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**Re: Concrete K_d Values Appropriate for the Tank Closure Performance
Assessment**

Abstract

Sorption K_d values of cementitious materials were measured for americium (Am), cadmium (Cd), cerium (Ce), cobalt (Co), cesium (Cs), mercury (Hg), iodine (I), neptunium (Np), protactinium (Pa), tin (Sn), strontium (Sr), technetium (Tc), uranium (U), and yttrium (Y). These measurements were conducted using 40-year old concrete, reducing grout, and an artificially aged reducing grout. Additionally, experiments were conducted under reducing and oxidizing conditions using simulants of young cement leachate (saturated $\text{Ca}(\text{OH})_2$) and old cement leachate (saturated CaCO_3). All measured K_d values followed expected geochemical trends.

The following general changes to the present K_d values of cementitious materials used in performance assessments (Kaplan 2006) are supported by this study. An important underlying principle in making these changes is that site-specific data is appreciably more reliable than literature data. Additionally, most of the reducing grout K_d measurements have never been performed. As such, the proposed changes to these K_d values will be based upon first-of-its-kind measurements, rather than by assuming chemical analogues, as has been past practices. The following are general changes to SRS performance assessment K_d values based on results of this study.

- Increase most Stage 1 & 2 (concrete during the two earliest stages of aging) K_d values under oxidizing conditions to approximately the measured values. The only K_d value to decrease was U, which was decreased from 1000 to 250 mL/g.
- Slightly increase all Stage 3 (concrete in its last stage of aging) values under oxidizing and reducing conditions. Measured values were all appreciably greater than the literature values, except U. However, the experimental conditions only captured early environmental conditions of Stage 3 and as such the experimental set up did not totally capture the anticipated evolution of environmental conditions expected over this cement aging stage.
- Increase most of the Stage 1 & 2 values under reducing conditions. This is to take account for the sorptive capacity provided by sulfide bonding that has traditionally not been included. Historically (Bradbury and Sarott 1995), K_d values for reducing grout were identical to those for regular (oxidized) cementitious materials, except that reducible contaminants, such as U and Tc, were assigned greater K_d values.

This memorandum was issued to provide early guidance for selecting K_d values of cementitious materials. A full report describing this work, as well as supporting research on plutonium and technetium sorption, and kinetic sorption and desorption studies will be issued.

Introduction

Concrete distribution coefficients (K_d values) were measured for several radionuclides in four environment conditions relevant to the Savannah River Site (SRS) performance assessments. The simulated environmental conditions were: 1) young oxidizing grout, 2) old oxidizing grout, 3) young reducing grout, and 4) old reducing grout. A detailed description of the mineralogy and aqueous chemistry of these four environments is presented in Kaplan (2006). It is important to note that this study does not address release of the radionuclides from the contaminated zone within a closed tank. Instead it deals with the geochemical parameters of the radionuclides once they are released into the aqueous phase. Radionuclide release from the sludge in the contamination zone is described by Denham (2007).

Scope and Objectives: The scope of this study was to measure the K_d /solubility values for the follow radionuclides and conditions:

- Radionuclides: Am, Cd, Ce, Co, Cs, Hg, I, Np, Pa, Sn, Sr, Tc, U, and Y,
- Cementitious Solid Phases: 40-year old aged concrete recovered from demolition activities at the SRS, reducing grout, and an artificially-aged reducing grout, and
- Aqueous phase chemistry: portlandite and calcite equilibrated conditions to simulate Stages 1 & 2, and Stage 3 in the cement lifecycle, respectively.

I, Tc, Np, and U were included in these tests because they were found to be important risk drivers in preliminary calculations. Am, Cd, Ce, Co, Cs, Hg, Sr, Sn, and Y were included because a great deal of additional information could be collected from these tests with relatively little additional effort, i.e., they could be measured together using a single cocktail of gamma-emitting isotopes. These gamma emitters were analyzed to provide information about the sorption of some radionuclides of interest to the performance assessment (Co, Ce, Cs, Am, Pa, Sr, and Y), as well as provide indirect information for surrogate radionuclides. For example, they provided insight into how monovalent cations (Cs) divalent cations (Hg, Co, and Sr), trivalent cations (Ce and Am), and tetravalent cations (Sn) sorb to cementitious materials.

