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004 EPA STAR Graduate Fellowship Conference *Next Generation Scientists—Next Opportunities*

olid-State NMR Studies of Hydroxyl-Promoted Kaolinite Dissolution under Simulated Tank Waste Conditions

Environmental Problem

he Vadose Zone at the Hanford Reservation R) in Richland, WA is contaminated with zardous and radioactive contaminants.

- e HR is one of the most contaminated sites in the world.
- clear weapons production led to the release of 1.7 trillion s of liquid waste to the ground at Hanford.
- % of waste storage tanks are known to have leaked or are ently leaking.
- ^{137}Cs and ^{90}Sr are the main contributors to site radioactivity.
- hough not used for human consumption, HR water feeds into Columbia River posing a threat to salmon habitats.

^{137}Cs/^{90}Sr fate and mobility may be affected mineral weathering processes at the site uck et al. 2004 and Chorover et al. 2003).

- eld and laboratory studies suggest that mineral weathering ctions produce mineral phases which can entrap Cs and Sr.
- rate-cancrinite, a Cs and Sr sorbent, was found in Hanford sludge.
- nited data exists pertaining to mineral weathering processes urring under HR geochemical conditions, thus more studies needed to better assess potential threats and to help develop er remediation and preventative strategies.

References

ck, E.C. and B.K. McNamara, *Environ. Sci. Technol.* **2004**, *38*, 1.

orover, J. et al., *Environ. Sci. Technol.* **2003,** *37*, 2200.

Hypothesis

- Hydroxyl-promoted dissolution of minerals in simulated tank waste leachates (STWL) lead to the formation of new phases that can influence the mobility of Cs and Sr in Hanford soils.

Research Plan

- Evaluate the weathering behavior of Kaolinite (K) in STWL.
  - Prepared batch reactors of simulated tank waste leachate with ^{133}Cs and ^{90}Sr and monitored kaolinite dissolution.
  - ^{27}Al nuclear magnetic resonance (NMR) spectroscopy, in conjunction with other spectroscopic and microscopic methods, was used to evaluate the weathering behavior of kaolinite and the formation of new mineral phases in reactors.
  - ^{27}Al NMR was used to assess the effects of ^{133}Cs and ^{87}Sr concentrations on neophase formation.
  - ^{27}Al NMR was used to quantify the relative amount of neophases forming in the reactor.
- Propose mechanisms of ^{133}Cs and ^{87}Sr removal from STWL leachate
  - ^{133}Cs and ^{87}Sr have different hydration properties and different solubilities thus, removal mechanisms are likely different.

Results

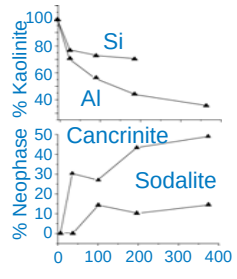

Fig. 1: ^{29}Si and ^{27}Al NMR of kaolinite dissolution in STWL (top); ^{27}Al NMR of cancrinite (C) and sodalite (S) formation in STWL (bottom).

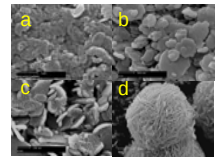

Fig. 2: SEM images:
(a) 0 days kaolinite,
(b) 1 day w/ S and K,
(c) 30 days w/ S, C, and K,
(d) Unidentified aluminum phase at 190 days.

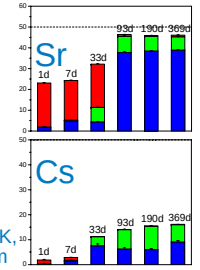

Fig. 3: Sr and Cs uptake from solution.

Conclusions

- Hydroxyl-promoted dissolution reactions of kaolinite are incongruent and lead to the formation of cancrinite and sodalite.
- The overall rate of dissolution is determined by Si release to solution.
- Sr removal is rapid and likely occurs via precipitation.
- Cs removal is gradual, occurs via an ion-exchange mechanism, and correlates to the formation of cancrinite and sodalite.
- Cs likely absorbs to cancrinite and sodalite phases.

Research Implications

- Knowledge gained from this simulated system can be combined with other studies to better decide how to allocate resources for the environmental management of this site.
- ^{137}Cs and ^{90}Sr are contaminants at the DOE Savannah River site, and as such, the results of this study should provide knowledge about the likelihood of radionuclide transport at this site.

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