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Newsletter, Issue 4

CEMENT-BASED MATERIALS, PROPERTIES, EVOLUTION, BARRIER FUNCTIONS

May 2019

ebama is a research and innovation action granted by the European Commission under the Horizon 2020 Research and Training Programme of the European Atomic Energy Community (EURATOM). The project started in June 2015 and finishes in June 2019.

The overall objective of Cebama is to support the implementation of geological disposal by significantly improving the knowledge base on the use of cementitious materials for the Safety Case for European repository concepts. Scientific/technical research in Cebama is largely independent of specific disposal concepts and addresses different types of host rocks, as well as bentonite backfill. Cebama is not focusing on one specific cementitious material, but is studying a variety of important cementbased materials in order to provide insight on general processes and phenomena which can then be transferred to several different applications.



Details on the structural and formal issues of the project can be found in our website (www.cebama.eu).

Ce <mark>ba</mark> ma

Project website: www.cebama.eu

Newsletter submitting organization :

Amphos 21 c/ Veneçuela, 103, 2nd floor Eo8019 Barcelona (Spain)



27 beneficiaries consisting of Research Institutes, Universities and one SME from 9 EU members countries, Switzerland and Japan contribute to Cebama.



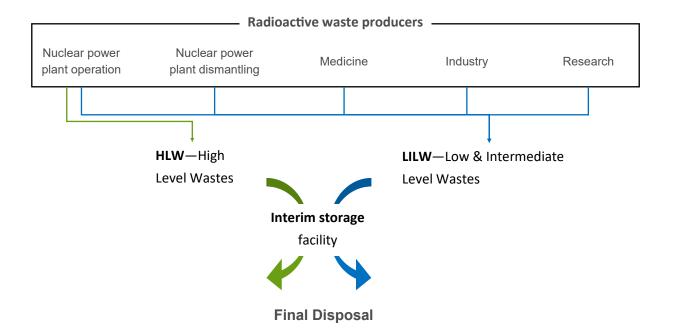
9 NWMO's support Cebama by codeveloping the work plan, participating in the **End-User Group (EUG)**, granting cofunding to some beneficiaries, and providing for knowledge and information transfer.



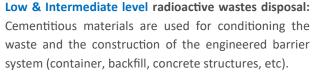
3 organizations joined the project as **Associated Groups (AG)**. AG are not receiving funding for participating but have interest on attending the Workshops and exchanging information related with Cebama.

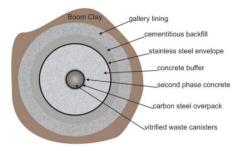


Why CEBAMA ?

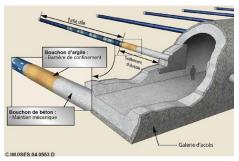


High level radioactive wastes disposal: Cementitious materials are mainly used for the construction of the engineered barrier system (backfill, plugs, liners, etc).

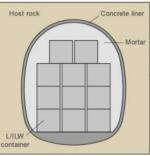




Belgian supercontainer concept for present high-level waste types (Cebama D1.03)



French concept of the HLW repository— Cigeo (Andra)



Swiss concept for a L/ILW repository (Cebama D1.03)



Spanish repository for LILW— El Cabril



CEBAMA objectives

To improve the knowledge base for the Safety Case for European repository concepts

"The safety case and supporting safety assessment provide the basis for demonstration of safety and for licensing of radioactive waste disposal facilities, and assist and guide decisions on siting, design and operations." (IAEA, 2012 - SSG-23)

Specific objectives have been defined to improve the knowledge on the **modelling** of transport properties (e.g. porosity, permeability, diffusion parameters) of **cement-based materials** in contact with the engineered and natural barriers of repositories in crystalline and argillaceous host rocks, as well as, the alteration that **radionuclide** retention processes could have on these materials.

CEMENTITIOUS MATERIALS:

- 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.
- To assess the impact on physical (transport) properties (e.g. porosity, water and gas transport properties).

WASTE MATERIALS:

- To study radionuclide retention processes in high pH concrete environments.
 - To analyse the retention of some specific radionuclides in high pH concrete environment, especially: Be, C, Cl, Ca, Se, Mo, I, Ra.
 - To assess the impact of chemical alterations (e.g., high pH concrete ageing, carbonation, transition from oxidizing to reducing conditions) on radionuclide retention.

