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Insight of the reactive transport models developed to understand the corrosion processes of high-level nuclear waste

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Processes occurring during weathering of the rocks, diagenesis of sedimentary deposits or radioactive waste containment take place over a variety of time and space scales. To circumvent such scaling issues that are usually beyond measurements, one approach consists of acquiring data in the laboratory and/or in the field and analysing the results with reactive transport models to test our understanding of processes.

In France, the concept of high-level waste (HLW) disposal is based on a multi-barrier system involving the host rock (Callovo-Oxfordian claystone level of the Bure site - COx), a cement-grout filling material (CGF), the carbon steel liner and overpack and finally the glass containing the radionuclides poured in a stainless steel container. When the glass is exposed to the water of the site, the various barriers will have already reacted with this water and modified them (ageing, neoformation of corrosion products (CP) and other secondary phases, etc.). Therefore, it is of paramount importance to study and quantify the interactions between "aged" and “unages” environmental materials (COx/CGF/steel-PC) and SON68 glass, an inactive surrogate for R7T7 glass.

The complex evolution of the chemical composition of a multi-materials (Glass/Steel/CGF/COx) experiment was modelled using Marthe-PhreeqC considering the glass and iron corrosion layer-by-layer. The modelling results agreed with glass corrosion thicknesses measured by Scanning Electron Microscopy and corroborated mechanisms highlighted in laboratory and on field by previous studies. These computational results enable to extrapolate the simulation over time at the scale of the canister to assess the glass corrosion after 10,000 years of interactions. The results showed the glass alteration rate evolved over the first 500 years and remain nearly constant thereafter, while the iron corrosion rate was nearly constant over the entire simulation time.

The sensitivity analyses made on generic cases evidenced that the corrosion products were quantitatively and qualitatively dependent on the corrosion rates and the chemistry of the surrounding environment. Overall, this study showed that the upscaling of an experiment at the waste package scale is achievable and that the mechanisms identified in laboratory are still reliable on larger scales of time and space.