Colloid-Facilitated Transport of Radionuclides Through the Vadose Zone

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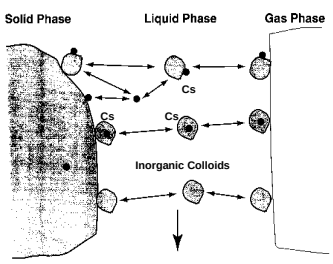
Introduction

Radioactive and hazardous waste stored in the underground tanks at the Hanford site has leaked into the vadose zone. Colloid-facilitated transport is not uncommon in the movement of contaminants in the environment. The potential for *in situ* formation of colloids and subsequent colloid-facilitated transport of radionuclides is high at the Hanford site. The extreme chemistry of tank waste supernatants, e.g. high pH, ionic strength, and aluminate concentration will result in dissolution of silica from native minerals in the sediments and subsequent formation of alumino-silicate colloids. These colloids would play an important role as carrier of radionuclides. Hence, in order to better understand the transport behaviors of radionuclides at the Hanford site and to develop more effective remediation technologies, the chemistry and physics of both contaminant-colloid interactions and their transport through the vadose zone bears detailed investigation.

Objectives

- Determine the structure, composition, and surface charge characteristics of colloidal particles formed under conditions typical of the Hanford tank leaks.
- Characterize the mutual interactions between colloids and contaminants, i.e. Cs.
- Evaluate mobility of colloids through soil under different degrees of water saturation and solution chemistry.
- Provide scientific basis for accurately modeling radionuclide movement at the Hanford site and developing of more effective remediation technologies.

Role of Mobile Colloids



Materials and Methods

- Hanford composite sediment was immersed in aluminate solution (100 mg/L Al in 0.1M NaOH solution). After various time intervals, the fine particles was collected, and organic matter/oxides was removed, followed by the extraction of < 2 μm colloids.
- Colloid Characterization: XRD, electrophoretic mobility and particle size (latter with Malvern Zetasizer).
- Colloid-Cs Interaction: Sorption study by batch technique.
- Column Studies: columns packed with Hanford sediment and sand are used for the study of colloid-facilitated transport of Cs under various conditions (water contents, electrolyte concentration, flow rate, pH).

Colloid Characterization

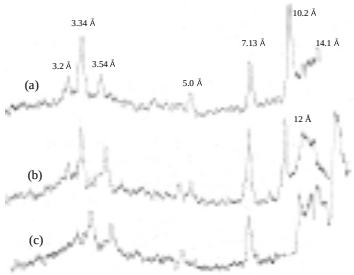


Figure 1. X-Ray Diffraction Patterns of Colloids extracted from Hanford Sediments after being reacted with aluminate solution: (a) unreacted; (b) after 1day at 25°C; (c) after 10 days at 50°C

Figure 1 shows that the alkaline aluminate solution significantly reacts with the 14-Å mineral (chlorite) extracted from the Hanford sediment. The average particle size of the resulting colloids range from 300 to 500 nm equivalent spherical diameter.

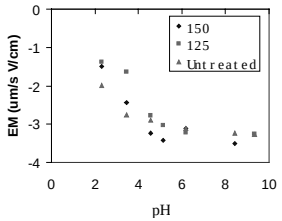


Figure 2. pH *vs* electrophoretic mobility (EM) of colloids extracted from Hanford Sediments after being reacted with aluminate solution (untreated colloids; 150: after 1 day at 50°C; 125: after 1 day at 25°C).

Cs Sorption

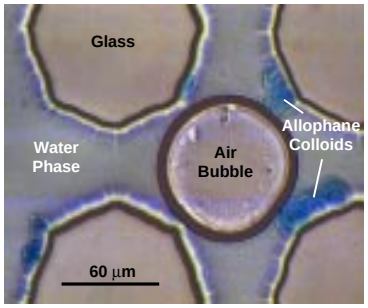
Table 1. Distribution Coefficients (K_d) of Cesium on Colloids Extracted from Hanford Site Sediments After Reacting with Aluminate Solution

Sample #	Description *	K_d (mL/g) #
0000	Unreacted	45671
1025	10 days at 25°C	33621
1050	10 days at 50°C	25970
2525	25 days at 25°C	29137
2550	25 days at 50°C	33622
Smectite(SWy-1)	Raw, < 2 μm, Na-, (CMS)	5057
Illite (Fithian, IL)	Raw, grounded, Na-, (Ward's Co)	420
Vermiculite(Enoree, SC)	Raw, grounded, Na-, (W.R. Grace Co)	4894

NOTE: * 100 g of Hanford composite sediment was immersed in 100 mL aluminate solution (100 mg/L Al in 0.1 M NaOH solution) at 25 and 50°C for 10 and 25 days, respectively.
K_d was determined at one-point concentration of Cs (2 mg/L, as $CsNO_3$), 40 mg in 25 mL distilled water, shaken for 6 hours, centrifuged and assayed by atomic emission spectrometry.
$ Equilibrium pH values fall in the range of 5.0-6.0

Table 1 indicates that, compared to model clay minerals, colloids extracted from Hanford Sediments after being reacted with aluminate solution still exhibit high affinity for cesium. This suggests that colloids at the Hanford Site might play an important role in radionuclide transport in the subsurface. The observed low sorption of illite and vermiculite is presumably due to their larger particle size. Results of the electrophoretic mobilities (**Figure 2**) of the colloids indicate that colloids are negatively-charged and likely tend to be quite mobile under the pH conditions at the Hanford Site.

Transport Study: (1) Micromodel

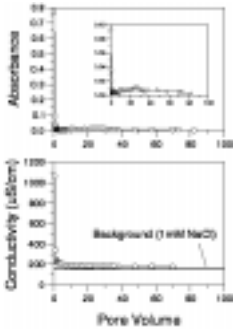


- Etched glass micromodel
- Allophane in 1mM NaCl, pH 7
- No sorption to liquid-gas interface

Transport Study: (2) Packed Column

- Hanford sediments sieved through 53 µm
- Elution with 1 mM NaCl
- Monitoring *in situ* mobilized colloids in outflow

In situ Mobilized Colloids



Conclusions

- The alkaline aluminate solution alters both mineralogical and chemical properties of the Hanford sediment colloids.
- Colloids exhibit high affinity for Cs, suggesting their potential as carriers of Cs during migration in vadose zone.
- Pretreatment and equilibration will be critical to performing transport experiments in packed columns.

Acknowledgments

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