Modeling of Geochemical Evolution and Reactive Transport of Radionuclides in the Near-Field of a Disposal Cell for High-Level Vitrified Waste in the Callovo-Oxfordian Claystone

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Introduction

Andra (the French national radioactive waste management agency) is planning to dispose of high-level vitrified radioactive waste (HLW) in a deep-underground geological facility engineered in the Callovo-Oxfordian (COx) claystone [1], called Cigéo (Figure 1). Quantitative calculations of radionuclide (RN) release, reaction, transport and impact on the biosphere are necessary to evaluate the safety of the repository concept. Physical and chemical processes taking place within the Engineered Barrier System (EBS) and in the surrounding host-rock may significantly affect RN transfer into the near-field of the facility.

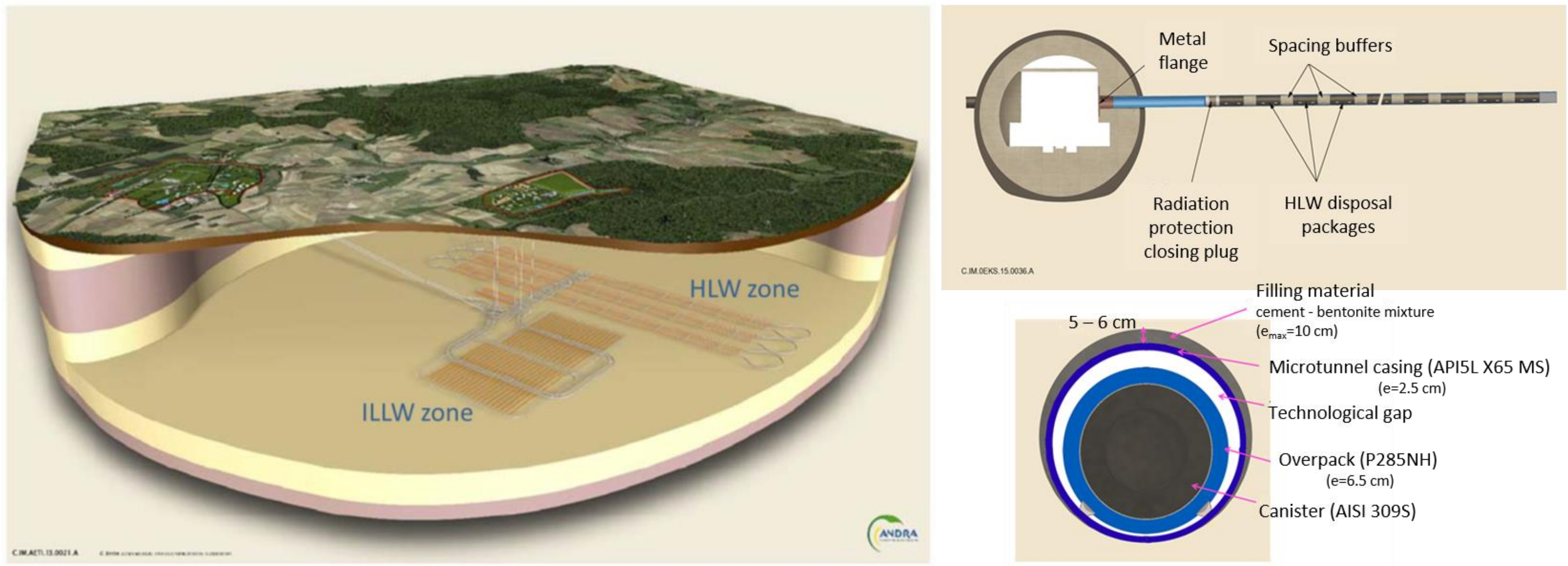

Figure 1. Schematic view of the deep disposal project (Cigéo) (left). Overall representation of the HLW repository architecture (right).

Objective

Many key processes (such as thermal, glass dissolution, steel corrosion, cementitious material degradation, porosity changes due to mineral dissolution/precipitation, or RN retardation) are closely coupled. Moreover, the geometry of the system can result in complex patterns of dilution and preferential transport effects. The objective of this work was therefore to:

1. Develop a reactive-transport model integrating detailed experimental information within a modelling framework capable of representing coupled chemical and transport processes, as well as geometrical details of the engineering design.
2. Simulate the long-term geochemical evolution of a single cell for HLW and the RN release and migration in the near-field COx.