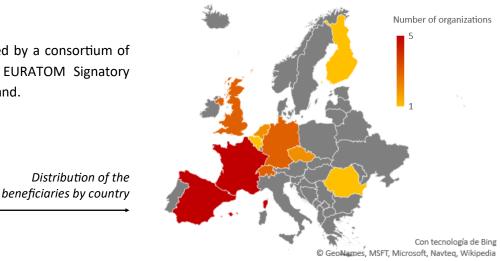
MODELLING:

• To improve validity of numerical models to predict changes in transport properties of cement-based materials and host rocks due to their geochemical interaction.

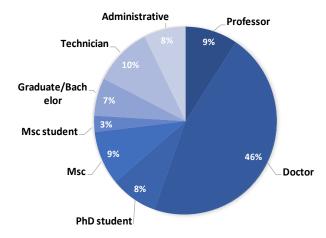
Further objectives cover **dissemination** of key results to scientific and non-scientific oriented stakeholders as well as training and education of young professionals for carrying over the expertise into future implementation programmes.

Who are we ?

The project is implemented by a consortium of 27 Beneficiaries, from 9 EURATOM Signatory States, Japan and Switzerland.



Additionally, 9 National Waste Management Organizations (NWMO) participate in the End-User Group (EUG) and 3 organizations joined the project as Associated Groups (AG).

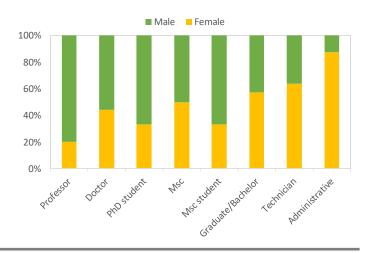


46% of the researchers involved in the project hold a PhD. Significant efforts were devoted to provide training to young scientists (11% of Msc and PhD students).

Professional profiles

Balanced gender distribution, with an overall 47-53% female and male, respectively.

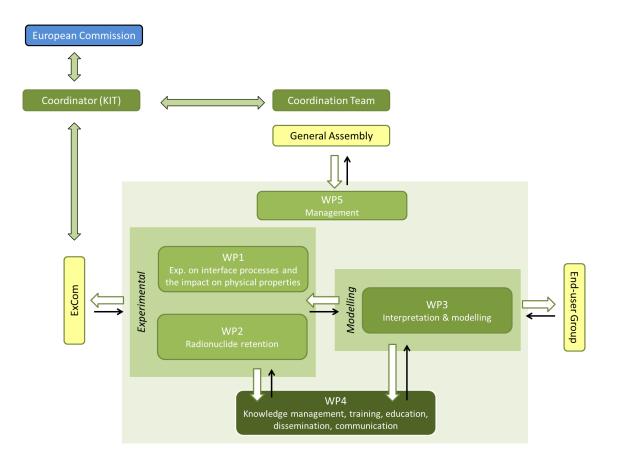
Gender information



How to reach CEBAMA goals?

The project is structured into 6 working groups (called work-packages, WP): three of them are devoted to carrying out scientific studies to fulfil the main project objectives. The forth is mainly focused on management and dissemination of the main outcomes of the project. WP5 is on management and a WP6 was defined after the start of CEBAMA on Ethics deliverables

All working groups interact with each other to build up an integrated approach to reach the main objective of this project, from the experimental design to the dissemination of the results obtained.



- Coordination Team: it is composed by 4 people which responsibility is to provide overall project implementation (e.g. communication with EC, technical activities, monitoring of economical resources, etc.).
- ExCom (Executive Committee): it consists of the Coordinator Team and the Work Package leaders (9 persons). They assesse the status and progress of each WP and monitor the effective and efficient implementation of the project, also in view of scientific quality.
- EUG (End-User Group): It is a group of 9 persons affiliated with European Waste Management Organizations set up to represent the interests of the end users to the project and its desired outcome.
- General Assembly: it consists of one member from each partner organization to the project and it has the ultimate responsibility regarding approval of management structure, project direction and Consortium Agreement.