This memorandum is intended to provide early guidance to modelers of the input values to use for the tank closure performance assessment. A more in depth report is being prepared. This report will provide:

- more background information about how the values reported in this memorandum were measured,
- new input values for Pu and Tc, which are presently being measured,
- desorption rates (kinetics) of radionuclides from cementitious materials,
- adsorption rates (kinetics) of radionuclides from cementitious materials, and
- kinetic information describing the rate that reducing grout converts an oxidizing system to a reducing system.

Materials and Methods

Materials

All radionuclides used in this study were purchased from Eckert & Ziegler Analytics (Atlanta Georgia), except ^{238}Pu , which originated from site activities and provided by David Hobbs (SRNL).

The reducing grout was prepared by Chris Langton (SRNL) and was identified as sample OPDEXE-X-P-O-BS. It contained 210 lbs/yd³ slag, 60 gallons/yd³ (500 lbs/yd³) of water, 75 lbs/yd³ of Portland cement, 375 lbs/yd³ fly ash, 2300 lbs/yd³ sand, 90 oz/yd³ of Adva-380, 275 g/yd³ of KelcoCrete, and 2.1 lbs/yd³ sodium-thiosulfate. This material was initially broken up with a chisel and hammer to ~1-cm sized particles. Then ~100 g of these particles were placed in a jaw crusher (Retsch Jaw Crusher Type BB51 with tungsten carbide plates) for ten minutes. The jaw crusher samples were then placed in a shatter box (Spec 8510 Shatterbox) for 10 seconds. The <1000- μm and >75- μm sieve fraction was used for these studies.

- The resulting particles had a Brunauer, Emmett, Teller (BET) surface area of $7.11 \pm 0.02 \text{ m}^2/\text{g}$ (2 replicates, 5-point isotherm).
- A 1:1, water: reducing grout suspension had a pH of 11.16 and an Eh (redox potential) of 36.5 mV.

An aged reducing grout was created by bubbling aerated water for one week into a beaker containing 30 g of the ground reducing grout.

- A 1:1, water: reducing grout suspension had a pH of 11.24 and an Eh of 325.3 mV.

This high platinum electrode Eh value, compared to that of the fresh reducing grout Eh value noted above was appreciably greater, suggesting that the material had in fact been oxidized.

The aged concrete was approximately 40-years old and was cored from a building pad located on the SRS. To separate the aggregate from the cement, a portion of the core was crushed with a hammer and then the cement was loosened from the aggregate with the use of an awl and screw driver. Separation of these two phases within the concrete was relatively easy to accomplish; the task took about one hour. After separation, the cement phase was broken up into smaller more uniform particles with a Spex 8510 Shatterbox. The <1000 μm and >75 μm sieve fraction was used for these experiments. It is important to note that the cement and not the aggregate was used in these measurements.

- The resulting particles had a BET surface area of $5.88 \pm 0.02 \text{ m}^2/\text{g}$ (2 replicates, 5-point isotherm).
- A 1:1 water:concrete suspension had a pH of 11.99 and an Eh of 346.6 mV.

Ca(OH)₂-Saturated Leaching Solution:

Background: This solution was used in the sorption tests to simulate the first and second stage of cement aging.¹ During these cement aging stages, portlandite (Ca(OH)₂), and calcium-silicate-hydrate gels are the key solid phases controlling aqueous leachate chemistry.

Because CO₂ in air is very soluble in water at high pH and the resulting dissolved carbonate will precipitate as calcite in the Ca(OH)₂- saturated solution, care was taken to minimize contact of the solution with atmospheric air. Excess solid Ca(OH)₂ was undesirable because it would buffer the pH at an abnormally high pH level.

Procedure: About 1-L of D.I. water was deoxygenated by boiling and then purge with Ar gas for 30 min. The deoxygenated water was then placed in a Teflon bottle (because this material has a low air diffusion coefficient) within an Ar atmosphere. A sufficient quantity of fresh Ca(OH)₂ (~1.26 g/L @ 25°C) was added to the deoxygenated water to just saturate the solution. To minimize exposure to air, the head space was purged with Ar. Additionally, it was necessary to periodically skim off precipitates that formed at the water/air interface.

