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► **To cite this version:**

Dauren Sarsenbayev, Christophe Tournassat, Carl I Steefel, Haruko M Wainwright. Building confidence in models for complex barrier systems for radionuclides. Proceedings of the National Academy of Sciences of the United States of America, 2025, 122 (27), <10.1073/pnas.2511885122>. <insu-05143813>

HAL Id: insu-05143813

<https://insu.hal.science/insu-05143813v1>

Submitted on 4 Jul 2025

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Building confidence in models for complex barrier systems for radionuclides

Dauren Sarsenbayev^{a,b} , Christophe Tournassat^{b,c} , Carl I. Steefel^{b,1} , and Haruko M. Wainwright^{a,d}

Edited by Donald Canfield, Syddansk Universitet, Odense M., Denmark; received May 13, 2025; accepted June 3, 2025

The modeling and simulation of the Cement–clay Interaction–Diffusion field (CI-D) experiment at the Mont Terri site in Switzerland presented here demonstrates that it is possible to capture the multiscale physical and chemical features of natural and engineered barrier systems for radionuclides. The simulations are successfully carried out with the newly developed CrunchODiTi high-performance computing software that accounts for multiple continua, including a continuum representing the electrical double layer (EDL) developed along negatively charged clay particles in clay rock. The simulation also accounts for both the complex three-dimensional (3D) geometry, expected as the norm in a geological waste repository, and the anisotropy of the geological formation. In addition, the high resolution of the model makes it possible to include “skin effects” developed at the interface between highly reactive materials, in this case between the high pH cement and the circumneutral but electrostatic Opalinus Clay. The successful history matching with the field experiment demonstrates that the distinct geochemical and physical properties of the cement and the Opalinus Clay in the CI-D experiment can be accounted for. Such analyses are essential for developing a defensible safety case for the underground storage of radioactive waste.

geological waste repository | radionuclide transport | barrier systems | anion exclusion | high-performance computing

The predictive modeling of radionuclide migration in geological formations is a fundamental aspect of nuclear waste management (1). A significant challenge now is to develop this capability in the context of natural and engineered barrier systems consisting of multiple materials that are typically distributed in complex 3D geometries (2, 3). The resulting heterogeneous transport properties (permeability and diffusivity) and general lack of data on these in the subsurface lead to challenges in predicting the migration rate of radionuclides from geological nuclear waste repositories (4). These physical complexities are further compounded by a range of chemical processes that influence the retardation of the migration rate of radionuclides (5).

The problem of designing a safe geological nuclear waste repository and predicting its behavior into the future, often over geological time scales, is made additionally challenging by the fact that the physical and chemical properties of interfaces between materials evolve over time. Often, these interfaces (e.g., between cement at pH 12 to 13 and clay-rock between pH 8 to 9) involve smaller-scale (mm to cm) reactive transport processes that require higher spatial resolution as compared to the dimensions of the repository. Each of the materials requires its own set of process models to describe radionuclide migration and time-dependent property changes that ideally would go well beyond the standard probabilistic linear distribution (or K_d) approaches (1, 6).

Reactive transport codes often do not consider the inherently multiscale nature of the problem, with processes occurring at the mm (or less) scale affecting both near-field and far-field migration of radionuclides into the biosphere. In addition, a particular challenge associated with geological repositories that is only rarely addressed is that of charged porous media (7). This oversight is particularly significant in low-permeable, compacted clay environments, which are integral to high-level nuclear waste repository designs worldwide due to their ability to impede radionuclide migration (8). In such formations, ion mobility is not merely a function of concentration gradients, but is also influenced by electrostatic interactions (9). Indeed, the consideration of the presence of an electric double layer (EDL)—the formation of a diffuse layer (DL) bordering negatively charged clay particles, leading to partial anion exclusion and a cation accumulation within these layers (7, 10)—is crucial for accurately simulating the transport of charged species, including significant anionic activation and fission products like $^{36}\text{Cl}^-$, $^{129}\text{I}^-$, $^{99}\text{Tc}^-$, and $^{79}\text{Se}^-$ —key dose contributors with high mobility and long half-lives (11). In this respect, the introduction of CrunchClay, now CrunchODiTi (for Off-Diagonal Terms integration), an evolving branch of the established CrunchFlow code (1), represents a significant evolution in the field of geochemical transport modeling.