Technical WP in detail

Each technical WP is focused on achieving specific objectives:

- WP1 is focused on investigating the interfaces between cementitious materials and natural host-rocks or engineered barrier components. It aims at quantifying the relevant alteration processes and their impact on physical properties, especially on the diffusive and advective transport properties for pore water and dissolved species.
- The objective of WP2 is to study radionuclide retention processes in high pH concrete environments. The
 aim is to provide insight on general processes and phenomena and their couplings in overall interaction
 mechanisms, which can then be transferred to different disposal situations and water access scenarios in a
 high pH repository environment with cementitious materials. It also assesses the impact of chemical
 alterations (e.g., high pH concrete ageing, carbonation, transition from oxidizing to reducing conditions) on
 radionuclide retention.
- The main goal of WP3 is to contribute in filling critical gaps by modelling and interpretation of
 experimental results generated within the project. The focus is mainly on physical and chemical processes
 that can lead to changes in transport properties both in the cementitious systems as well as their
 interfaces with clays or compacted bentonite.

The abovementioned objectives are achieved by the following **studies**:

- <u>WP1</u>: Preparation and characterization of cementitious samples and performance of different experiments devoted to study specific properties.
- <u>WP2:</u> Sorption experiments under high pH concrete environments are carried out considering various radionuclides (Be, C, Cl, Ca, Se, Mo, I, Ra, Tc) and relevant cement phases and alteration products. The following phenomena are covered: redox, reaction kinetics, changes in solubility and speciation, release mechanisms and solid solution formation.
- <u>WP3:</u> Development of reactive transport models for simulating the cement-clay interactions and the governing chemical, mechanical and hydrodynamics processes of the system. These models are calibrated with data obtained from experimental WPs. Different software are used by the group members and a benchmark exercise is performed to compare how each model works.

Different teams are collaborating and exchanging knowledge to investigate on the abovementioned objectives:

- <u>WP1:</u> 19 organizations; it is leaded by VTT (FI), BRGM (FR) and University of Bern (CH).
- <u>WP2:</u> 11 organizations; it is leaded by Armines/Subatech (FR).
- <u>WP3:</u> 13 organizations; it is leaded by Amphos 21 (ES).

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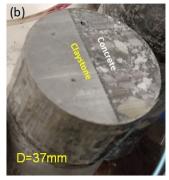
Preparation of samples



Preparation of the reference **concrete** (VTT)



Preparation of a **bentonite** suspension (UJV)



Interface between COx claystone and concrete (LML)



Concrete and **mortar** samples used for the experiments (CIEMAT)

Experimental set-up



Percolation experiment (SCK·CEN)



Experimental design (UAM)



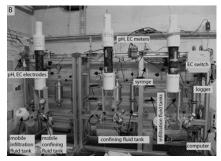
Punch test (UJV)



Experimental gallery of the concrete elements (ANDRA)



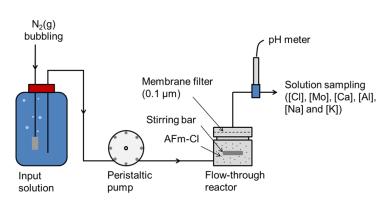
Analysis of tracer flow with GeoPET (HDZR)



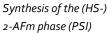
Infiltration devices (UNIBERN)

WP2 in images

Experimental set-up







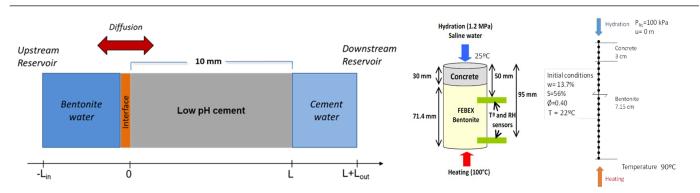


Synthesis of C3A, precursor of a AFm phase (A21)

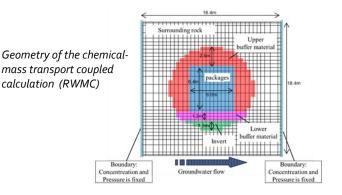
The experimental apparatus. Flow rates, pH as well as Cl, Mo, Ca, Al, Na and K concentrations were monitored as function of time (BRGM)

WP3 in images

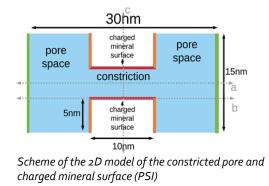
Modelled systems



Schematic representation of the through diffusion experiments (KIT)



Setup of the concrete-bentonite HB column tests (left) and scheme for the numerical model (right). (UdC)