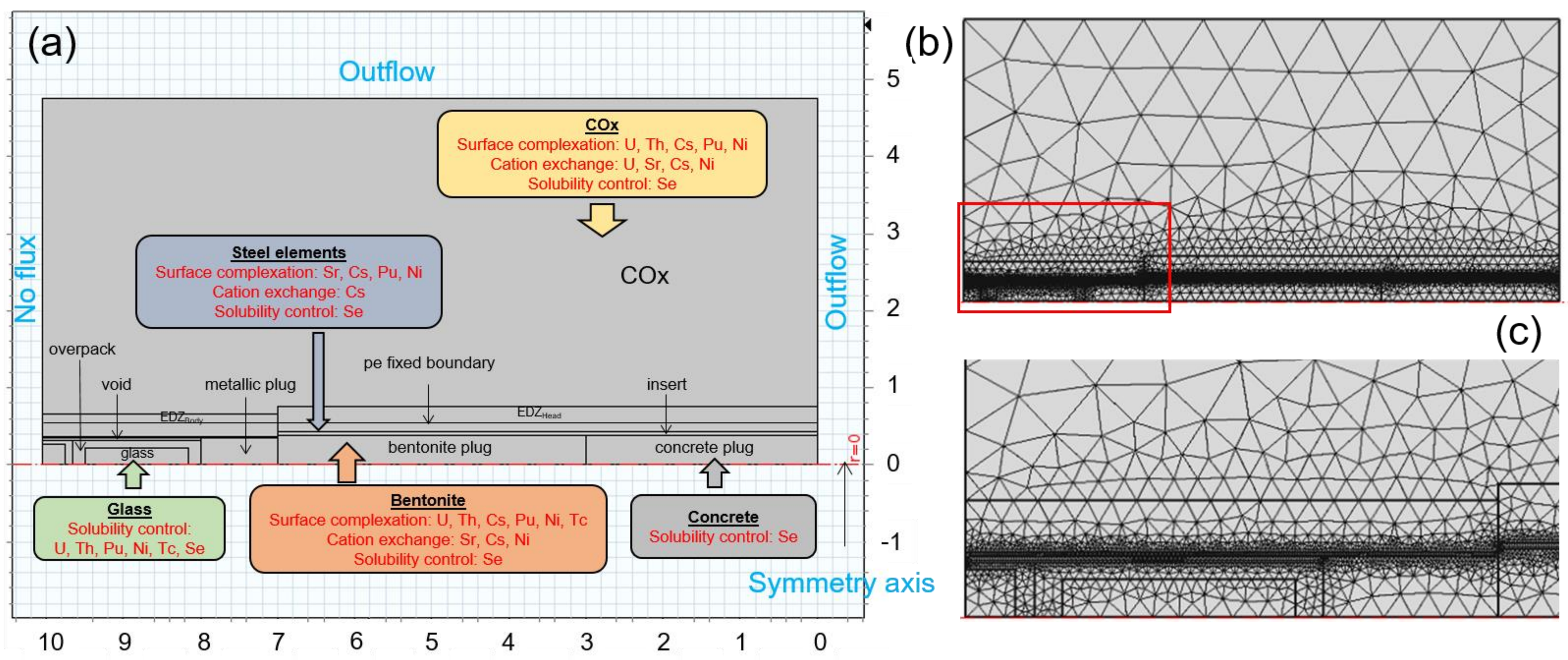

Figure 2. (a) Geometry and mechanisms of RN retention, and (b) computational mesh of the problem (c, zoom in).

Modeling approach

A reactive transport model was implemented in iCP [3] (a numerical interface that couples COMSOL Multiphysics [4] and PhreeqC [5])

iCP interface COMSOL PHREEQC

- ❑ 2D axisymmetric domain over ca. 10 m (Figure 2).
- ❑ Chemically complex system of multiple materials (vitrified waste, steel, cementitious material, compacted bentonite and claystone).
- ❑ RN release from glass follows a kinetic rate law.
- ❑ Coupling with multicomponent Fickian diffusive transport: 22 aqueous components including 8 RN (U, Th, Sr, Cs, Pu, Ni, Tc and Se).
- ❑ Chemical reactions are temperature-dependant and the impact of porosity changes due to dissolution and precipitation reactions on diffusion is explicitly included.
- ❑ Thermodynamic database *Thermochimie* (http://www.thermochimie-tdb.com/).
- ❑ 20 equilibrium and 8 kinetic phases precipitate/dissolve simultaneously.
- ❑ A mechanistic mineral-specific retardation model is defined for each RN in each compartment (Figure 2a).
- ❑ Selected retardation competition effects from the major background dissolved components (e.g., Ca, Mg, K, Sr).

