The role of excellent science in geological disposal system optimization – view and perspectives from EuradScience

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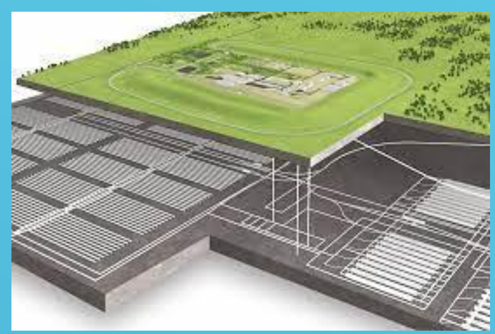


https://www.gov.uk/guidance/gdf-geological-disposal-facility

Optimisation is a **central element** in the **stepwise** design, construction and operation of a geological disposal facility.

According to the International Commission on Radiological Protection (ICRP 122, 2013), optimization needs to be understood in the **broadest sense**, *"as an iterative, systematic and transparent evaluation of options (…) for enhancing the protective capabilities of the system and for reducing impacts (radiological and others)"* .

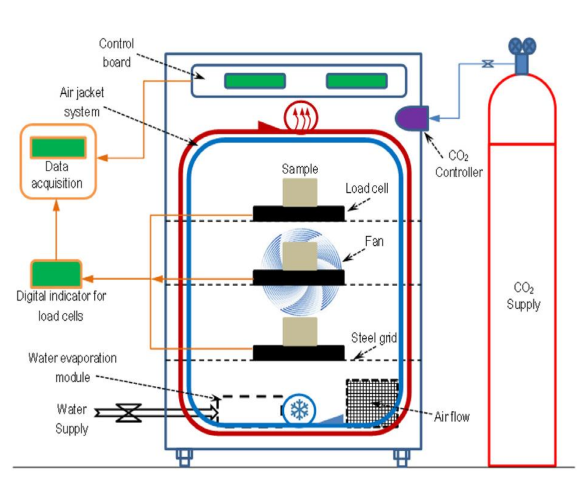
This poster presents several **case studies** illustrating how **scientific or technical advancements** or breakthroughs can assist in the **iterative evaluation of optimization, reducing conservatisms and elucidating uncertainties**, or even fundamentally affecting disposal system design and associated safety functions. The case studies are based on recent (national and international) studies and projects, highlighting the continued need to support scientific advancements in the context of waste management and disposal.

Advancing science for a better assessment of long-term durability and performance of cementitious materials for radioactive waste conditioning and disposal

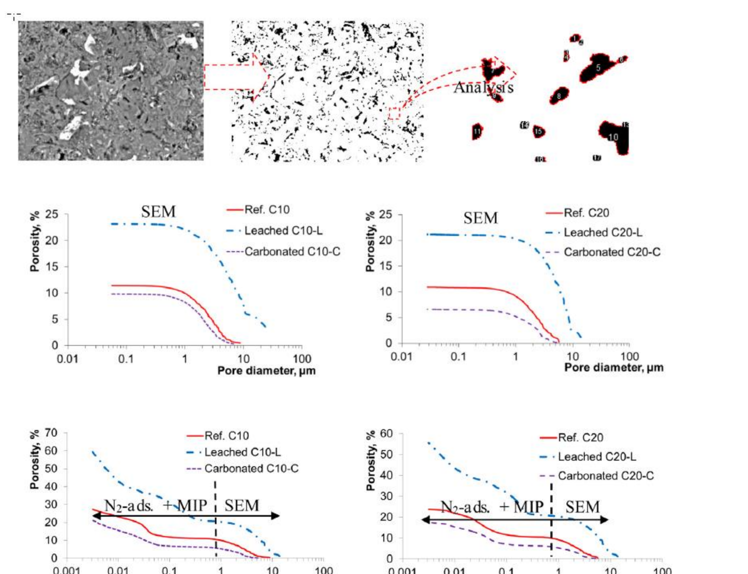
Context: **Cementitious materials** are omnipresent in many radioactive waste disposal facility concepts as conditioning matrix for radioactive waste and/or as engineered barriers. **Safety functions are assigned to cementitious SSCs** (system, structures and components) both during operational activities in the predisposal and operational phase and post-closure phase. Cementitious materials pose important challenges for the scientific-technical community because of their **multi-scale nature** and the strongly coupled **multi-physics** behavior.