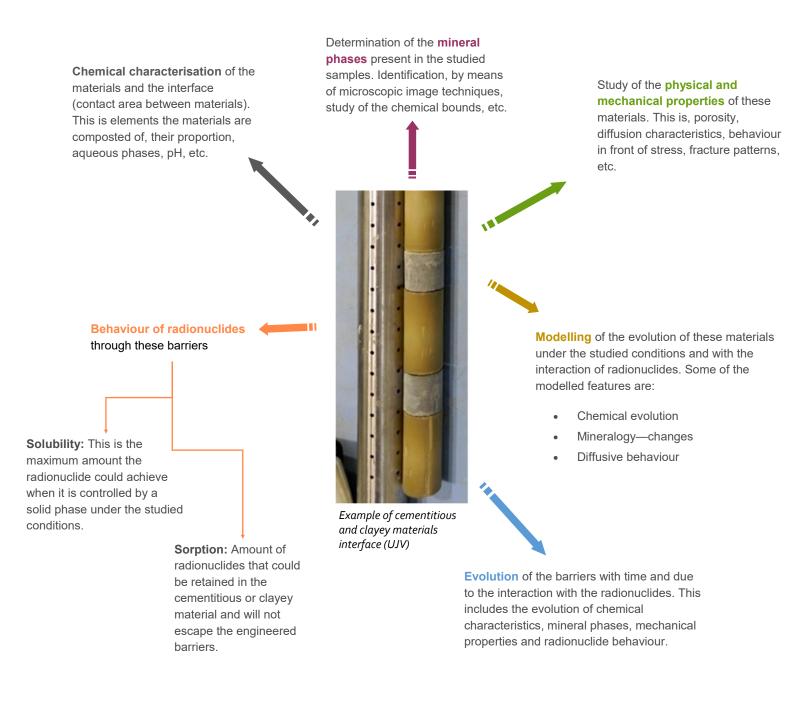


Note: All pictures are obtained from the 1st, 2nd and 3rd Annual Proceedings of CEBAMA

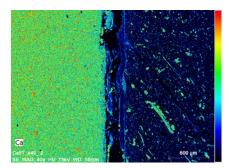
What did we find?

Cebama has generated a huge amount of data relevant on the **behaviour of cementitious and clayey materials** under repository conditions and provides information to improve the understanding on the **processes** that occurs at the interface between both type of materials.

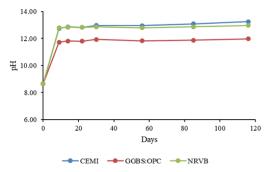
A summary of the type of data generated in this project is provided in the following scheme.



Chemical characterization

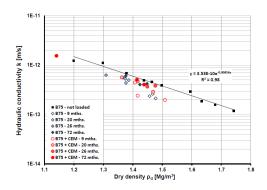


Map distribution of Ca - cement and argillite disks in contact (IRSN)

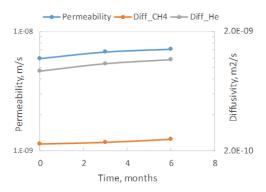


Changes in granitic GW pH due to interaction with cementitious materials (SURREY)

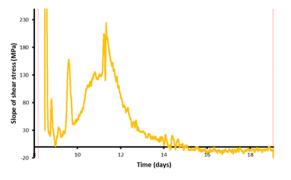
Physical and mechanical properties



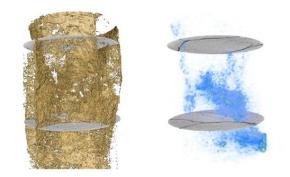
Hydraulic conductivity vs. bentonite dry density for bentonite and bentonite in contact with cement (UJV)



Evolution of transport properties of backfill materials during batch experiment (BRGM)

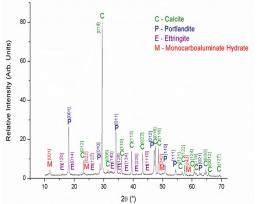


Slope of shear stress during shearing of the concrete / Cox interface (BGS)

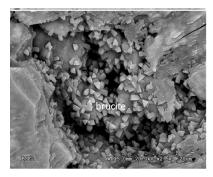


Fracture surface and tracer concentration - Brine labelled with ¹⁸F through a fractured halite drill core (HDZR)

Mineralogy

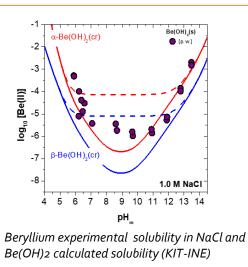


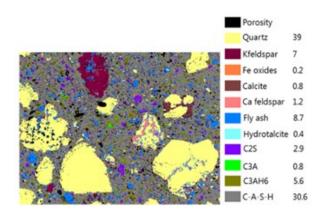
XRD pattern for a cementitious sample at 12 days of curing. Crystalline phases are labelled (USDF)



