Evolution of the geochemical background of a HLW cell in the Callovo-Oxfordian formation

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Introduction

The French National Waste Management Agency (Andra) envisages the safe disposal of High-Level Waste (HLW) and Intermediate-Level Long-Lived Waste (IL-LLW) through deep geological storage using a multibarrier system (Figure 1). The radioactivity confinement and the waste storage are based on a homogeneous and continuous low permeability clay formation, called the Callovo-Oxfordian formation (CallOx) .

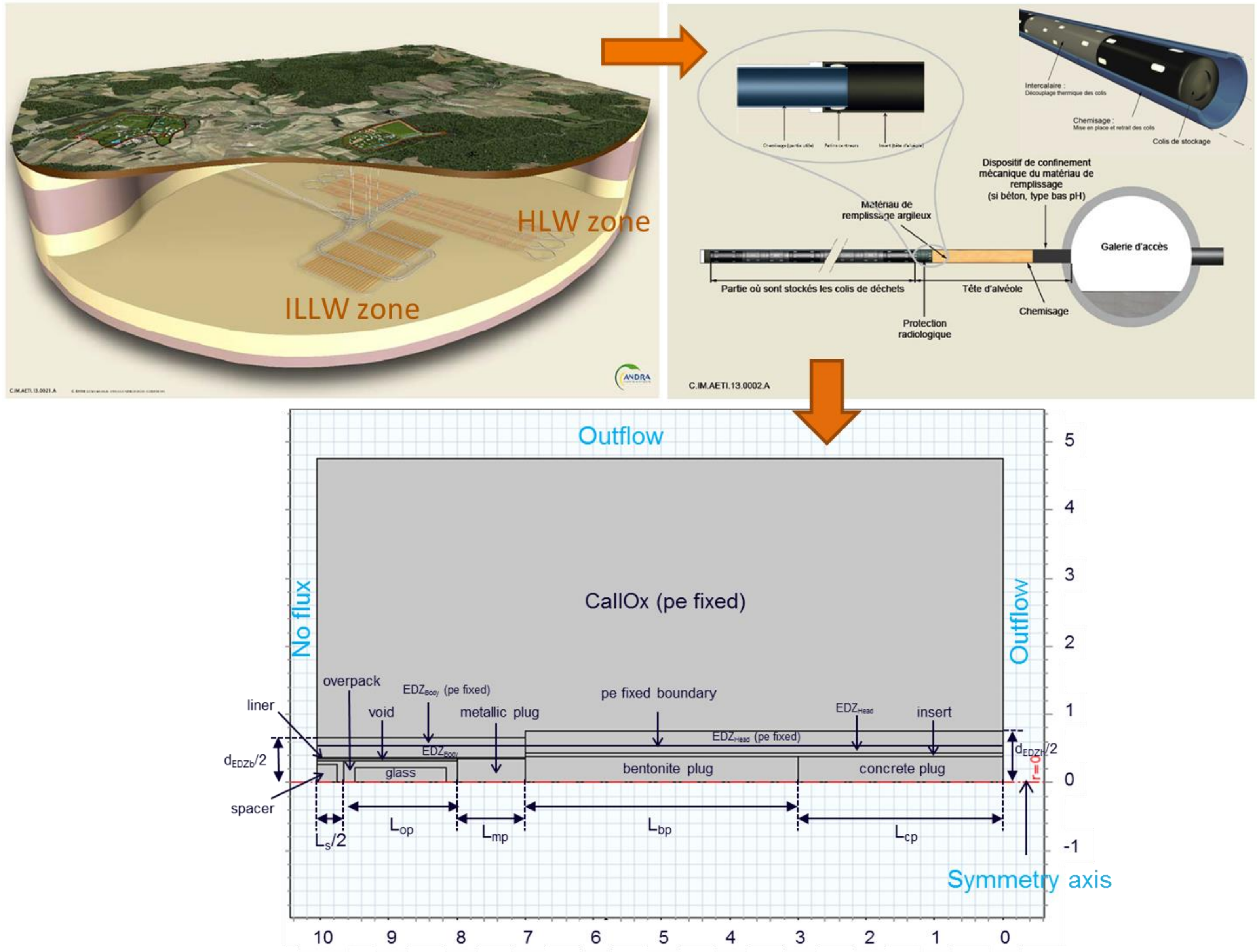

Figure 1. Schematic view of the deep disposal project (CIGEO) (top left). Overall representation of the HLW and IL-LLW repository architecture (top right). HEAD model domain, geometry, materials (subdomains) and boundary conditions considered in the present modelling (bottom).