Problem statement: How to **accurately predict the evolution** of cementitious materials in a disposal environment and **assess the consequences of this evolution on their performance** over extreme long time scales, ranging from a few hundreds to several tens of thousands of years?

Results (i): well-controlled conditions in a **laboratory set-up** allow for in-depth understanding of the consequences of ageing processes, by studying the complementarity of well-designed accelerated degradation set-ups, microstructural characterization, and determination of hydraulic and transport properties for assessing degradation rates.

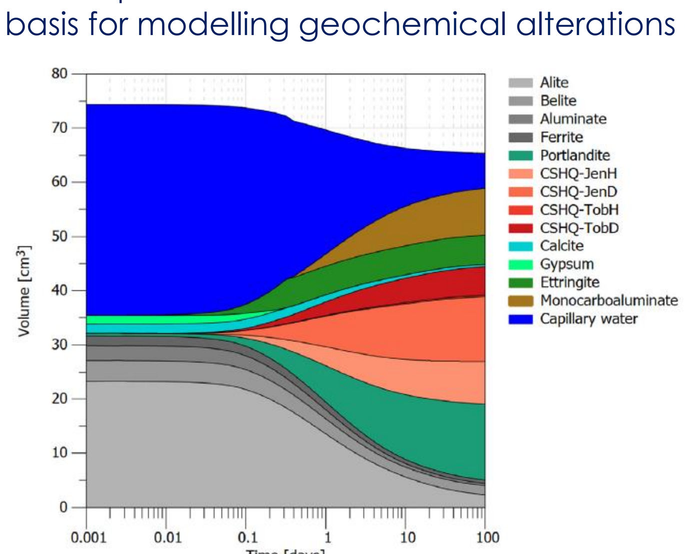


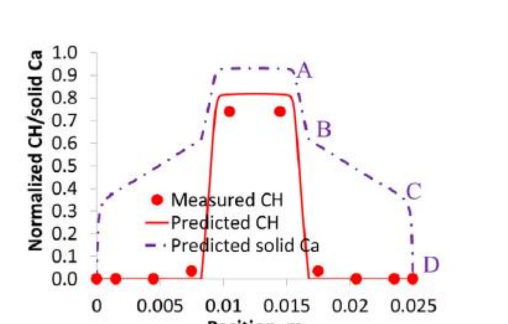
Temperature, relative humidity and CO_2 controlled climate chamber allowing to automatically log experimental variables such as sample mass changes via a digital acquisition system



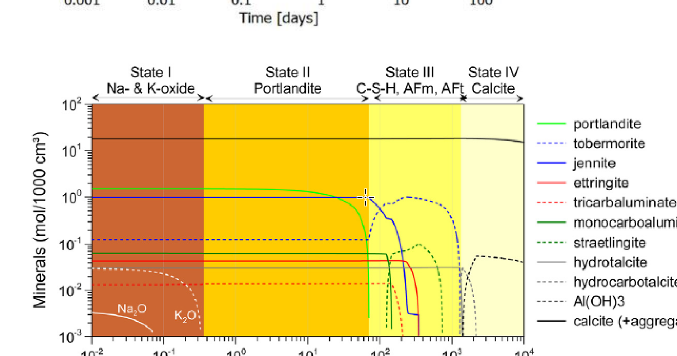
(Top) Segementation and pore identification from SEM (Middle) Average cumulative porosity derived from SEM (Bottom) Combining N_2-adsorption, MIP and SEM

Results (ii): Key element in **modelling** ageing of cement-based materials is the representation of its multi-scale heterogeneity. This can be captured by different models each focusing on different aspects of this multi-scale nature. Thermodynamic calculations of the cement matrix is the basis for modelling geochemical alterations at the continuum, meso and pore scale.