Calcite-Saturated Leaching Solution:

Background: This solution was used in the sorption tests to simulate the third stage of cement aging. During this stage, portlandite has been fully dissolved/reacted and the solubility or reactions of the calcium-silicate-hydrate gel with the infiltrating water controls the pH of the cement pore-water/leachate. The pH continues to decrease until it reaches the pH of the background sediment, pH 5.5. Radionuclides tend to sorb least during this stage.

¹ Cement is described in the performance assessment as aging in three progressive stages. These stages have unique mineralogy and leachate chemical conditions. How cement aging is treated in the performance assessment is described in Kaplan (2006).

By preparing the solution at a slightly elevated temperature the possibility of calcite precipitation during the test at room temperature will be minimized. (Calcite undergoes retrograde solubility.) There was no need to minimize contact of this solution with the atmosphere. Calculated equilibrium values for this solution are: pH = 8.3, $\text{Ca}^{2+} = 20 \text{ mg/L}$, TIC = 58 mg/L, $\text{PCO}_2 = 0.0003 \text{ atm}$ (fixed).

Procedure: Added 1 L of DI water on a stir/heating plate and raised the temperature of the solution by 3 to 10 °C from ambient room temperature. As the solution was being stirred with a stir bar, 5.6 mg of CaCO_3 was added to the heated DI water. The mixture was left on the stir/heating plate for 24 hr. This suspension was then passed through a 0.45- μm membrane.

Methods

There were four near identical experiments conducted, in which different radionuclides were added to the three different cementitious materials:

1. ^{237}Np , ^{125}I and ^{235}U ,
2. ^{99}Tc ,
3. ^{238}Pu (results not reported in this memorandum),
4. a suite of gamma-emitting radionuclides, $^{109}\text{Cd(II)}$, $^{60}\text{Co(II)}$, $^{139}\text{Ce(III)}$, $^{230}\text{Hg(II)}$, $^{113}\text{Sn(IV)}$, $^{137}\text{Cs(I)}$, $^{241}\text{Am(III)}$ and $^{89}\text{Sr(II)}$.

Radionuclides were added in these groupings to ease analysis. Each of these experiments was conducted in an oxidizing environment (on the lab counter top) and a reducing environment (within an Ar-atmosphere).

Batch sorption tests were conducted in a suspension including 0.5 g solid (reducing grout, aged reducing grout, and aged cement) and 12 mL solution (calcite-saturated or Ca(OH)_2 -saturated). Prior to adding the radiological spike solutions, each solid was pre-equilibrated with two 12-mL aliquots of their respective solution phases. After the one week equilibration period, a third 12-mL aliquot was added, and the resulting suspension was then spiked with $\sim 330 \mu\text{L}$ of a stock radiological solution. All spiked solutions, except Tc and I, was then pH adjusted by adding $\sim 500 \mu\text{L}$ of 1 M NaOH. This was necessary because the background solutions for the spikes were acidic. Spiked suspensions were mixed twice daily by vigorously shaking for ~ 10 seconds. At the end of this spike-equilibration period, the solids were permitted to settle for 1 hour and then the aqueous phase of each tube was passed through a 0.1- μm filter. These solutions were then analyzed for their respective radionuclides using traditional analytical techniques.

All tests were conducted in triplicate. A positive control was included with each set of experiments. These were “no-solids” control samples that were used to determine if precipitation of the radionuclides occurred during the experiments. The extent of sorption, K_d value, was calculated using Eq. 1, which calculates sorption by subtracting the concentration of radionuclide added from the concentration remaining in solution at the end of the one-week equilibration period.

$$K_d = \frac{(C_i - C_f) \times V}{C_f \times m_{\text{soil}}} \quad (\text{Eq. 1})$$

where C_i and C_f are the aqueous concentrations of the initial spike and the final equilibrium concentration of each radionuclide (Ci/mL); V = volume of liquid in the final equilibrated suspension (mL); and m_{soil} is the soil mass (g).

