

**Evaluation of the Field Exchange Capacity of Hanford Sediments with Implications for  $^{137}\text{Cs}$  Migration.** C.I. Steefel<sup>1</sup>, S.A. Carroll<sup>1</sup>, S.B. Yabusaki<sup>2</sup>, and P.C. Lichtner<sup>3</sup>. <sup>1</sup>Lawrence Livermore National Laboratory, L-204, P.O. Box 808, Livermore, CA 94551 ([steefel@llnl.gov](mailto:steefel@llnl.gov)), <sup>2</sup>Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, <sup>3</sup>Los Alamos National Laboratory, MS F649, Los Alamos, NM 87545.

**Introduction:**  $^{137}\text{Cs}$  is a significant contaminant in many high-level nuclear wastes (HLW) in the United States, Canada, Europe, and the former Soviet Union. While  $\text{Cs}^+$  is known to sorb strongly to the mica and clay fraction of soils, there is evidence at the Hanford nuclear site in the United States of enhanced migration of  $\text{Cs}^+$  in the vadose zone below leaking HLW tanks, especially at the SX-108 tank. Explanations for the enhanced migration of  $\text{Cs}^+$  at Hanford can be broadly separated into those involving primarily physical processes (fast pathways or exclusion of immobile zones within the sediment under low liquid saturation conditions) and those involving primarily chemical processes (competition between  $\text{Cs}^+$  and other cations present at high concentrations in the leaking tank fluids). The potential importance of competing cations is suggested by batch adsorption experiments carried out on Hanford sediments, which demonstrate the applicability of a classical ion exchange model to describe  $\text{Cs}^+$  adsorption [1]. The competitive effect of  $\text{Na}^+$  in particular may be important because of the very high  $\text{NaNO}_3$  concentrations in the tank leaks (up to saturation with solid  $\text{NaNO}_3$  at the SX-108 tank). To evaluate the relative importance of physical and chemical processes in driving enhanced  $\text{Cs}^+$  migration, we have 1) carried out column experiments involving  $\text{Cs}^+$  transport at a variety of  $\text{NaNO}_3$  and  $\text{KNO}_3$  concentrations, and 2) performed three dimensional simulations of non-isothermal multiphase flow and heat transport coupled to multicomponent ion exchange at several of the leaking tank sites where subsurface data is available.

**Column Experiments:** Plug flow column experiments have been carried out at a variety of  $\text{NaNO}_3$ ,  $\text{KNO}_3$ , and  $\text{CsNO}_3$  concentrations using composite Hanford sediment similar to that used in batch adsorption experiments [1]. Experimental results indicate a retardation of  $\text{Cs}^+$  of about 41 at 1M  $\text{NaNO}_3$  and  $10^{-4}\text{M}$   $\text{CsNO}_3$ , while a similar experiment carried out using 5M  $\text{NaNO}_3$  shows a substantially lower retardation of 7.8 (Fig. 1). Flow rate was varied in two of the experiments and had no effect on retardation, suggesting that kinetic effects are not important under these experimental conditions. This conclusion is also supported by the observation that desorption of  $\text{Cs}^+$  is rapid (Fig. 2).

**Field Exchange Capacity:** Three-dimensional modeling of multiphase flow, heat transport, and multicomponent ion exchange and transport were used to evaluate the exchange capacity of the Hanford sedi-

ments in the field. The modeling focused on a 57,000 gallon leak lasting one week at the SX-115 tank. Comparison of the simulation results with data collected from a borehole adjacent to the leaking tank suggests that the field exchange capacity is quite high, close to the CEC determined using conventional batch experimental techniques. This conclusion is based on the strong retardation of both  $\text{Cs}^+$  and  $\text{Na}^+$  at this site. The enhanced migration of  $\text{Cs}^+$  at the nearby SX-108 site, therefore, is likely to be at least partly a chemical effect related to the higher  $\text{NaNO}_3$  and  $\text{Cs}^+$  concentrations there.

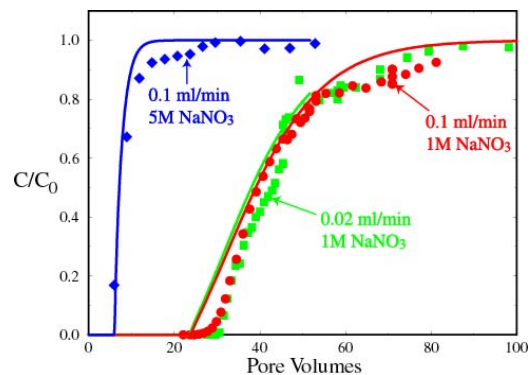


Figure 1:  $\text{Cs}^+$  breakthrough in Hanford sediment, influent =  $10^{-4}\text{M}$   $\text{CsNO}_3$ .

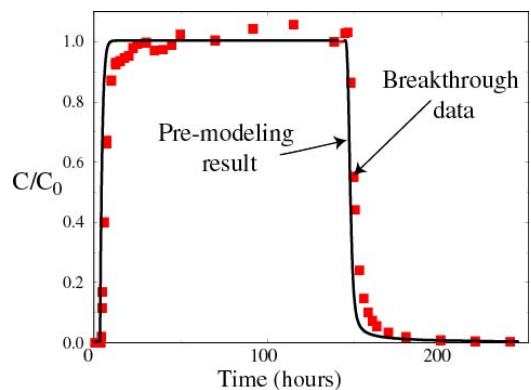


Figure 2:  $\text{Cs}^+$  breakthrough and desorption in Hanford sediment, influent = 5M  $\text{NaNO}_3$ .

This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

**References:**

[1] Zachara J.M., Smith S.C., Liu C., McKinley J.P., Serne R.J., Gassman P.L (submitted). *GCA*.

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