

Dispersion-induced traveling fronts

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1. Introduction

Emergent phenomena are of great concern in the risk analysis of nuclear waste storage sites. The unpredicted and **the fast transport of radionuclides** in the subsurface has been observed in the vicinity of some nuclear waste repositories in the United States. The phenomenon has been attributed to physical factors, i.e., precipitation and fracture flow, and to geochemical factors, such as:

- **Organic ligands** that form strong complexes with radionuclides enhancing mobility;
- **Sorption capacity** of sediments that might retard migration, but in presence of complexing ligands drastically reduces it.
- Low **pH** that might favor the desorption of adsorbed radionuclides;
- **Colloids** formation (e.g., PuO) which travel at the main flow velocity.

The **combination of flow transport and geochemical models** of the subsurface has been used in this contest and has highlighted the role of both the dispersion and the reaction at the liquid/solid interface to affect radionuclide mobility [1].

3. Modeling

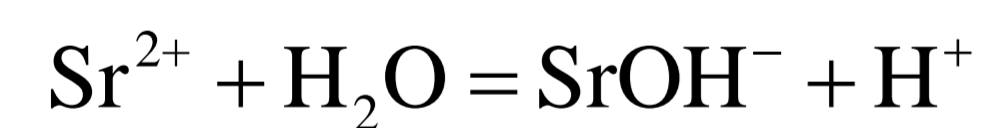
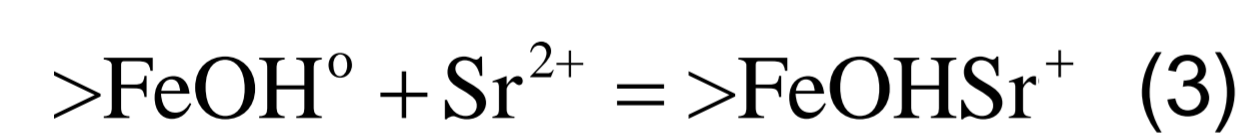
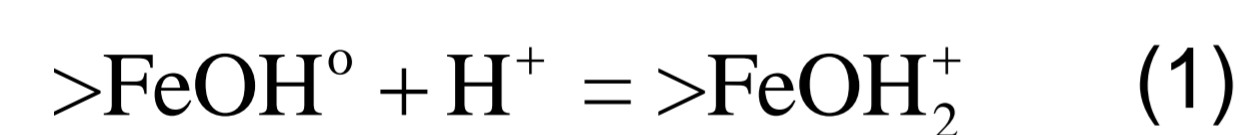
Reactive transport equations, strontium (Sr²⁺) and acidity:

$$\phi \frac{\partial(C_{Sr} + Z_{Sr})}{\partial t} + u \frac{\partial C_{Sr}}{\partial x} - D \frac{\partial^2 C_{Sr}}{\partial x^2} = 0$$

$$\phi \frac{\partial(C_H - C_{OH} + Z_H)}{\partial t} + u \frac{\partial(C_H - C_{OH})}{\partial x} - D \frac{\partial^2(C_H - C_{OH})}{\partial x^2} = 0$$

where C_i and Z_i are the concentrations of the aqueous and adsorbed specified species; u is the Darcy velocity; D is the dispersion coefficient; ϕ is the media porosity.

