

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 662147.

# CEMENT-BASED MATERIALS, PROPERTIES, EVOLUTION, BARRIER FUNCTIONS

Newsletter, Issue 4 May 2019

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**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 cement-based 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).



**Project website:** www.cebama.eu

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



# Newsletter submitting organization :

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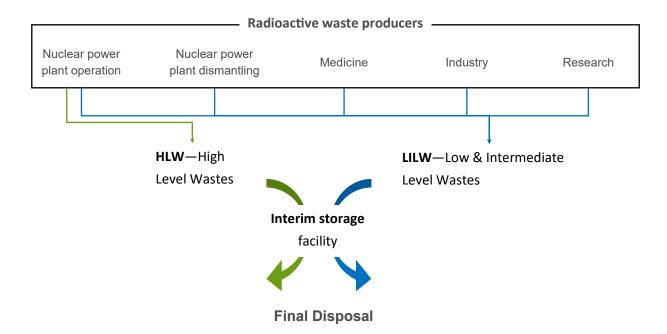
**9** NWMO's support Cebama by codeveloping the work plan, participating in the **End-User Group (EUG)**, granting cofunding to some beneficiaries, and providing for knowledge and information transfer.



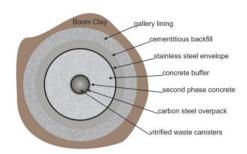
**3** organizations joined the project as **Associated Groups (AG)**. AG are not receiving funding for participating but have interest on attending the Workshops and exchanging information related with Cebama.



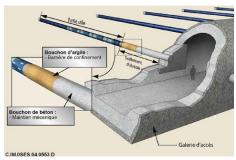
## Why CEBAMA?



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

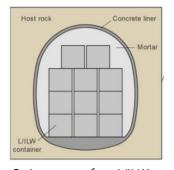


Belgian supercontainer concept for present high-level waste types (Cebama D1.03)



French concept of the HLW repository— Cigeo (Andra)

#### Low & Intermediate level radioactive wastes disposal: Cementitious materials are used for conditioning the waste and the construction of the engineered barrier system (container, backfill, concrete structures, etc).



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:**

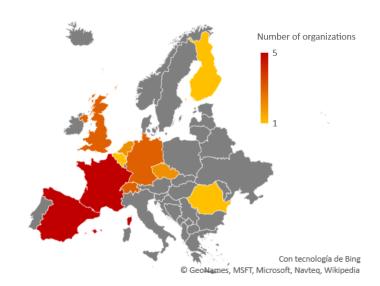
• To improve validity of numerical models to predict changes in transport properties of cement-based materials and host rocks due to their geochemical interaction.

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.

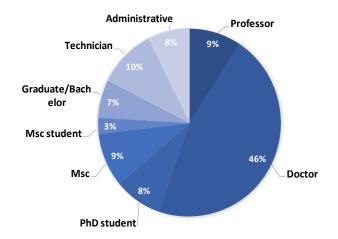
## Who are we?

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

Distribution of the beneficiaries by country



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

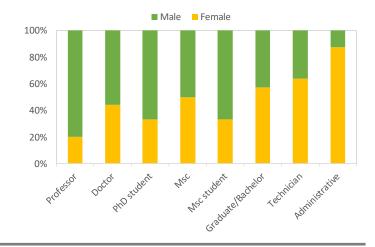


46% of the researchers involved in the project hold a PhD. Significant efforts were devoted to provide training to young scientists (11% of Msc and PhD students).

Professional profiles

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

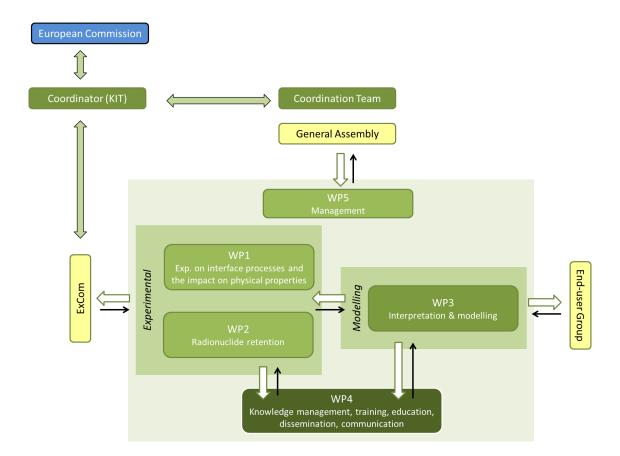
Gender information



## 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:

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

# WP1 in images

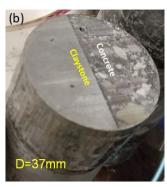
#### **Preparation of samples**



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)