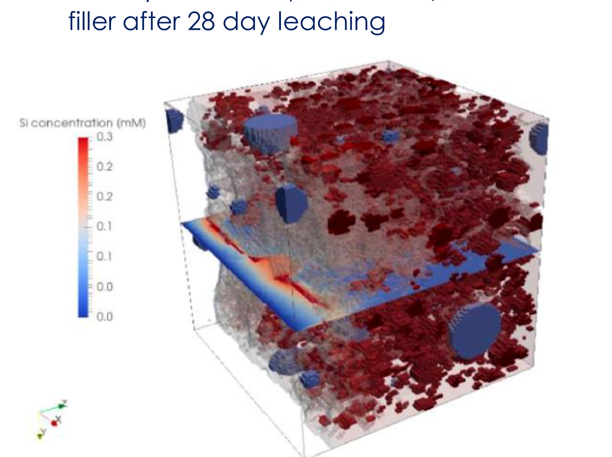




Evolution of Ca in solid phases and portlandite content along depth of sample with w/c ratio of 0,375 and 10% limestone filler after 28 day leaching

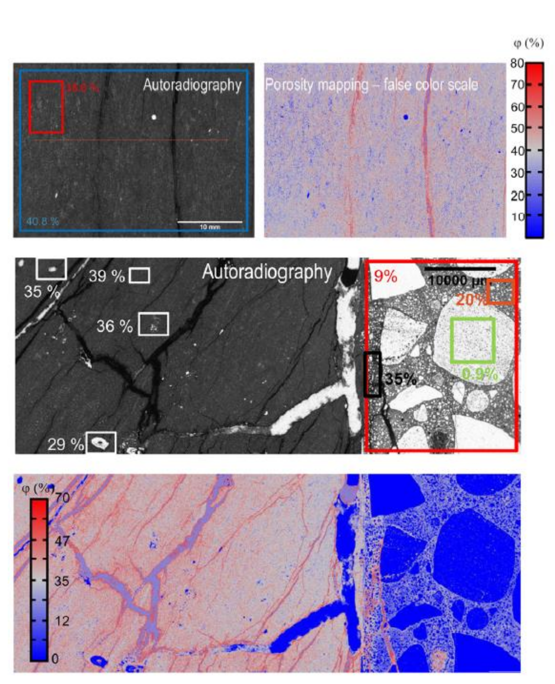


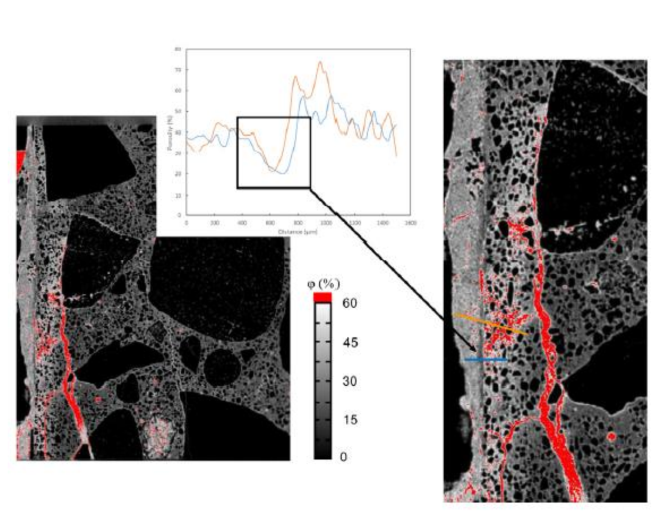
(Top) Cement phase composition during hydration of 100 g of CEM-1 OPC clinkers
(Bottom) Cement phase evolution during calcium leaching with rain water. Calculations are performed using CEMDATA 18 database, in HPGeochemistry (PHREEQC-version embedded in HPx)



Partially leached microstructure of hardened cement paste. The colours represent different phases of hardened cement paste.

Results (iii): The modelling techniques discussed above can then be **validated** in the context of ageing cement-based materials under disposal conditions,against samples taken from **analogues** of a real repository (such as existing underground research facilities).





(Left) Porosity maps of Boom Clay obtained by autoradiography (Top) and spatial distribution of porosity at concrete, clay sides and the interface (Middle) and porosity (bottom) mapping of concrete and clay after 14 year interaction at in-situ conditions
(Right) Evidence of concrete porosity decrease at clay interface after 14 year interaction with Boom Clay
(All measurements by BRGM)

Conclusion: **Scientific and technological advances** have led to a **better understanding** and a more accurate prediction, leading to a **decrease of uncertainties in the safety case and the performance assessment** of radioactive waste repositories. An approach based on synergy between (i) experimental studies, (ii) multi-scale modelling and (iii) coupled reactive transport models proved to be necessary and a pre-requisite to better assess the long-term performance of cementitious materials under repository conditions.