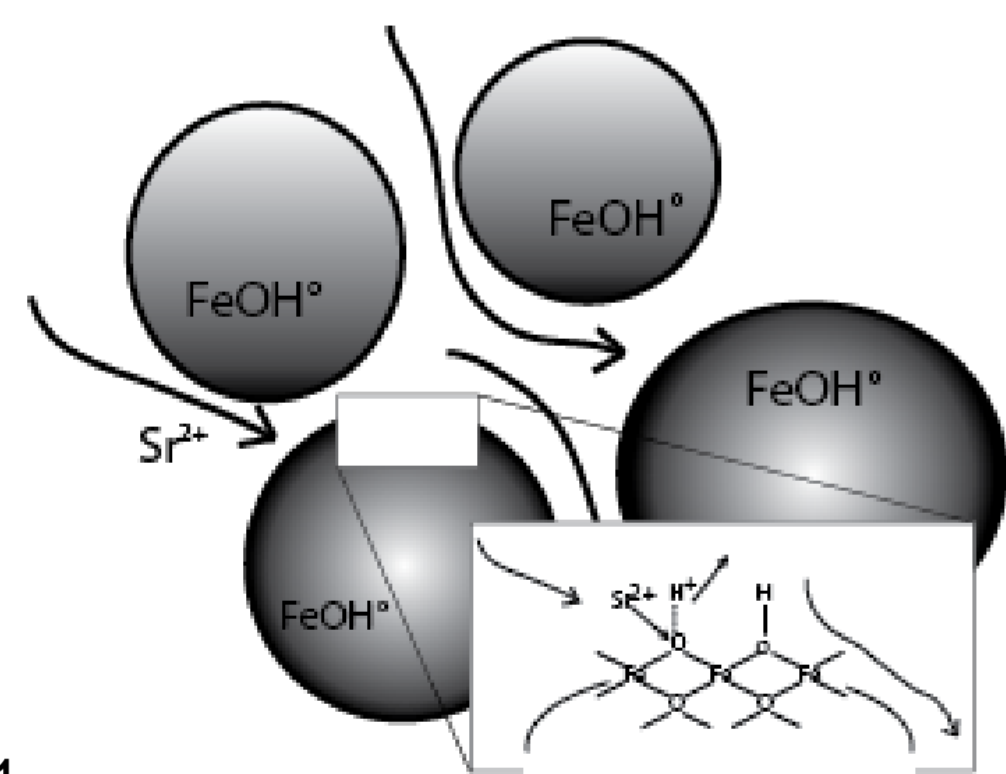
Geochemical model:



At 25°C, $K_1=10^{-7.3}$, $K_2=10^{-8.9}$, $K_3=10^{5.1}$

Adsorption isotherms:

$$Z_{Sr} = \frac{a_{Sr} K_3 Z_{tot}}{1 + a_H K_1 + a_{Sr} K_3}, Z_H = \frac{a_H K_1 Z_{tot}}{1 + a_H K_1 + a_{Sr} K_3}$$