Objectives

The objective of this work was to update a previously developed reactive transport model accounting for the chemical and thermal evolution of a HLW cell. The main improvements consist of a more detailed description of the surface complexation and ion exchange processes considered in the geochemical model. Different corrosion rates are assumed for different steel barriers. Also, the diffusion through the steel domains increases with time as corrosion proceeds, according to the experience.

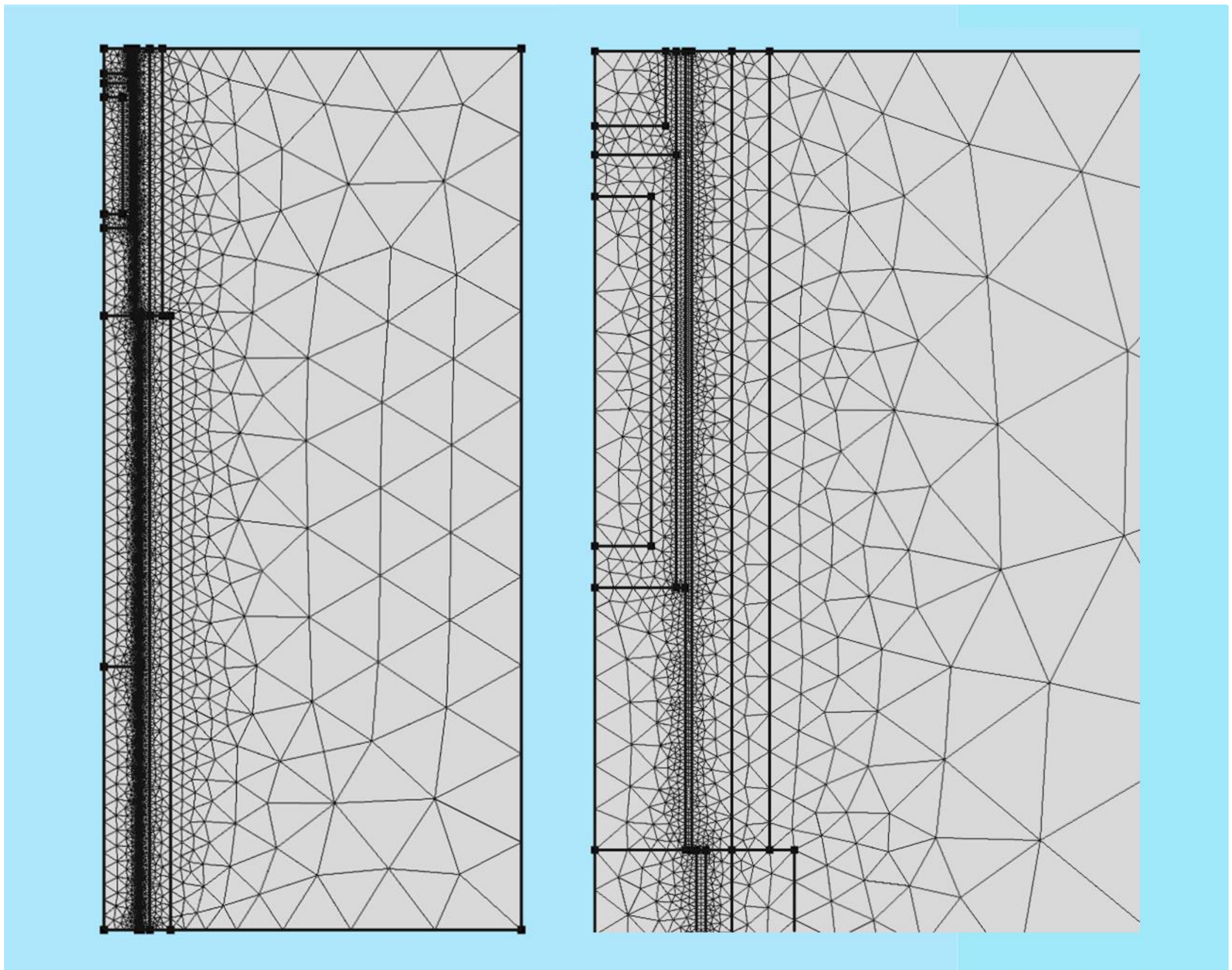

Figure 2. Detailed finite element mesh of the HLW cell and the surrounding CallOx (left) with a zoom around the glass and overpack (right).