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Author contributions: D.S., C.T., C.I.S., and H.M.W. designed research; D.S., C.T., and C.I.S. performed research; D.S., C.T., C.I.S., and H.M.W. analyzed data; C.T. primary developer of software CrunchODiTi; C.I.S. contributed to development of CrunchODiTi software; and D.S., C.T., C.I.S., and H.M.W. wrote the paper.

The authors declare no competing interest.

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Published July 3, 2025.

Given these complexities, it is critical to i) apply a new generation of modeling tools such as CrunchODiTi that are capable of capturing reactive transport phenomena over large spatial and temporal scales and ii) validate these models where possible against field and laboratory experiments. The long-term (13-y) cement–clay interaction–diffusion (CI-D) experiment at Mont Terri in Switzerland provides an excellent opportunity for such validation, consisting as it does of multiple engineered and natural materials arranged in asymmetric 3D geometries in the subsurface at Mont Terri. The CI-D experiment, which is part of the broader CI project at the Mont Terri site, consists of four boreholes (12). A recirculation system into the early boreholes was used for the injection of radiotracers, including the uncharged tritiated water (HTO) and the isotopolog of the anion chloride, $^{36}\text{Cl}^-$. The experiment consists of out-diffusion of the radiotracer “cocktail” into the cement and then into the Opalinus Clay.

Results

Here, we report for the first time the results of a large 3D reactive transport modeling simulation (1.5×10^6 grid cells) with a complex geometry (Fig. 1A) that takes into account the influence of the EDL developed in the Opalinus Clay on radionuclide transport. During the first 450 d, the plume is primarily confined within the isotropic cement, while after 450 d the anisotropy of the Opalinus Clay and the negative net charge of the clay matrix begins to impact the charged solute’s transport (Fig. 1C). The simulated evolution of the relative borehole concentrations of the tracers over 900 d was in good agreement with the experimental results (Fig. 1C). We simulated the cement–clay interface by explicitly setting a 0.14% porosity of the “skin” interface of 1 cm in thickness. These “skin effects” are likely due to chemical interactions between the high-pH cement and the circumneutral Opalinus Clay that lead to mineral precipitation that reduces the porosity and thus diffusivity at the interface. From Fig. 1C, it is apparent that the gradient of the concentration curve of $^{36}\text{Cl}^-$ begins to steadily decrease at 450 d, indicating anion exclusion of

the chloride in the negatively charged Opalinus Clay. This is further demonstrated by the fact that the neutral HTO does not exhibit gradient flattening in the charged medium, indicating transport unaffected by EDL interactions.

Discussion

We show that it is possible to replicate observed radiotracer transport behavior in a multicomponent, anisotropic geometry that includes a high-resolution (cm-scale) treatment of the cement–Opalinus Clay interface using data from the Mont Terri CI-D experiment. By accounting for key physicochemical processes—such as electrostatic effects in clay, sorption in cement, ionic strength variations, and interfacial skin effects—our results support the reliability of modeling radionuclide migration across evolving material interfaces. We demonstrate the potential of this approach through large-scale 3D simulations performed with the massively parallel CrunchODiTi software. This approach establishes confidence in modeling radionuclide behavior in complex barrier systems relevant to deep geological repositories being considered or deployed worldwide. Our findings point to the limitation of methods that ignore the differing reactivity and transport properties of the multiple materials typically expected in repositories consisting of engineered and natural barrier systems. They also reinforce the conclusions drawn in earlier studies that the electrostatic effects on radionuclide (especially anionic) transport in clay-rich rocks cannot be neglected (13).