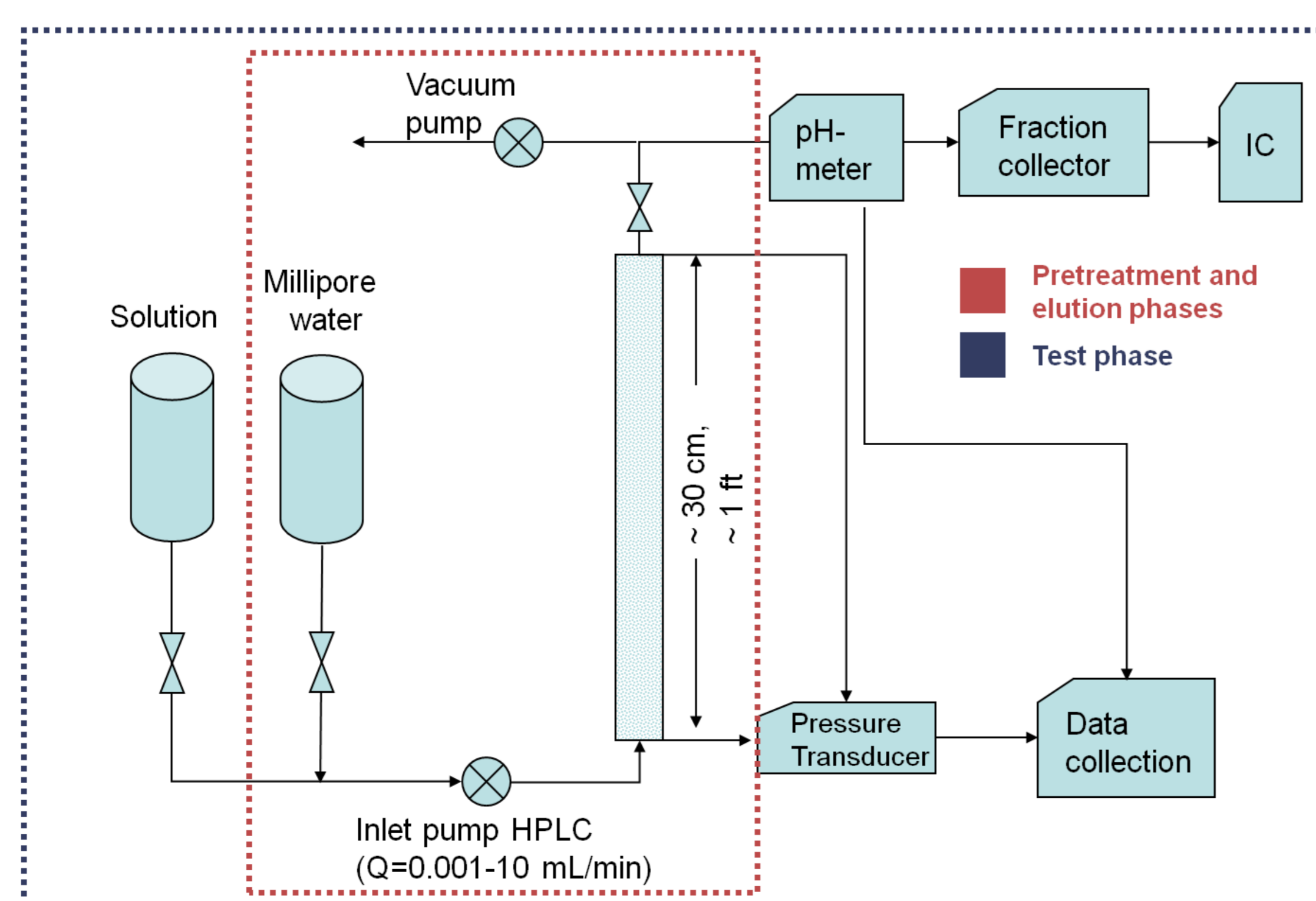


5. Experimental

Core-scale experiments will be performed to investigate the transport of Sr²⁺ throughout a column packed with silica beads coated with **amorphous ferric oxyhydroxide (HFO)**. pH, concentration of Sr²⁺, and total dissolved solids (TDS) will be measured.

The **effect** of the following variables on Sr²⁺ transport will be investigated:

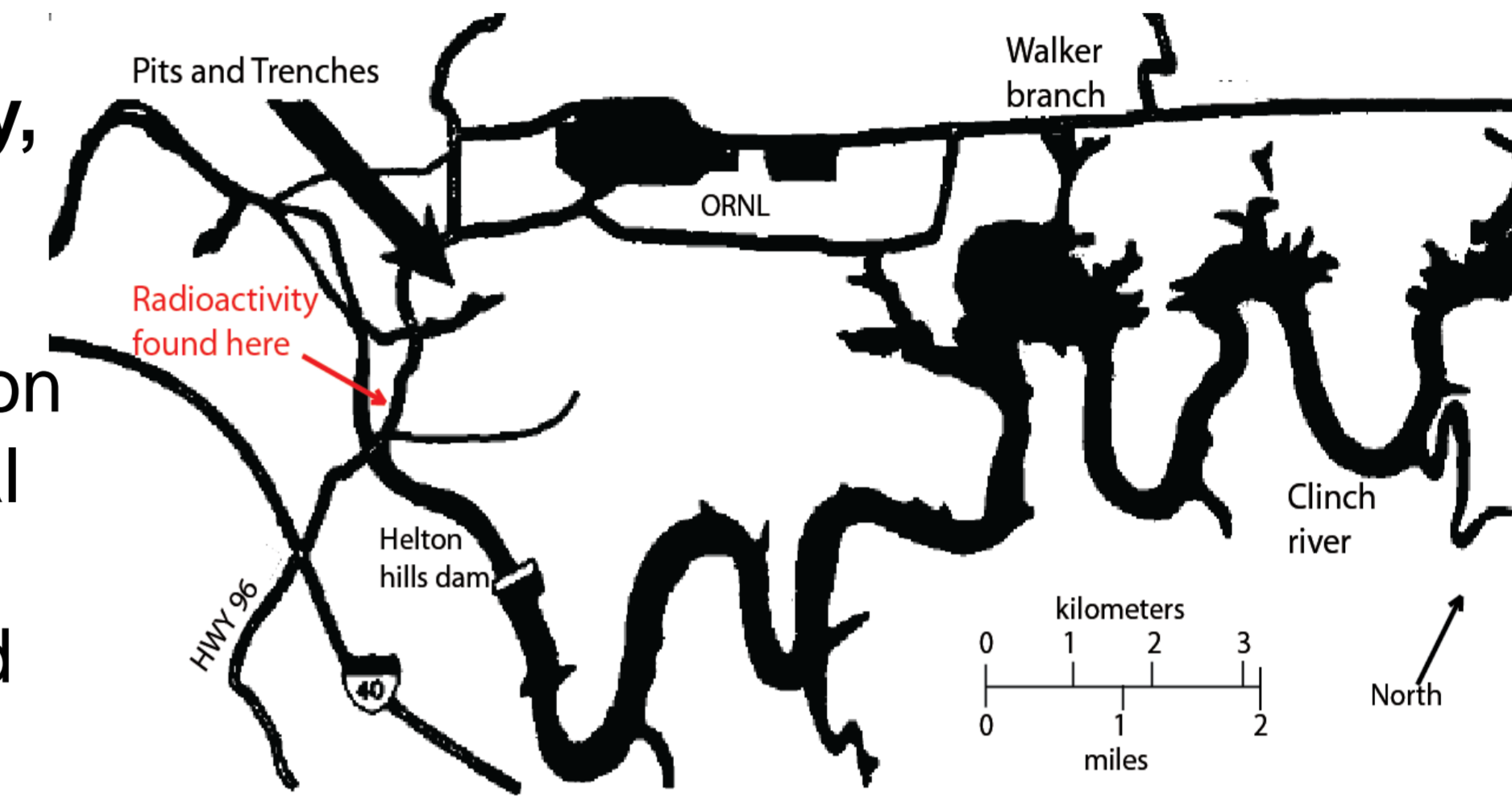
- pH
- Porosity
- Volume fraction of coated-beads
- Ionic strength
- Counter ions
- Flow velocity