Modeling approach

The updated geochemical conceptual model of the HLW cell with surrounding parts of the CallOx formation was implemented in iCP (Nardi et al., 2014), a high performance computing framework based on the coupling of COMSOL Multiphysics® and PHREEQC (Parkhurst and Appelo, 2013).

The model is developed for the Head configuration of the HLW cell considering the last design of the HLW cell and materials parameters provided by Andra. Reactive transport calculations focused on the “background” geochemical evolution for a period of 100,000 years. Subdomains discretization is optimized to improve the quantification of reactive transport through the different barriers at a reasonable CPU time (Figure 2).

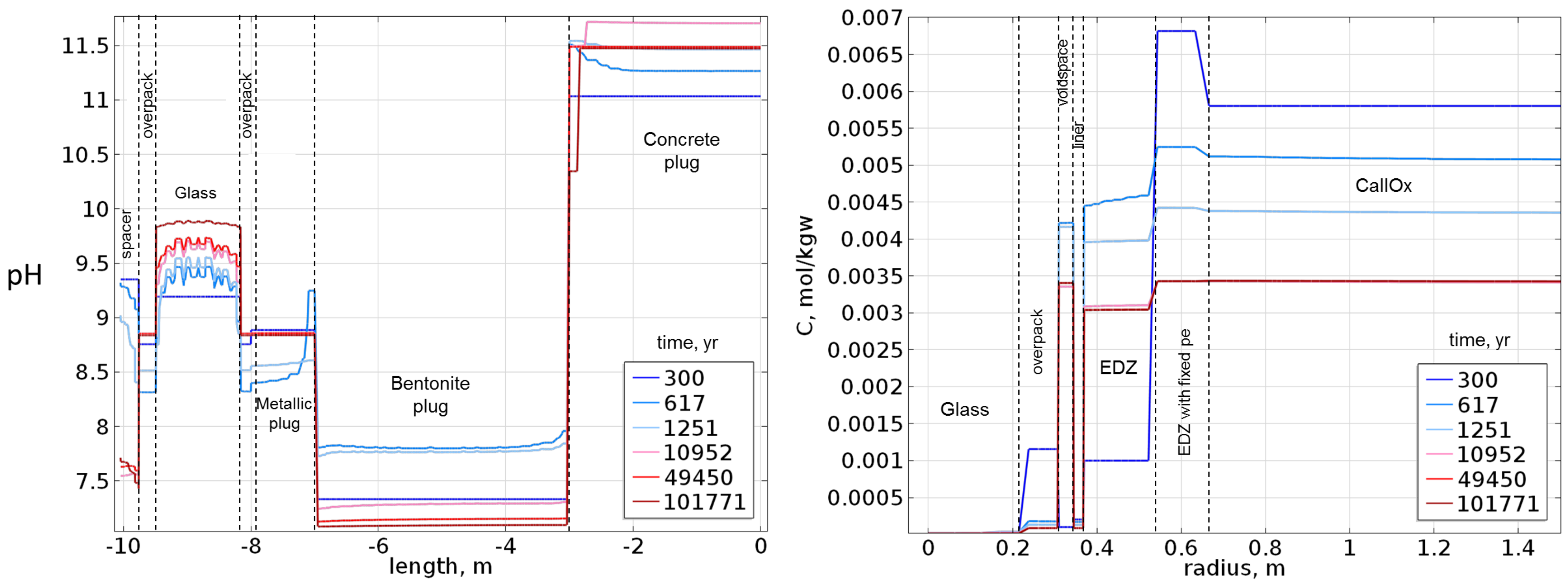

Figure 3. Evolution of pH along the HLW cell (left) and carbonates radial profile at the glass package position (right).

Results and conclusions

The changes in the chemical composition of waters and mineral phases are due to a combination of chemical reactions, diffusion of aqueous species and thermal effects. The impact of temperature decrease during the initial 10,000 years of calculation is significant. Depending on the particular reaction in consideration, the decrease of temperature may trigger mineral dissolution (e.g. gibbsite) or precipitation (e.g. calcite) affecting the overall aqueous composition of the system. A significant effect related with temperature decrease is the increase in pH due to impact on water stability constant (Figure 3).