Selected results

Simulations allowed to study the effect of assumptions and details of the EBS on the chemical evolution of the disposal cell and RN transfer, over a period of 100,000 years. Temperature decrease causes a significant increase in pH due to impact on water stability constant (Figure 3a). Important drivers for the system's reactivity include: the dissolution of the waste glass, anoxic steel corrosion and major dissolved concentration gradients between the HLW cell and the surrounding COx claystone (Figure 3b).

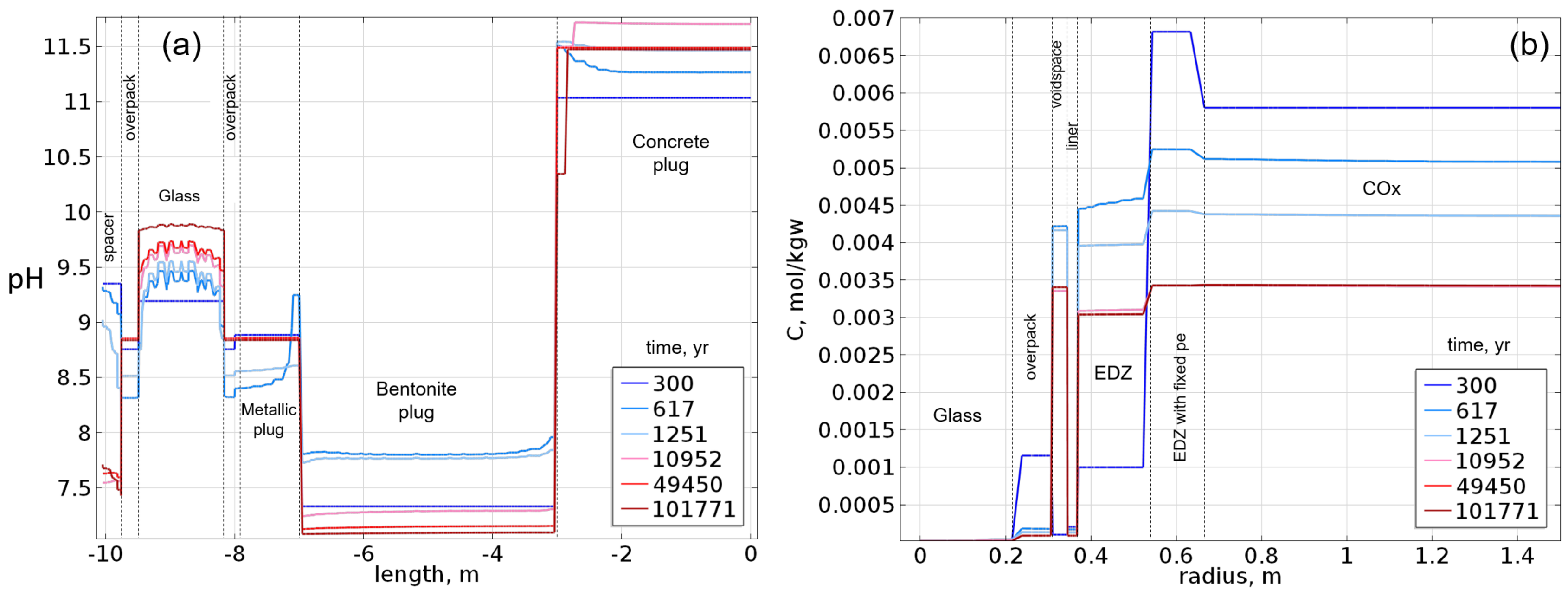

Figure 3. Evolution of (a) pH along the HLW cell and (b) carbonates radial profile at the glass package position.

As a result of multi-solute diffusion a complex pattern of mineral precipitation/dissolution is created across the model domain (Figure 4a). Retardation by surface complexation, cation exchange and mineral dissolution/precipitation affects significantly the transport of U (Figure 4b), Th, Pu, Cs and Ni. For these elements retardation in COx plays an important role during transport through the disposal system and the adjacent host-rock.