Results

Before discussing the results, it is important to mention that the K_d values, as they are defined in Eq. 1, include not only adsorption or absorption, but also (co)precipitation. Some (co)precipitation of the radionuclides was noted in the control samples that did not include any solids. That is to say, aqueous radionuclide concentration decreases were observed in the absence of solids. Therefore the measured K_d values, did not describe only readily reversible sorption reactions, as is implicitly implied by this construct. Instead, this process and (co)precipitation was measured; (co)precipitation tends to have a much faster forward reaction (precipitation), than a backward reaction (dissolution). The implications of this are that the resulting K_d values when used in reactive transport modeling will underestimate the true retardation of the contaminant. Furthermore, these would be considered conservative values for the groundwater risk pathway.

Measured K_d results are presented in Table 1. There are some general trends in this data.

- K_d values generally increased in the following order of valence state:

$$\begin{aligned} -I (I, \text{TcO}_4^-) < +I (\text{Cs}^+) < +II (\text{Cd}^{2+}, \text{Co}^{2+}, \text{Hg}^{2+}, \text{Sr}^{2+}, \text{and } \text{UO}_4^{2+}) \\ < +III (\text{Am}^{3+}, \text{Ce}^{3+} \text{ and } \text{Y}^{3+}) = +IV (\text{Sn}^{+4}) \end{aligned} \quad (2)$$

This trend is largely consistent with geochemical principles.

- K_d values in the reducing grout were generally much larger than those in the basemat or aged reducing grout. Some notable higher reducing grout K_d values, as compared to the two oxidized grouts, are Am, Cd, Ce, Co, Pa, Sn, U, and Y. The exceptionally high reducing grout K_d values for Cd, Sn, and all three trivalents (Am, Ce, and Y), are likely due to the presence of sulfide in the reducing grout, and their absence in the oxidized grouts.
- For the radionuclides which have varying oxidation states (Np, Pa, and U; no data for Tc), K_d values were appreciably greater with the reducing grout than the basemat or the aged reducing grout. This trend was anticipated.
- Greater K_d values were generally measured in the $\text{Ca}(\text{OH})_2$ systems, the “younger” cement scenario, than in the calcite-saturated system, the “older” cement scenario. Again, this trend was expected.

The existing oxidizing and reducing cementitious K_d values used in past performance assessment calculations are presented in Table 2. These are literature K_d values, originating primarily from Bradbury and Sarott (1995) and Krupka and Serne (1998). Previously, no sorption experiments were conducted with SRS cementitious materials. Furthermore, there have been very few sorption experiments conducted with reducing cementitious solids at SRS or elsewhere. It is commonly assumed that radionuclide sorption to reducing cementitious solids is the same as that of oxidizing cementitious solids, except for those radionuclides that sorb more strongly in a reduced state, such as Pu(IV), Tc(IV), and U(IV) (Bradbury and Sarott 1995, Krupka and Serne 1998). It is also commonly

assumed that sorption in Stages 1 and 2 is generally greater than sorption in Stage 3 of cementitious solids (Bradbury and Sarott 1995, Krupka and Serne 1998). Both of these assumptions are reflected in the values reported in Table 2.

A comparison of the measured K_d values to the literature K_d values presently used in the performance assessment is presented in Table 3. The measured K_d values were similar to the literature values for the Oxidizing Cementitious Solids/Stages 1 & 2, except for Cd, Cs, and Sr. For these exceptions, the measured K_d values were appreciably greater than the literature values. The literature K_d values for the Oxidizing Cementitious Solids/ Stage 3 are all consistently less than the measured values. The cause for this difference may be that there are few experimental results of “aged” cements and therefore there is little information by which scientists can predict sorption at this stage. Also, the experimental set up for the Stage 3 (i.e., the basemat in CaCO_3 solution) does not exactly match the conceptual environment of Stage 3. Notably, the mineral assembly in the samples used for these measurements has not been weathered (aged) for thousands of years.