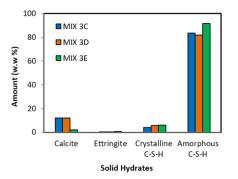
Identification of brucite in a concrete sample (UAM)

Radionuclide behaviour — Solubility

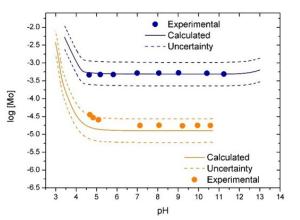




Quantitative mineralogical map for the concrete far from the interface with bentonite (SCK-CEN)

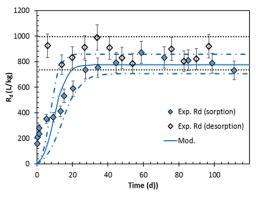


Crystalline and amorphous phase composition of three different low pH cement pastes (KIT-INE)

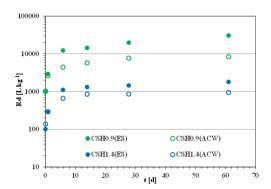


Molybdenum experimental solubility in NaClO4/cementitious water and Powellite calculated solubility (Amphos21)

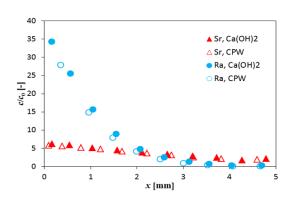
Radionuclide behaviour - Sorption



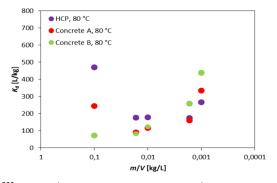
C-14 sorption onto non-carbonated hardened cement paste samples (Subatech/Armines)



²²⁶Radium uptake on CHS phases in two different solutions (JUELICH)

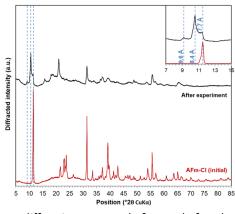


Strontium and radium sorption profile on cement samples in Portlandite or synthetic cement water (CTU)

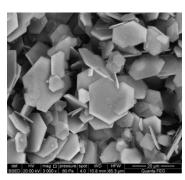


²²³Ra uptake on cementitious materials at 80°C (CTU)

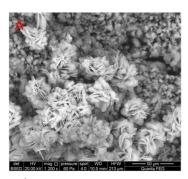




X-ray diffraction patterns before and after the experiment (BRGM)

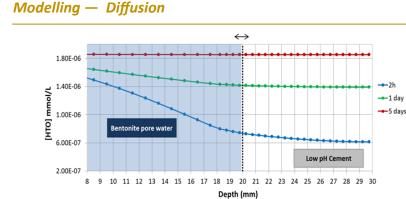


SEM picture of an AFm pase (JUELICH)

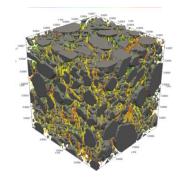


Selenium precipitation to CSH surface (SURREY)

-2h

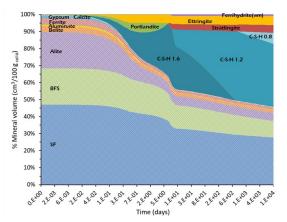


HTO diffusion in the low pH cement / bentonite interface (KIT-INE)



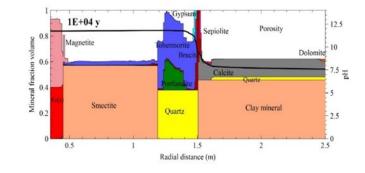
Diffusive flux streamlines of conservative tracer (JUELICH)

Modelling — Mineralogy

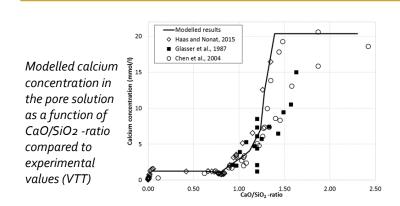


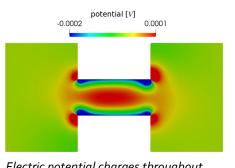
Evolution of main cement hydrates as a function of hydration time (Amphos21)

Modelling — Chemical parameters



pH and mineral volume fractions modelled from the canister surface (left) to the clay formation, across the bentonite barrier and the concrete liner (UdC)





Electric potential charges throughout the domain (PSI)

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Note: All pictures are obtained from the 1st, 2nd and 3rd Annual Proceedings of CEBAMA

Training of young scientists

The project promotes training of young professionals through training-on-the-job funding within the Cebama work. 5 researchers have been awarded the mobility measures for stay at a organization from the project consortium.