2. Field observations

The rapid transport of radionuclides in the subsurface has been observed at, e.g.:

- **Nevada test site.** 828 underground nuclear tests were conducted between 1956 and 1992. Plutonium and other radionuclides traveled unexpectedly hundreds of meter away due to colloid-facilitated transport [2].
- **Oak Ridge National Laboratory, Tennessee.** Low-radioactivity waste tranches mixed with high pH solution to favor immobilization were disposed. However, several radionuclides migrated several hundreds of meters in seeps and streams [3].
- **Radioactive Waste Management Complex at INEEL, Idaho.** The transport of radionuclides modeled assuming only retardation by adsorption on the sediments underestimated the migration distance because the model was based on oversimplified determinations of adsorption coefficients [4].

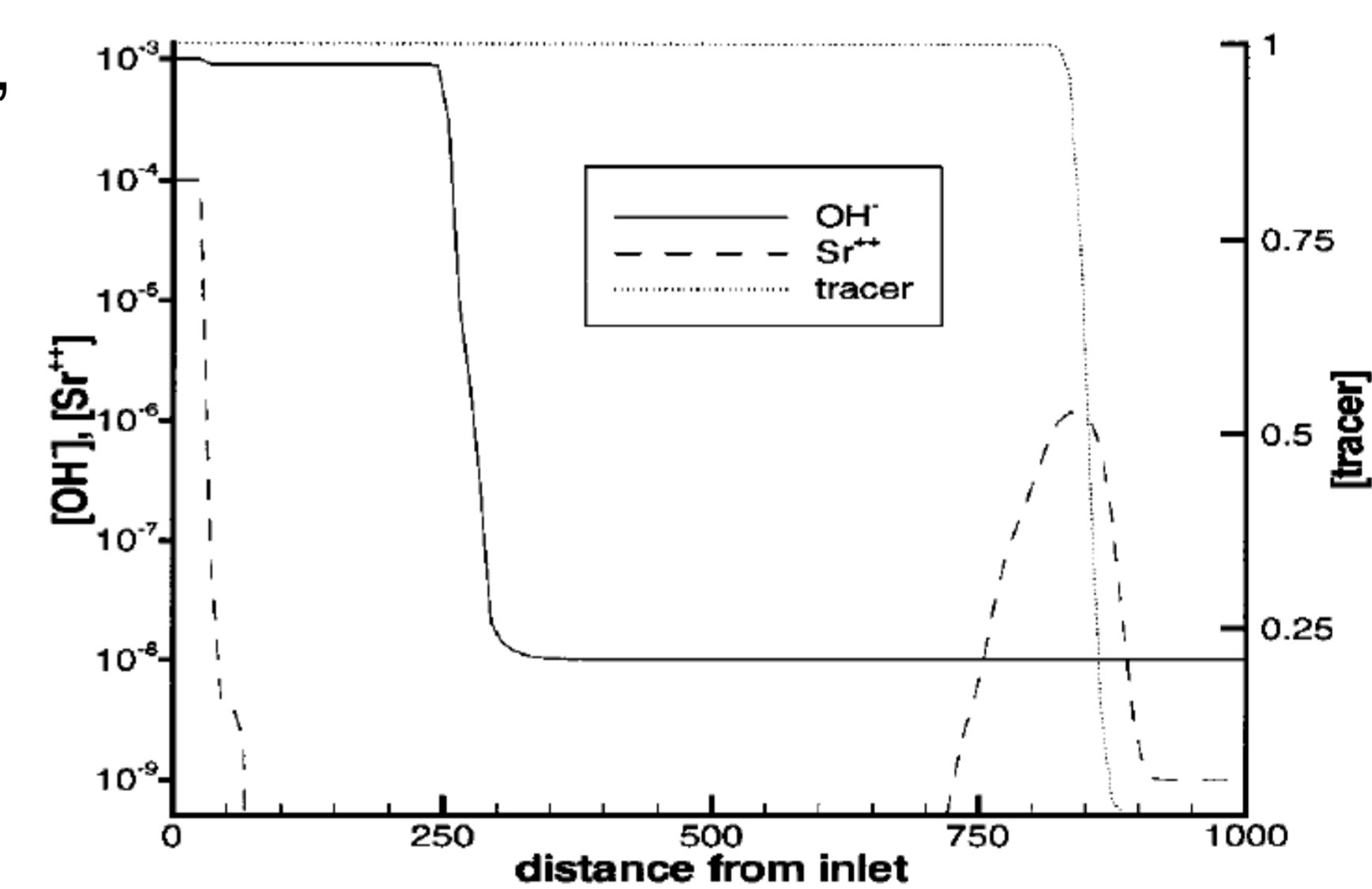
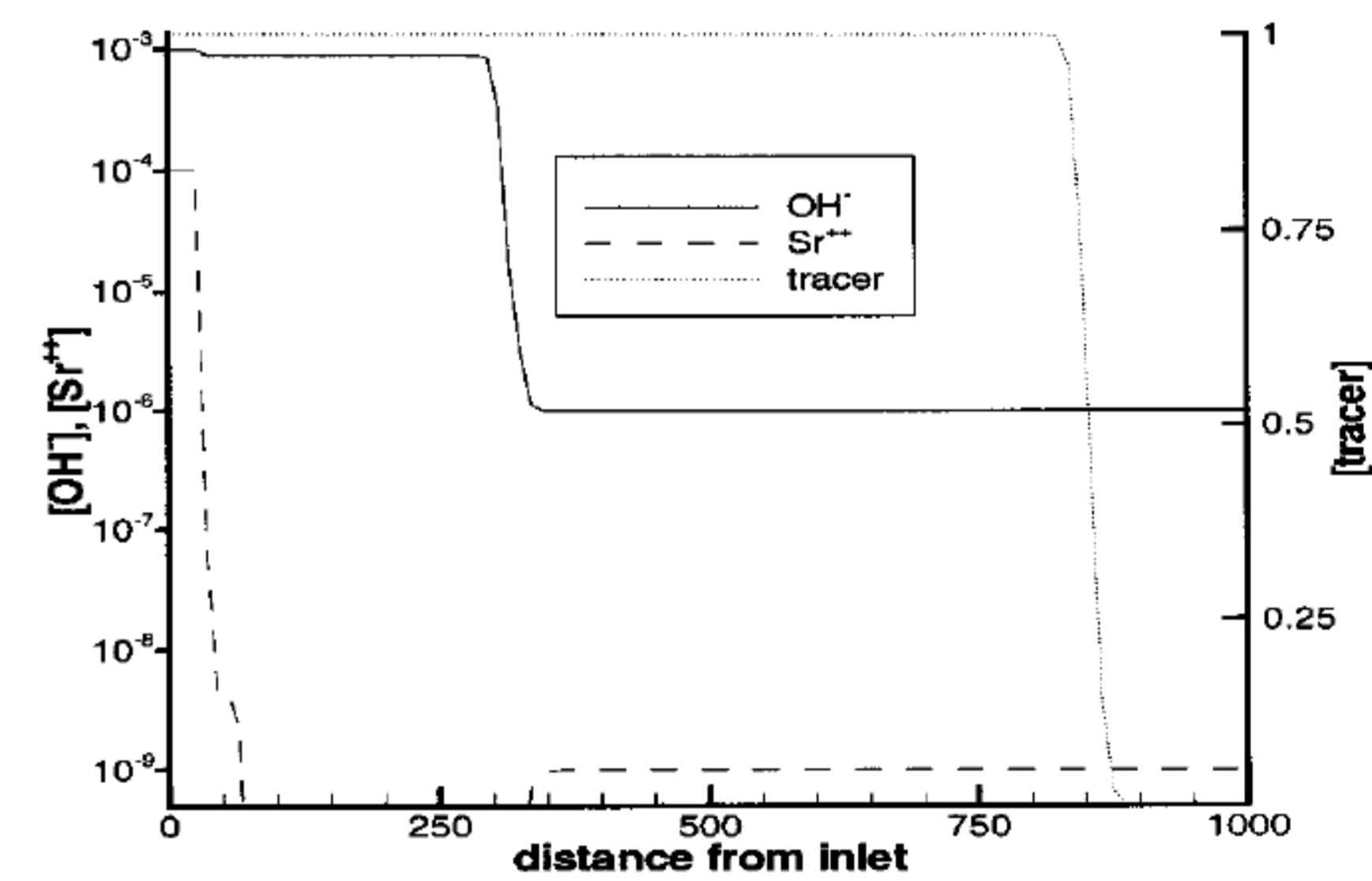