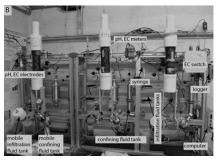
Punch test (CTU)



Experimental gallery of the concrete elements (ANDRA)



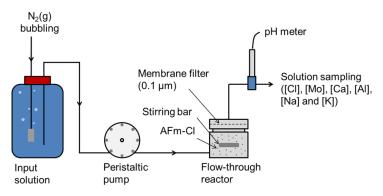
Analysis of tracer flow with GeoPET (HDZR)



Infiltration devices (UNIBERN)

## WP2 in images

#### **Experimental set-up**



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



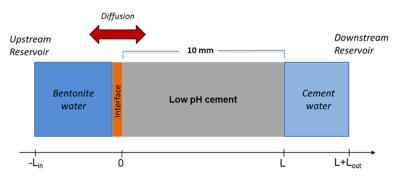
Synthesis of the (HS-) 2-AFm phase (PSI)



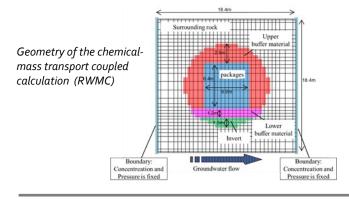
Synthesis of C<sub>3</sub>A, precursor of a AFm phase (A<sub>21</sub>)

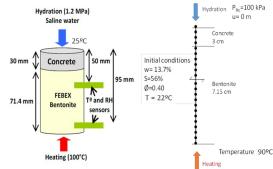
## WP3 in images

#### **Modelled systems**

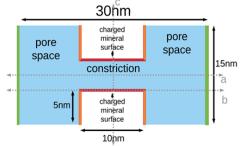


Schematic representation of the through diffusion experiments (KIT)





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



Scheme of the 2D model of the constricted pore and charged mineral surface (PSI)

## 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 characterisation of the materials and the interface (contact area between materials). This is elements the materials are composted of, their proportion, aqueous phases, pH, etc.

phases present in the studied samples. Identification, by means of microscopic image techniques, study of the chemical bounds, etc.



Study of the **physical and mechanical properties** of these materials. This is, porosity, diffusion characteristics, behaviour in front of stress, fracture patterns, etc.

Behaviour of radionuclides through these barriers

Solubility: This is the maximum amount the radionuclide could achieve when it is controlled by a solid phase under the studied conditions.

Sorption: Amount of radionuclides that could be retained in the cementitious or clayey material and will not escape the engineered barriers.



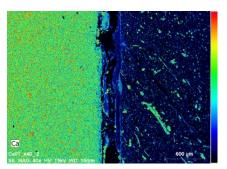
Example of cementitious and clayey materials interface (CTU)

**Modelling** of the evolution of these materials under the studied conditions and with the interaction of radionuclides. Some of the modelled features are:

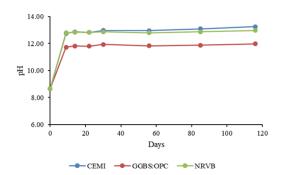
- Chemical evolution
- Mineralogy—changes
- Diffusive behaviour

**Evolution** of the barriers with time and due to the interaction with the radionuclides. This includes the evolution of chemical characteristics, mineral phases, mechanical properties and radionuclide behaviour.

#### Chemical characterization

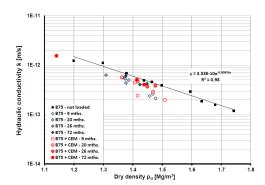


Map distribution of Ca - cement and argillite disks in contact (IRSN)

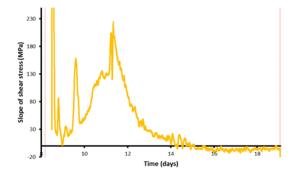


Changes in granitic GW pH due to interaction with cementitious materials (SURREY)

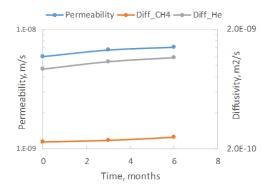
#### Physical and mechanical properties



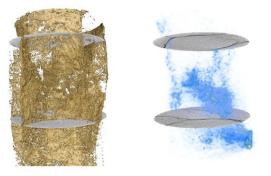
Hydraulic conductivity vs. bentonite dry density for bentonite and bentonite in contact with cement (UJV)