Outlook: insights obtained during these studies also led to important new developments, for example by using novel binder systems as alternative immobilization matrices → the intensive development and study of **alkali-activated materials (AAMs) as (more sustainable) alternatives to OPC**, among others for stabilizing and solidifying radioactive waste.

Ref: Jacques *et al.*, (2021) "Towards a scientific-based assessment of long-term durability and performance of cementitious materials for radioactive waste conditioning and disposal." Journal of Nuclear Materials, 557, 153201
Duro *et al.* (2020) "Contribution of the results of the CEBAMA project to decrease uncertainties in the Safety Case and Performance Assessment of radioactive waste repositories." Applied Geochemistry, 112, 104479

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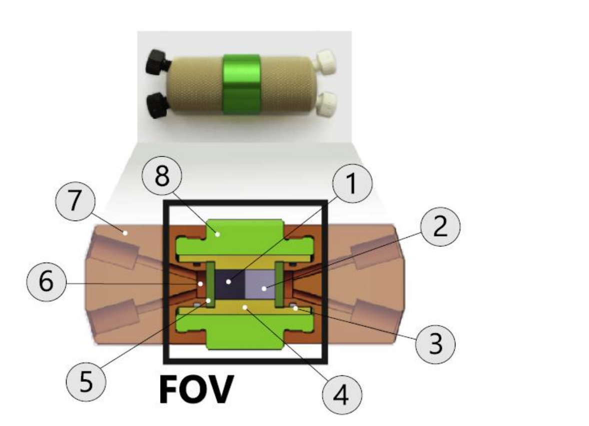
New scientific approaches to studying interface processes between cement and clays under geological repository conditions

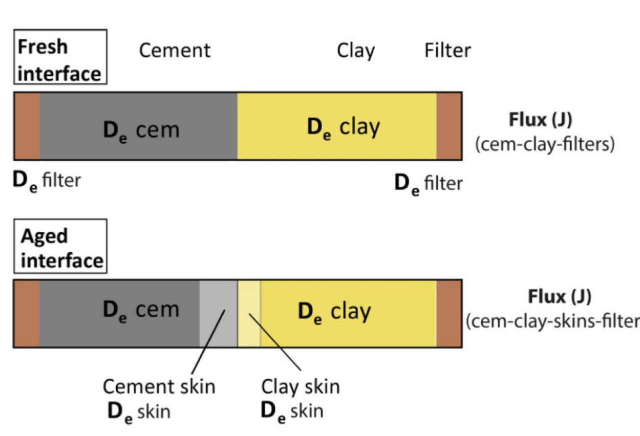
Context: Cement and clays are proposed as engineered barrier materials in underground repositories for radioactive waste. When cement and clay come into contact, chemical gradients between their very different pore water compositions lead to diffusive exchange of solutes, which can result in mineral transformations and alterations of transport properties at the materials interface. Lack of accurate descriptions of the **interface processes between two contrasting materials**, and difficulties in assessing the **impact of these processes on their performance**, frequently lead to **conservative approaches** during Performance Assessment.

Problem statement: How to **better bound** the evolution of the **interface** between clay/concrete and its impact on the transport and mechanical properties of both materials, to allow for **reduction of conservatism** in Performance Assessment.