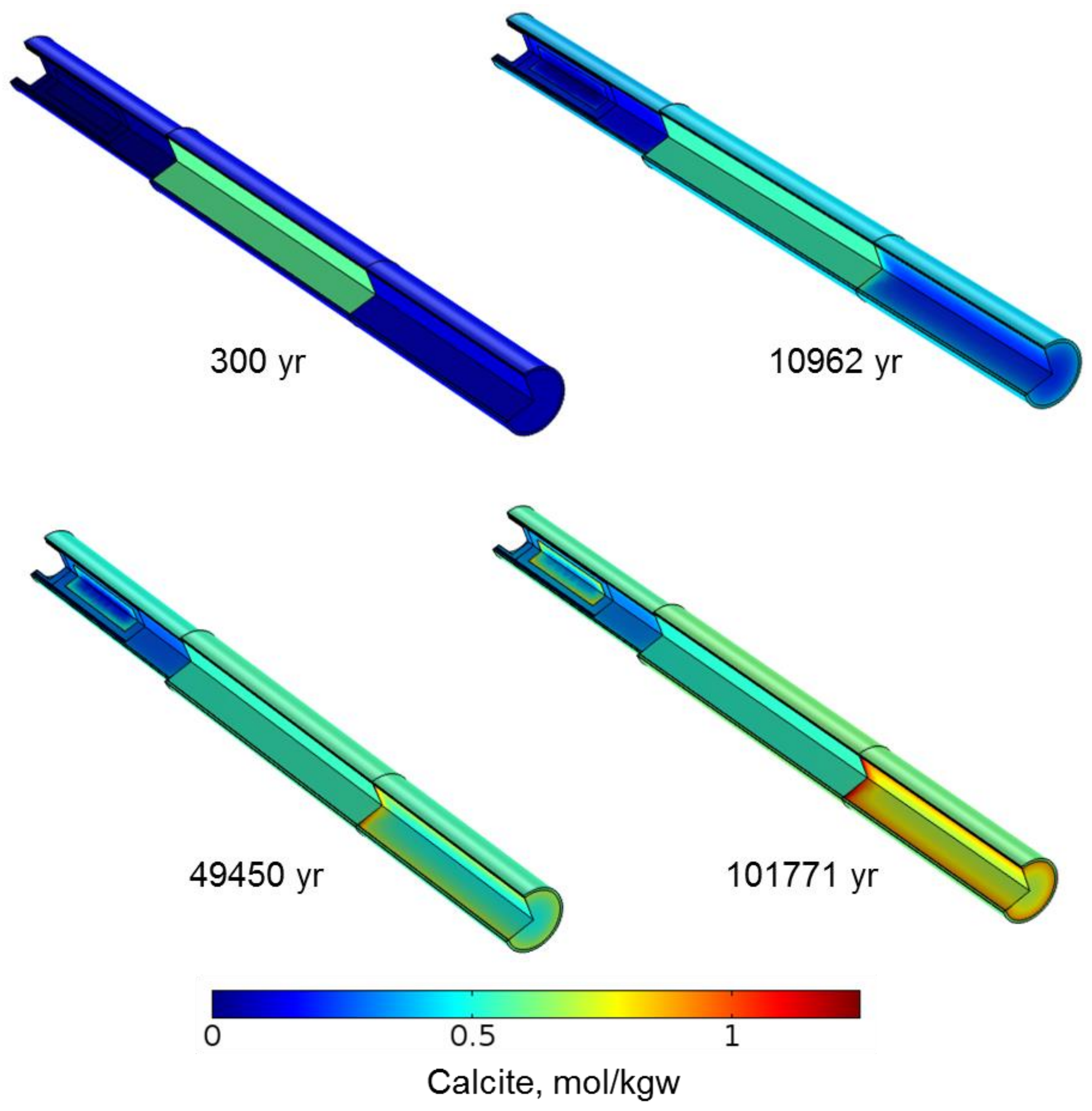

Figure 4. Evolution of calcite concentration through the HLW cell.

Important drivers for the system’s reactivity include: the dissolution of the waste glass, anoxic steel corrosion coupled with hydrogen generation, and major dissolved concentration gradients between the waste cell and the surrounding CallOx claystone (Figure 3). As a result of multi-solute diffusion a complex pattern of mineral precipitation/dissolution is created across the model domain (Figure 4).

References

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Parkhurst, D. L., and Appelo, C. A. J., 2013. *Description of input and examples for PHREEQC version 3—A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations*, U.S. Geological Survey Techniques and Methods, book 6, chap. A43, 497 p., available only at http://pubs.usgs.gov/tm/06/a43/

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