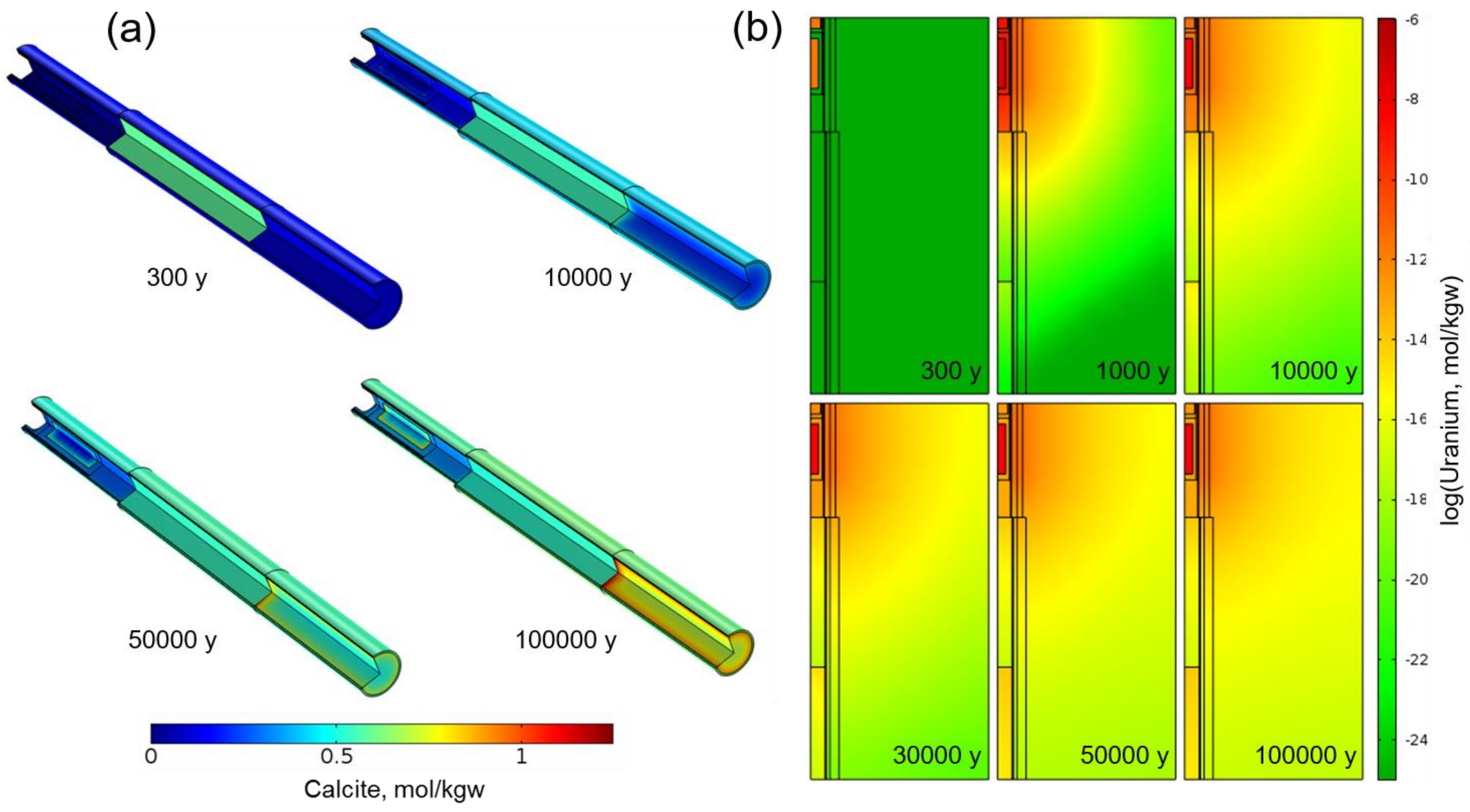

Figure 4. Evolution of (a) calcite in the HLW cell and (b) uranium in the HLW cell and COx.

Diffusion through the liner increases in time due to corrosion. Thus, for most RN that are strongly retained in the COx, the liner constitutes a preferential pathway that dominates transport in the axial direction. The above is combined with the relatively limited retardation onto magnetite resulting from the liner corrosion. This behaviour is however not the case for Ni, which is strongly retarded by surface complexation onto magnetite. This also results in competition with other RN (Cs, Sr and Pu), which has the effect to promote their preferential transport through the liner.

References

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