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

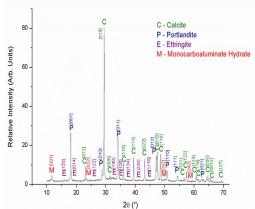


Evolution of transport properties of backfill materials during batch experiment (BRGM)

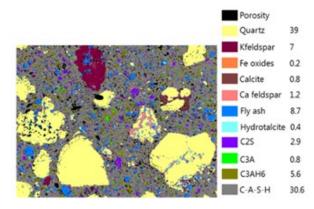


Fracture surface and tracer concentration - Brine labelled with <sup>18</sup>F through a fractured halite drill core (HDZR)

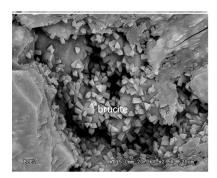
#### Mineralogy



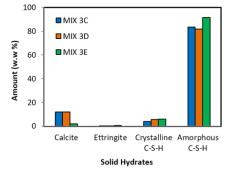
XRD pattern for a cementitious sample at 12 days of curing. Crystalline phases are labelled (USDF)



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

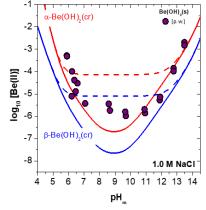


Identification of brucite in a concrete sample (UAM)

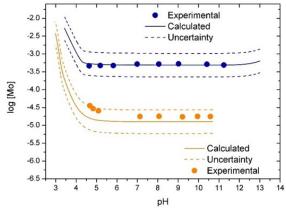


Crystalline and amorphous phase composition of three different low pH cement pastes (KIT-INE)

#### Radionuclide behaviour — Solubility

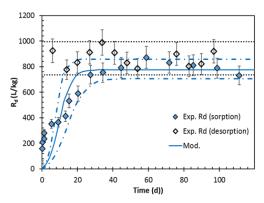


Beryllium experimental solubility in NaCl and Be(OH)2 calculated solubility (KIT-INE)

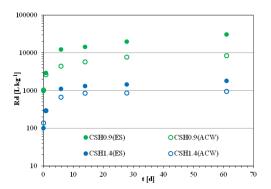


Molybdenum experimental solubility in NaClO4/cementitious water and Powellite calculated solubility (Amphos21)

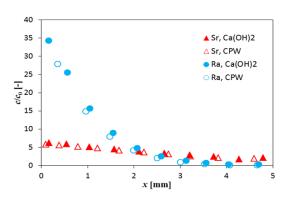
#### Radionuclide behaviour — Sorption



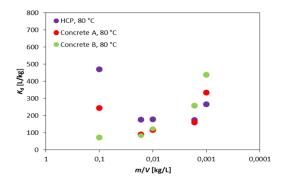
C-14 sorption onto non-carbonated hardened cement paste samples (Subatech/Armines)



<sup>226</sup>Radium uptake on CHS phases in two different solutions (JUELICH)

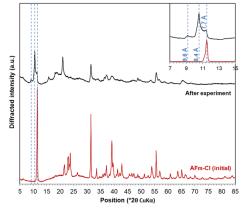


Strontium and radium sorption profile on cement samples in Portlandite or synthetic cement water (CTU)

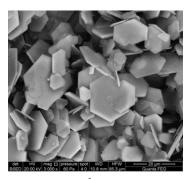


<sup>223</sup>Ra uptake on cementitious materials at 80°C (CTU)

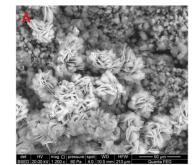
#### **Solid phase evolution** due to radionuclide interaction



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

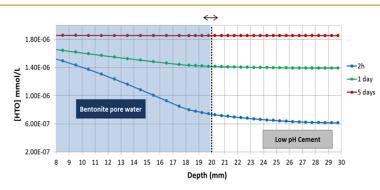


SEM picture of an AFm pase (JUELICH)

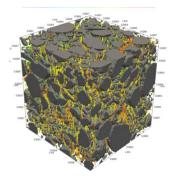


Selenium precipitation to CSH surface (SURREY)