Jana Kittnerová (CVUT, Czech republic to JUELICH, Germany) - Period: January-April 2018

Synthesis, preparation and characterization (e.g. SEM, XRD) of calcium silicate hydrate (CSH) phases, the major hydration phases in cementitious materials. These materials were then used for experiments to get further insight into the uptake mechanisms of ²²⁶Ra, ¹³³Ba and ⁹⁰Sr in cementitious barriers. Besides these experiments we also dealt with the influence of carbonation on various cementitious materials and its consequences on the sorption properties for ²²⁶Ra.

Stephan Rohmen (JUELICH, Germany to PSI, Switzerland) - Period: February-March 2018

Discussions in pore-scale reactive transport modelling to improve the Lattice-Boltzmann based reactive transport code developed within CEBAMA called iPP. A benchmark activity to compare the results of iPP against another reactive transport code (Yantra) has been started. Application of iPP to simulate experiments performed within CEBAMA WP1, providing for an enhanced understanding of the coupling between chemical alteration processes in cementitious materials and changes in its physical properties.





Aku Itälä (VTT, Finland to AMPHOS21, Spain) - Period: February 2018

Discussions related to the modelling of the VTT experiments and hydration of reference cements. VTT's CSH model was applied to the hydration model of AMPHOS21 and tested the workability of the model. More discussions of the usability of the model of VTT for low pH values (below 0.8), the role of different ion activity models, the use of different databases and the evolution of relevant mineral phases took also place.

Enrique Rodríguez Cañas (UAM, Spain to BRGM, France) - Period: March-April 2018

Determination of the mineralogy and porosity of a 13 years in situ concrete/ bentonite contact by image processing of quantitative elemental mapping and ¹⁴C MethylMethAcrylate (MMA) impregnated samples. Imaging results were possible to compute mineral/phase maps based on procedures of chemical segmentation using ternary scatter plot projections to detect the mineralogical evolution at microscale related to the geochemical perturbation of the complex mineralogy at the bentoniteconcrete.



Training of young scientists



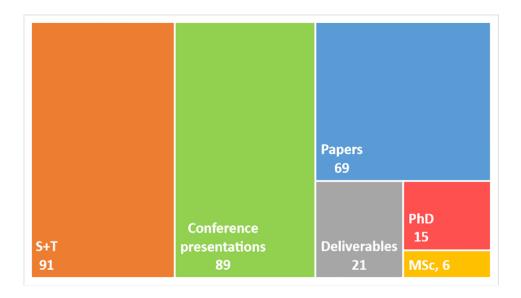
Marta López García (AMPHOS21, Spain to JUELICH, Germany) - Period: May-June 2018

Study of the adsorption of Mo onto specific aluminate phases representative for hydration products in cementitious materials, namely AFm and AFt and mixtures thereof, containing different anions (i.e. carbonate and sulfate). Characterization of synthesized solid phases by various spectroscopic and microscopic techniques including XRD, SEM-EDX, FTIR, and RAMAN as well as by TG-DSC.

Project dissemination & Training activities

Significant efforts were also devoted by the individual project participants to disseminate the technical information and knowledge generated in the frame of this project. **More than 60 articles** have been published, submitted or are in preparation for their submission in several peer review scientific journals. Additionally, researchers have attended to different international conferences and workshops to present their work by either oral talks or in poster sessions. CEBAMA has produced about **90 presentations (oral talks and posters)** at conferences and meetings, highlighting the results of individual or joint contributions between partners in CEBAMA. Additional presentations were given by the members of the Coordination Team on the overall CEBAMA project.

Additionally, it is important to highlight that 15 PhD students performed their thesis in the frame of this project. All of them had the opportunity of presenting their work in all the project workshops.



