm-Cl phases was studied and it was found to be congruent, i.e., Ca/Al ratio similar in the leaching solution and in the initial solid phase, for pH between 10 and 13. On the contrary, the dissolution was incongruent, with a higher ratio Ca/Al in solution than in the solid phase for pH around 9. This implies that in supporting models the process cannot be considered congruent for degraded stages of the system, when the pH falls below 10, and other processes accounting for reduction of Al concentration in solution, such as Al containing phases, must be incorporated. Cl seems to be a reasonable indicator for AFm-Cl dissolution rate, as the rate of dissolution based on Cl release was determined to be independent of pH.
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The results obtained so far contribute to ascertain kinetics of retention and release, as well as the extent of retention of several radionuclides on simplified solid systems.

4 Early contributions of WP3 to the Safety Case and Performance Assessment

This is the work package in which application to the Safety Case and Performance Assessment can be more easily evaluated. Some of the models are already being applied for preliminary evaluation of long-term performance of concrete structures by some NWMO. A critical value of this workpackage relies on the development of upscaling methods at two levels: space and time, at sizes and times beyond possible experimentation. Therefore, focus with application to Safety Case and Performance Assessment of WP3 is on:

- modelling the interrelation between chemical changes of the cement paste microstructure and its physico-chemical properties,
- upscaling the microstructural features of cement and concrete relation to incorporate them in macroscopic reactive transport models,
- extrapolating numerical models for long-term conditions.

The beneficiaries have worked on model developments from the onset of the project, preparing the numerical models and testing them in front of previously existing experimental data, given that the experimental data in WP1 and WP2 is not ready for modelling until the project is advanced. A summary of the safety assessment-relevant production at this early stage of the project by WP3 is given below.

- Verification in front of experimental data of the hydro-chemo-mechanical model for degradation of cementitious materials which is already being used by SKB for preliminary evaluation of long-term performance of concrete structures in SFL, the Swedish repository for LILW.
- Advances in upscaling from microstructure to macro-model have been made by:
    - Incorporating the effect of electric potential on ion-diffusion rates,
    - Selecting secondary zeolite minerals to include in the long-term models,

    - Diffusivity prediction in cement paste materials,
    - Evaluation of porosity, permeability and diffusion caused by geochemical reactions (dissolution/precipitation).
- Evaluation of the applicability of modelling tools to assess the effect of water compositions to a high-pH plume originating from degradation of cementitious materials was studied. These results are already feeding Posiva to answer questions arising from the licensing applications to the Finnish regulatory authority.

5 Future work

In order to analyse the work of the project in the context of Safety and Performance Assessment, and for preparation of reporting the outcome of the project, a questionnaire is being prepared to be filled by the EUG on the issues of relevance for CEBAMA.

The objective is to compile information on how each specific issue is being dealt with in the actual state of the SC/PA of each one of the organisation. This will allow a monitoring of how the project contributes, at its closure, to modify, update, improve the processes, concepts and parameters used by the agencies with direct impact on the Safety Case and the Safety Assessment of each one of the concepts of interest.

The questionnaire will be discussed in the Second Annual Workshop of the CEBAMA project, to be held in Helsinki, May 2017, and will be worked throughout the third year to be finalised and filled in with input generated from CEBAMA and presented at the end of the project.