For the Reducing Cementitious Solids, literature K_d values were almost all less than those measured in this study, except for uranium and iodine. As mentioned above, there has been little research evaluating the extent of solute sorption to reducing grouts. The few studies that have been conducted have been limited to investigations of reducible contaminants, such as Tc(VII), Pu(V/VI), and U(VI). The extent of greater sorption by non-reducible contaminants has not been evaluated. The presence of sulfides in the reducing grout would be expected to increase the sorption of soft transition metals (metals towards the bottom of the periodic chart), such as Cd, Hg, and Sn, and would have relatively little effect on the sorption of hard metals, such as Cs, Sr, and U(VI). As was the case with the Oxidizing Cementitious Solids in Stage 3, the literature K_d values for the Reducing Cementitious Solids in Stage 3 were less than the measured values. Again, the likely cause for this is: 1) the SRS reducing grout formulation is different than those previously used in sorption experiments (primarily the amount and type of slag used is unique), and 2) there has been an unwritten assumption that enhanced sorption to reducing cementitious solids is only due to favorable redox changes in the contaminant, and enhance sorption to the presence of sulfides were not accounted for.

K_d values measured in saturated Ca(OH)_2 were all greater than those listed as Reducing Cementitious Solids in the 3rd Stage (Table 3). Finally, it needs to be recognized that the experimental K_d results include both (ad/ab)sorption and precipitation. In some cases, such as Am, Pa, U, Np, Sn, and Y, appreciable amount of precipitation likely occurred.

Conclusions

The following general changes to the K_d values of cementitious solids in Kaplan (2006) are supported by this study. An important underlying principle in making many of these suggested changes is that site-specific data is appreciably more accurate than literature data.

- Increase most Stage 1 & 2 values under oxidizing conditions to approximately the measured values. The only important exception is for U, which will be decreased from 1000 to 250 mL/g.
- Slightly increase all Stage 3 values, except U, under oxidizing and reducing conditions. As discussed above, the measured values were all appreciably greater than the literature values,

except U in Stage 3. The literature K_d values were based on little experimental data. These measured values greatly increase the pool of existing literature reducing-grout K_d values.

- Increase most of the Stage 1 & 2 values under reducing conditions. This is to account for the sorptive capacity provide by sulfide bonding. Sorption values for iodine will not be changed, and those for Cs and U will be moderately reduced.

These recommended changes are presented in Table 4.

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Krupka, K. M., and R. J. Serne. 1998. Effects on Radionuclide Concentrations by Cement/Ground-Water Interactions in Support of Performance Assessment of Low-Level Radioactive Waste Disposal Facilities. NUREG/CR-6377. U.S. Nuclear Regulatory Commission, Washington, DC.

Table 1. K_d values appropriate for young ($\text{Ca}(\text{OH})_2$) and old (CaCO_3) cementitious materials (mean and standard deviation of triplicate measurements; units = mL/g).

	Basemat - K_d				Aged Reducing Grout- K_d				Reducing Grout K_d			
	$\text{Ca}(\text{OH})_2$		CaCO_3		$\text{Ca}(\text{OH})_2$		CaCO_3		$\text{Ca}(\text{OH})_2$		CaCO_3	
	avg	stdev	avg	stdev	avg	stdev	avg	stdev	avg	stdev	avg	stdev
Am(III)	6031	2145	4112	3742					42,887	29,994	17,113	19,602
Cd(II)	9907	3685	3180	1445					297,300	5293	17,510	13,838
Ce(III)	6003	2014	4652	4552					103,840	112,245	4564	1776
Co(II)	4343	969	3994	2888					15,065	10,665	6539	3137
Cs(I)	21	5.4	17.6	4.1					-2.3 ^(a)	0.7	15	2.2
Hg(II)	289	9.8	568	191					1173	328	1095	957
I(-I)	14.8	4.9	14.4	7.3	22.9	4.4	24.3	2.9	6.2	0.2	11	6.0
Np(V) ^(b)	1652	1101	1318	764	644.3	37.9	3265	3250	3949	565	2779	2000
Pa(V) ^(b)	1630	4.9	1074	303	1018	33	24,618	12,729	9890	4589	8049	6577
Sn(IV)	3900	63	3360	2579					71,762	8983	5520	4015
Sr(II)	28.1	2.8	39.1	4.2					2.9	0.3	54.6	9.9
Tc(-VII) ^(b)	0.8	1.2	1.4	5.8	7.0	0.3	4.9	0.1	(c)	(c)	(c)	(c)
U(VI) ^(b)	259	189	165	67	83.1	2.3	333.3	98.6	2489	2354	3182	2704
Y(III)	4830	1230	4738	4596					64,821	75,544	5336	2411