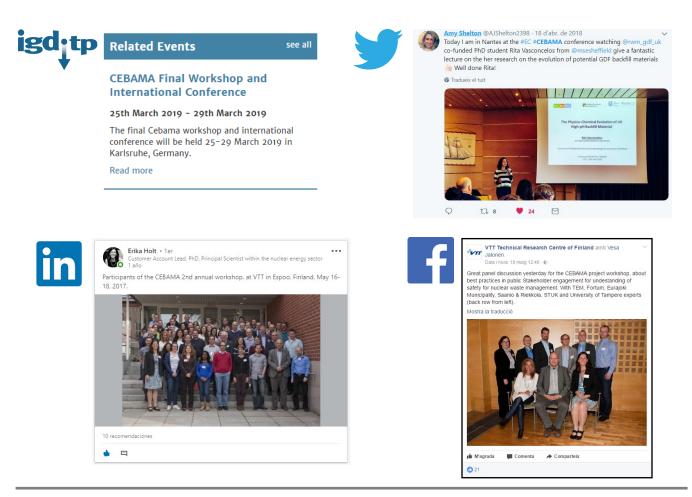
More information of Cebama

Detailed information on the project can be found at the project website. Reports produced are available at the deliverables section of the website)



Deliverables
Public deliverables of the project
WP1: Experiments on interface processes and the impact on physical properties
D1.01 Detailed description of scientific work
D1.02 Agreement and documentation of systems to be studied D1.03 State of the art report related with WP1 topics
 D1.05 State of the art report related with WP1 topics D1.04 Report on WP1 established experimental boundary conditions, experimental methods
 D1.05 Report on WP1 selected experimental materials to be used, including both new laboratory and aged in-situ samples
WP2: Radionuclide retention in high pH concrete
D2.01 Detailed WP2 description of scientific work
 D2.02 Agreement and documentation of systems to be studied D2.03 State-of- art report related to WP2 topics (initial)
D2.03 State-or- art report related to WH2 topics (initial)
WP3: Interpretation & modelling
 D3.01 Detailed description of scientific work D3.02 Review and definition of modelling approach to be followed in the project: scale of analysis, physico-chemical processes,
software, HPC resources
 D3.03 Consolidated plan for Upscaling Modelling Task D3.04 Description of and results from the modelling of external lab and/or field experiments
WP4: Knowledge Management, Training, Education, Dissemination,
Communication
D4 01 Generic poster presenting the project
D4.01 Generic poster presenting the project D4.02 Set up of project webpage
D4.04 Planning on interaction with socio-political stakeholders D4.05 Communication action plan
D4.05 Communication action plan D4.06 Planned dissemination of final results
D4.08 Draft of the 1st Annual Project Workshop Proceeding
D4.09 Basis for application to Safety Case and Performance Assessment

Additionally, information on specific events or progresses are published in several platforms and formats. Some examples below:

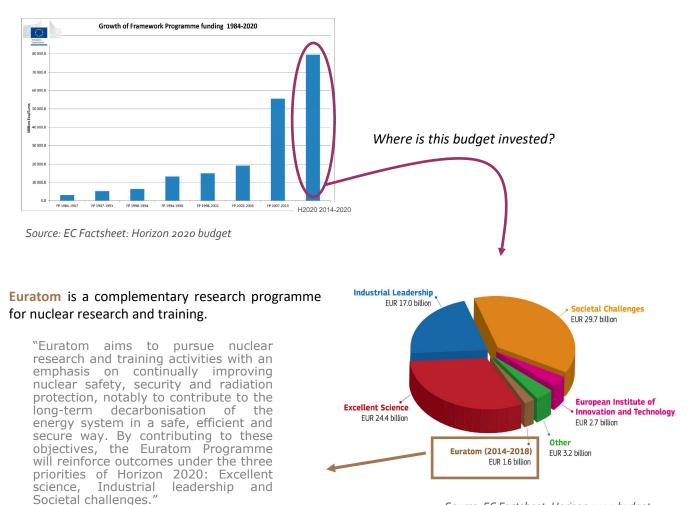


Acknowledgements

The research leading to these results has received funding from the European Union's Horizon 2020 Research and Training Programme of the European Atomic Energy Community (EURATOM) (H2020-NFRP-2014/2015) under grant agreement n° 662147 (CEBAMA).

EC Funding

Horizon 2020 is a research an innovation programme with about \in 80 billion of funding available over 7 years (2014 to 2020).



Source: EC Factsheet: Horizon 2020 budget

The programme is focused in two areas:

(European Commission)

- \Rightarrow Nuclear fission and radiation protection
- \Rightarrow Fusion research

