

INTER-OFFICE MEMORANDUM

Memorandum Number: SRNL-TR-2009-00019

Date: 1/16/2009

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Re: Tc and Pu Distribution Coefficients, \mathbf{K}_{d} Values, for the Saltstone Facility Performance Assessment

Recently, Tc and Pu distribution coefficients, K_d values, were measured using site specific sediments and cementitious materials (Kaplan et al. 2008a; 2008b). These results were reported after issuing the last geochemical data package (Kaplan 2006), which contains most of the geochemical input values that will be used in the Saltstone Facility Performance Assessment. The purpose of this document is to recommend new K_d values based on the new site-specific results (Table 1). The best estimates for Tc K_d values for sandy and clayey sediments were 0.6 and 1.8 mL/g, respectively. This reflects a six and nine fold increase, respectively, compared to best estimates (Kaplan 2006). The best estimates for Pu concrete K_d values were increased by a factor of at least two.

Table 1. Recommended Tc and Pu K_d values.

Rad	Solid	Cement Stage ¹	K _d Conservative	K _d Best Estimate	Comments
Тс	Sand	N/A	0.2	0.6	Used the median value of 47 Tc K _d values,
Tc	Clay	N/A	0.2	1.8	including 24 K _d values recently measured from 24 depth-discrete samples from one borehole recovered from E-Area (Kaplan et al. 2008a). Recommending similar Tc K _d values for sandy and clayey sediments because clay Tc K _d values were not significantly greater than those of the sandy sediments.
Pu(III/IV)	Oxidizing Concrete	1	5000	10,000	Reducing concrete (including Saltstone) K _d values for Pu are 10,000 mL/g for stages 1,2, and 3. Oxidizing concrete (concrete without blast furnace slag) Pu K _d values are 10,000 for stages 1 and 2, and 1,000 for stage 3. These Pu results are described using site specific materials (Kaplan et al. 2008b – Tables 4 & 5; Kaplan et al. 2007 – Table 4). Other labs have measured minimal differences between Pu sorption under reducing and oxidizing conditions (Allard et al. 1987). The primary mode of Pu sorption to cementitious materials is by precipitation; hence solubility values are more representative than K _d values. Concrete Pu solubility values are reported in Kaplan (2006; Table 12).
		2	5000	10,000	
		3	5000	1000	
Pu(V/VI)	Oxidizing Concrete	1	5000	10,000	
	00.000	2	5000	10,000	
		3	5000	1000	
Pu(III/IV)	Reducing Concrete	1	5000	10,000	
		2	5000	10,000	
		3	5000	10,000	
Pu(V/VI)	Reducing Concrete	1	5000	10,000	
		2	5000	10,000	
		3	5000	10,000	
Pu(III/IV)	Sandy Sediment	N/A	200	300	Unlike concrete, sediment Pu K _d values are dependent on the oxidation state of Pu. Sediment K _d values have not changed from those reported by Kaplan (2006; Table 10). Aqueous Pu concentrations in sediments are primarily controlled by solubility; hence solubility values are more representative than K _d values. Soil Pu solubility values are reported in Kaplan (2006; Table 11). Calculations are to be conducted such that 99% of the Pu are Pu(III/IV) and 1% is Pu(V/VI). These percentages were taken from laboratory studies and reactive transport modeling (Kaplan et al. 2004).
	Clayey Sediment	N/A	3000	6000	
Pu(IV/V)	Sandy Sediment	N/A	11	16	
	Clayey Sediment	N/A	2500	5000	

¹ Concrete degradation is assigned three stages, or phases, during which porewater chemistry and concrete mineralogy are expected to be different. Stage 1 is the youngest stage during which calcium-silicate-hydrate gels harden and Portlandite. Stage 2 is in steady state with the calcium-silicate-hydrate gel and Portlandite; typically the longest stage. Stage 3 portlandite is fully dissolved or reacted and the solubility is control by the calcium-silicate-hydrate, eventually only leaving silica as the solubility control.

References

Allard, B., Eliasson, L., Hoglund, S., Andersson, K. 1984. Sorption of Cs, I, and Actinides in Concrete Systems. SKB Technical Report SKB/KBS TR-84-15, SKB, Stockholm, Sweden.

Kaplan, D. I., 2006. Geochemical Data Package for Performance Assessment Calculations Related to the Savannah River Site. WSRC-TR-2006-00004. Washington Savannah River Company, Aiken, SC.

Kaplan, D. I., B. A. Powell, D. I. Demirkanli, R. A. Fjeld, F. J. Molz, S. M. Serkiz, J. T. Coates. 2004. Enhanced Plutonium Mobility During Long-Term Transport Through an Unsaturated Subsurface Environment. Environ. Sci. Technol. 38:5053–5058.

Kaplan, D. I., Roberts, K., Shine, G., Grogan, D., Fjeld, R., Seaman, J. 2008a. Range and Distribution of Technetium K_d Values in the SRS Subsurface Environment. WSRC-STI-2008-00286, Rev. 1. Savannah River National Laboratory, Aiken, SC.

Kaplan, D. I., Roberts, K., Coates, J., Siegfried, M., Serkiz, S. 2008b. Saltstone and Concrete Interactions with Radionuclides: Sorption (K_d), Desorption, and Reduction Capacity Measurements. SRNS-STI-2008-00045. Savannah River National Laboratory, Aiken, SC.