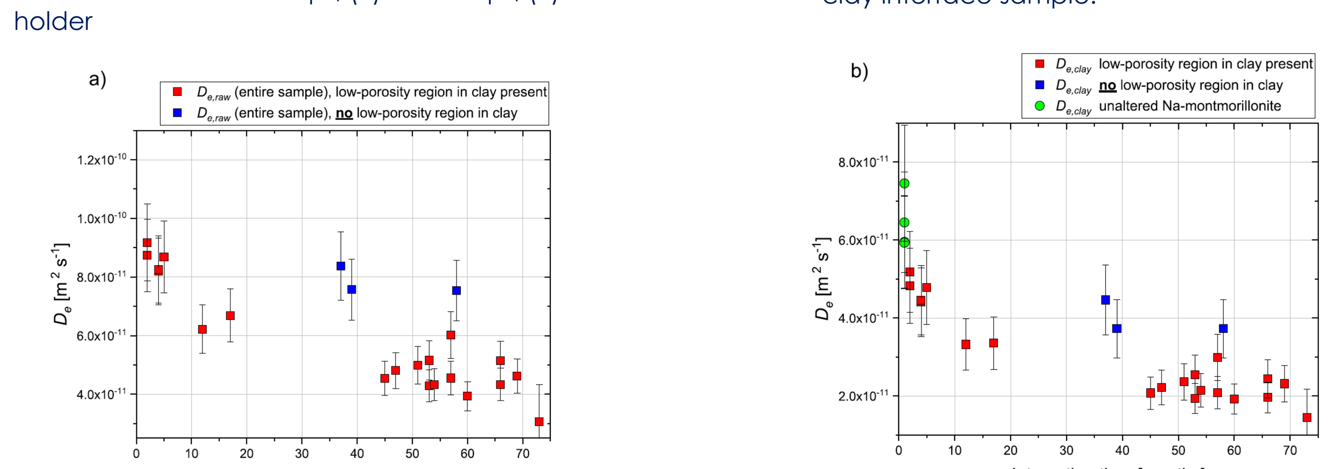
Results (i): **Novel reaction cells** containing cement-clay interfaces were prepared and let react over a period of six years. During this time, the changes in transport properties of the samples were periodically monitored by means of through-diffusion experiments and neutron radiography. The experiments allowed obtaining relevant information regarding the development of the diffusive properties and the reactivity of the interface system and the evolution of water filled porosity in cement and clay compartments. HTO and $^{36}Cl^-$ were used as tracers to study the evolution of both the total and the anion accessible porosity.



Reaction cell: (1) Cement sample, (2) clay sample, (3) O-rings, (4) PTFE sample holder, (5) PEEK frits, (6) solution reservoirs within PEEK caps, (7) PEEK caps, (8) aluminum holder

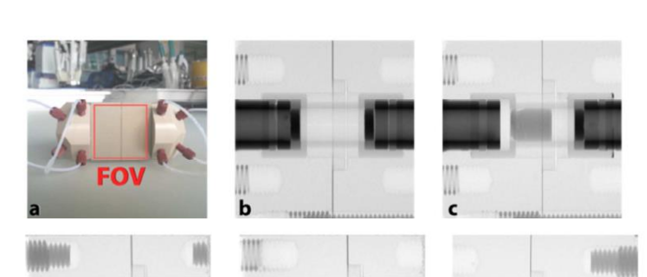


Schematic view of a fresh and an aged cement-clay interface sample.

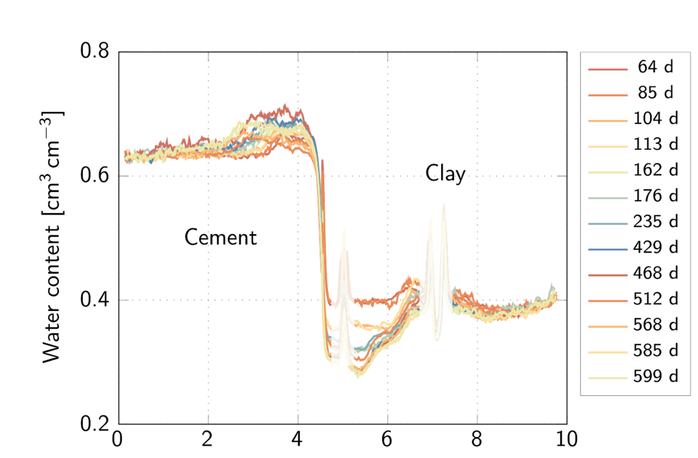


Evolution of the average diffusion coefficient (De) of HTO for (a) the entire system (De,raw) composed of filters, cement and clay); and (b) for the clay part only (De,clay).).

Results (ii): **Novel non-destructive analysis** using neutron radiography of several evolving cement-clay interfaces delivered quantitative data which allowed resolving local water contents within the sample domain. An increase in cement porosity was observed propagating ~2mm away from the interface within 600 days, and a considerable decrease in clay porosity within ~2 mm. The experiments provided unique information on the dynamics of the processes up to nearly two years and allowed for better constraining of reactive transport models.