#### Modelling — Diffusion

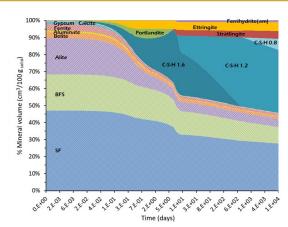


HTO diffusion in the low pH cement / bentonite interface (KIT-INE)

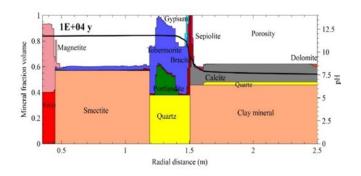


Diffusive flux streamlines of conservative tracer (JUELICH)

#### Modelling — Mineralogy

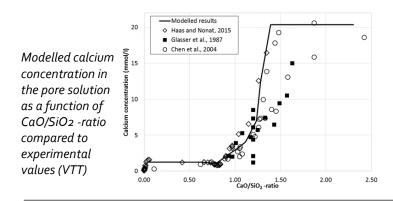


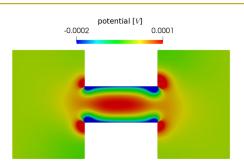
Evolution of main cement hydrates as a function of hydration time (Amphos21)



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

#### **Modelling** — Chemical parameters





Electric potential charges throughout the domain (PSI)

## **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 <sup>226</sup>Ra, <sup>133</sup>Ba and <sup>90</sup>Sr 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 <sup>226</sup>Ra.

#### 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 <sup>14</sup>C 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 bentonite-concrete.



## **Training of young scientists**



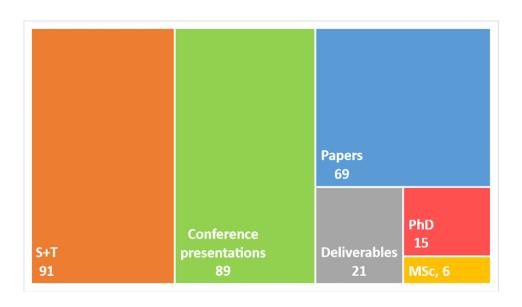
Marta López García (AMPHOS21, Spain to JUELICH, Germany) - Period: May-June 2018

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.

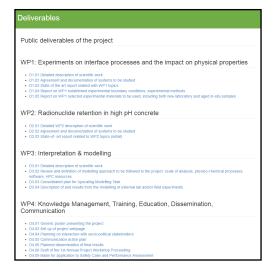
Additionally, it is important to highlight that 15 PhD students performed their thesis in the frame of this project. All of them had the opportunity of presenting their work in all the project workshops.



## **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)

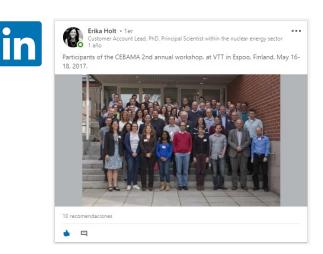




Additionally, information on specific events or progresses are published in several platforms and formats. Some examples below:









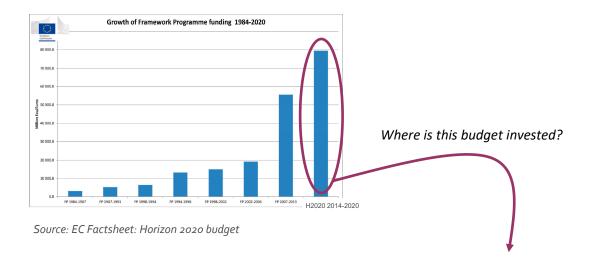


## **Acknowledgements**

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**

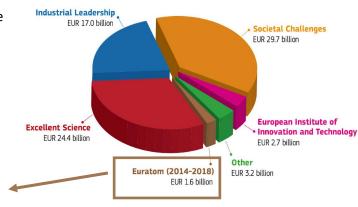
Horizon 2020 is a research an innovation programme with about €80 billion of funding available over 7 years (2014 to 2020).



# **Euratom** is a complementary research programme for nuclear research and training.

"Euratom aims to pursue nuclear research and training activities with an emphasis on continually improving nuclear safety, security and radiation protection, notably to contribute to the long-term decarbonisation of the energy system in a safe, efficient and secure way. By contributing to these objectives, the Euratom Programme will reinforce outcomes under the three priorities of Horizon 2020: Excellent science, Industrial leadership and Societal challenges."

(European Commission)



Source: EC Factsheet: Horizon 2020 budget

The programme is focused in two areas:

- ⇒ Nuclear fission and radiation protection
- ⇒ Fusion research