Summary of Methods

A model based on the massively parallel software CrunchODiTi is used that incorporates electrostatic effects on transport such as diffusion potentials and partial anion exclusion in DLs developed adjacent to negatively charged clay particles (7, 9, 10, 14, 15). The simulation grid has dimensions of 1.8 m \times 1.8 m \times 1.8 m, with a total of 1,468,435 variably sized grid cells (including boundary cells). The list of primary unknowns included 8 aqueous

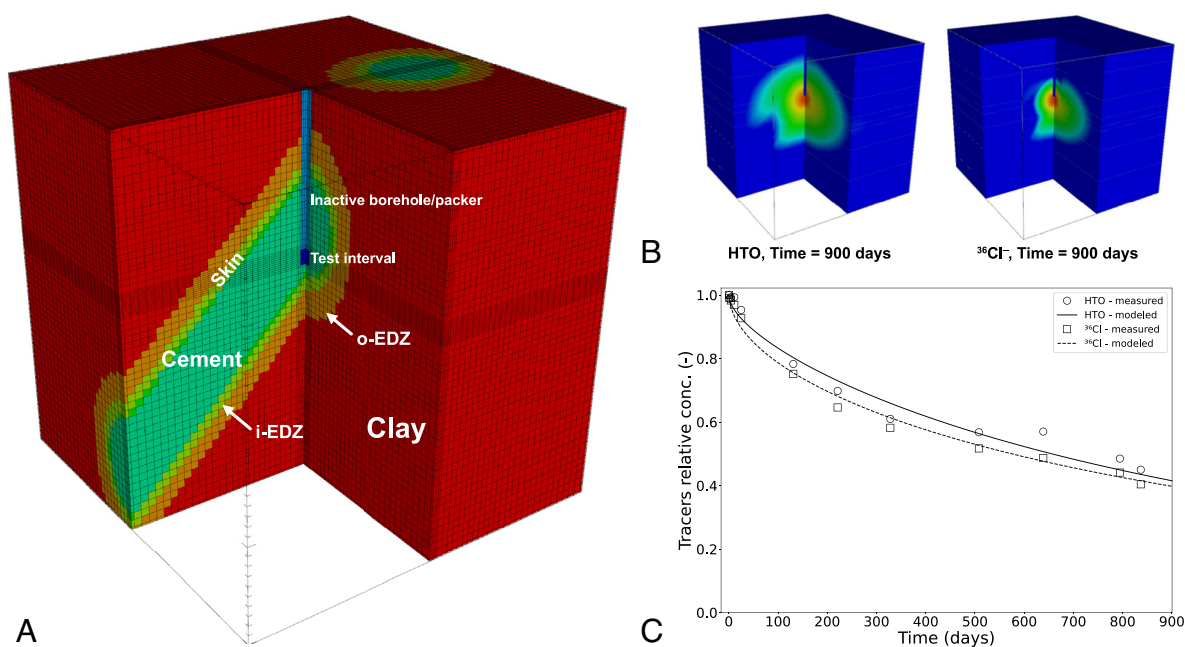


Fig. 1. (A) Computational domain and material distribution used in the simulations. The domain is rotated so that the Opalinus Clay strata are vertical. (B) 3D contour plots of neutral HTO (Left) and charged $^{36}\text{Cl}^-$ (Right) solutes at 900 d. (C) Comparison of observed (symbols) and simulated (lines) borehole concentrations using the 3D model.

components (H_2O , H^+ , Na^+ , Cl^- , HTO , Ca^{2+} , SO_4^{2-} , and Br^-), 1 solid phase upon which sorption occurs, 1 surface potential, and 1 DL potential. In addition, ionic strength, water activity, and diffusion potential (as described by the Nernst–Planck equation) were included, bringing the total number of primary variables to 14. This results in approximately 21 million degrees of freedom ($1,468,435 \times 14 = 20,558,090$). The code is parallelized using the implementation of Message Passing Interface (MPI) in the Portable, Extensible Toolkit for Scientific Computation (PETSc) (16). Further details are provided in the *SI Appendix*. The simulations are used to predict the concentrations of anionic chloride and HTO in the borehole (results shown in Fig. 1C) that are

influenced by the combination of multiscale and multicomponent processes discussed here.

Data, Materials, and Software Availability. All study data are included in the main text.

ACKNOWLEDGMENTS. Funding for this work has been provided by the Spent Fuel and Waste Disposition Campaign, Office of Nuclear Energy of the US Department of Energy, under Contract Number DE-AC02-05CH11231 with Lawrence Berkeley National Laboratory. Additional support for D.S. was provided by the US Department of Energy, Office of Nuclear Energy through the Ingenuity program at Berkeley Lab. D.S. and H.M.W. acknowledge the support from the US Department of Energy, Office of Nuclear Energy through the Nuclear Energy University Program.

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