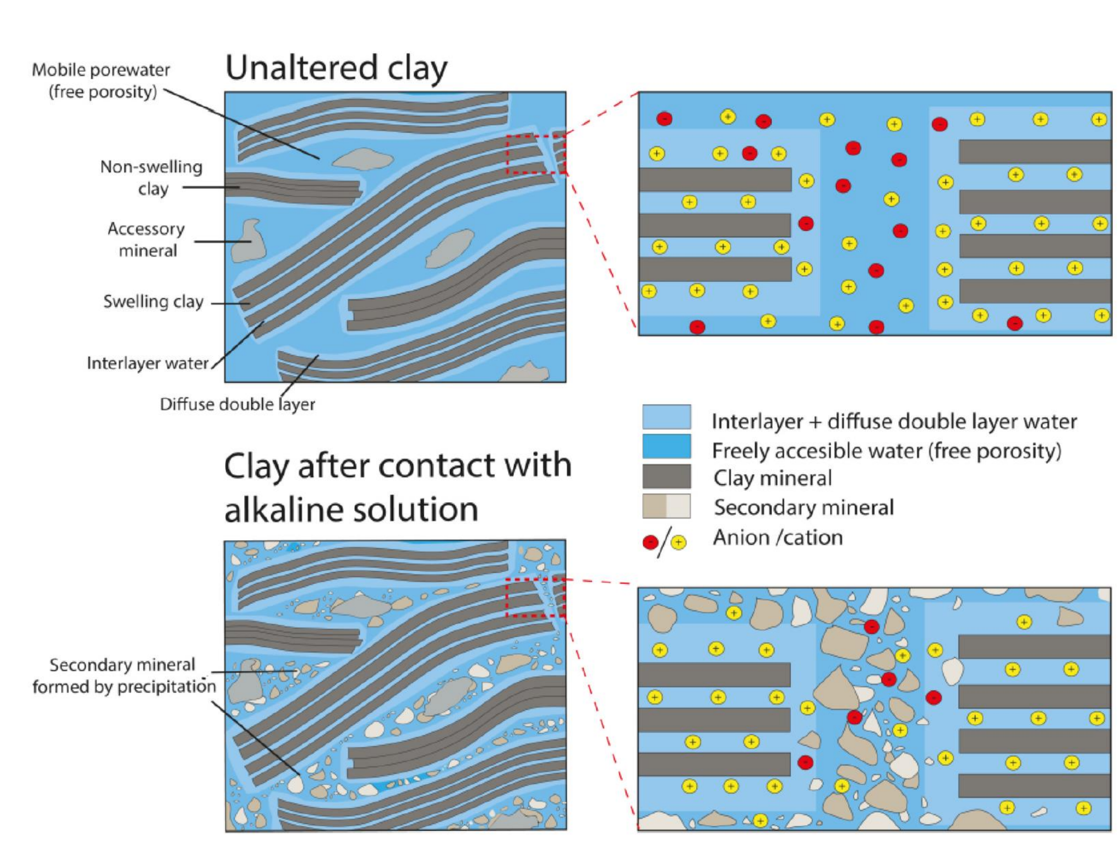


Quantification of water content in the sample by neutron radiography. Sample cell with the neutron field of view (FOV) shown as red square (a); transmission neutron radiographs of an empty cell
(b), a dry cement on the left (c), a dry clay on the right (d), a cement-clay interface at time d = 64 days (e) and the same interface at time d = 104 days (f).



Evolution of profiles of water content with reaction time (in days) across interface. The regions blanked out in the figure are artefacts originating from the joints of the aluminum holder of the cell

Conclusion: **Novel experimental** diffusive flux **set-ups** combined with **innovative non-destructive analysis** allowed to conclude that the changes of transport properties in the cement/clay sample are dominated by a narrow low porosity/low diffusivity alteration zone in the clay compartment close to the interface. The results can be used to **define bounding scenarios** allowing for a better and more accurate assessment of clay/cement interface evolution and its effect on transport properties.



Schematic illustration of clay minerals and respective pore environments for unaltered clay, and for clay after contact with alkaline solution (e.g. cement porewater). Precipitation location (free porosity only or interlayer/diffuse double layer) and precipitation type.

Ref: Luraschi, P., et al. (2020). "Evolution of HTO and Cl-36(-) diffusion through a reacting cement-clay interface (OPC paste-Na montmorillonite) over a time of six years." Applied Geochemistry 119.
Shafizadeh, A., et al. (2015). Quantification of water content across a cement-clay interface using high resolution neutron radiography Physics Procedia 69 (2015) 516 – 523
Shafizadeh, A., et al. (2020). "Time-resolved porosity changes at cement-clay interfaces derived from neutron imaging." Cement and Concrete Research 127.

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