4. Numerical results

In **absence of diffusion/dispersion**, the reactive transport model yields two shocks (i.e., of OH⁻ and Sr²⁺) as predicted from classical theory of chromatography travelling at velocity:

$$v_{Ci} = \frac{u / \phi}{1 + \frac{\Delta Z_i}{\Delta C_i}}$$

In **presence of diffusion/dispersion**, a pulse of Sr²⁺ arises. It travels faster than the Sr²⁺ shock. When the two shocks are near each other, diffusion/dispersion yields the formation of a metal flux through the pH shock towards the low-pH region that favors Sr²⁺ mobility.



6. Conclusions and outlook

The anomalous transport of radionuclides has been observed in some of the U.S. nuclear waste storage sites.

The numerical simulations of Sr²⁺ transport in a HFO porous media highlighted the importance of both dispersion and competitive adsorption of Sr²⁺ and H⁺ ions in generating the formation of a fast travelling front of Sr²⁺.

Core-scale experiments will be run under various operating conditions, and designed and analyzed with a reactive transport model coupled with a geochemical model for solution and solid/liquid interface.

References

- [1] Bryant, S.L.; Dawson, C.; van Duijn, C. J. (2000) Dispersion-induced chromatographic waves. *Ind. Eng. Chem. Res.* **39** 2682-2691.
- [2] Kersting, A. B.; Efurud, D. W.; Finnegan, D. L.; Rokop, D. J.; Smith, D. K.; Thompson, J. L. (1999) Migration of plutonium in groundwater at the Nevada Test Site. *Nature* **397** 56-59.
- [3] Sauders, J. A.; Toran, L. E. (1995) Modeling of radionuclide and heavy metal sorption around low- and high-pH waste disposal sites at Oak Ridge, Tennessee. *Applied Geochemistry* **10** 673-684.
- [4] INEEL (2003) INEEL subregional conceptual model. *Report INEEL/EXT-03-01171*.