^(a) For these experiments, negative K_d values should be assumed to be equal to 0 mL/g, and are likely attributed to analytical error.

^(b) Np(V), Pa(V), Tc(VII) and U(VI) were added as NpO_2^+ , PaO_2^+ , TcO_4^- , and UO_2^{2+} respectively.

^(c) Tc K_d measurements in reducing grout had to be redone because O_2 had entered the experimental system, thereby compromising the value of the data.

Table 2. Literature K_d values presently listed in the performance assessment's geochemical data package (Kaplan 2006; units = mL/g).

	Oxidizing Cementitious Solids ^(a)		Reducing Cementitious Solids ^(a)		Comments
	Cement Stage		Cement Stage		
	1 & 2	3	1 & 2	3	
Am(III)	5000	500	(b)	(b)	Not included in Kaplan (2006). Values assumed equal to that reported for Pb.
Cd(II)	500	250			
Ce(III)	5000	500			
Co(II)	1000	500			
Cs(I)	2 – 4 ^(c)	2			Not included in Kaplan (2006). Values assumed equal to that reported for Pb.
Hg(II)	500	250			
I(-I)	8 - 20	0			
Np(V) ^(d)	2000	200			
Pa(V) ^(d)	2000	200			
Sn(IV)	4000	2000			
Sr(II)	0.2 – 0.5	0.8			
Tc(-VII) ^(d)	0	0	5000	5000	
U(VI) ^(d)	1000	70	5000	5000	Not included in Kaplan (2006). Values assumed equal to other +3 solutes.
Y(III)	5000	500			

^(a) K_d values for Oxidizing Cementitious Solids are the "Best" estimates for Stage 3 taken from Table 13 in Kaplan (2006). K_d values for Reducing Cementitious Solids are the "Best" estimates for Stages 1&2 taken from Table 14 in Kaplan (2006).

^(b) K_d values not listed for the Reducing Cementitious Solids are the same as those reported for the Oxidizing Cementitious Solids.

^(c) Ranges of K_d values exist where the proposed K_d values reported in Kaplan (2006) for Stage 1 differ from those for Stage 2. All single K_d entries indicate that the K_d values for Stages 1 and 2 are the same.

^(d) Np(V), Pa(V), Tc(VII) and U(VI) were added as NpO_2^+ , PaO_2^+ , TcO_4^- , and UO_2^{2+} respectively.

Table 3. Comparison of the measured K_d values to the literature K_d values presently used in the performance assessment (units = mL/g).

	Oxidizing Cementitious Solids				Reducing Cementitious Solids			
	Stages 1 & 2		Stage 3		Stages 1 & 2		Stage 3	
	Kaplan (2006) ^(b)	Measured	Kaplan (2006) ^(b)	Measured	Kaplan (2006) ^(b)	Measured	Kaplan (2006) ^(b)	Measured
Am(III)	5000	6031	500	4112	5000	42,887	500	17,113
Cd(II)	500	9907	250	3180	500	297,300	250	17,510
Ce(III)	5000	6003	500	4652	5000	103,840	500	4564
Co(II)	1000	4343	500	3994	1000	15,065	500	6539
Cs(I)	2 – 4 ^(a)	21	2	17.6	2 – 4 ^(a)	-2.3 ^(c)	2	15
Hg(II)	500	289	250	568	500	1173	250	1095
I(-I)	8-20	14.8	0	14.4	8-20	6.2	0	11
Np(V) ^(d)	2000	1652	200	1318	2000	3949	200	2779
Pa(V) ^(d)	2000	1630	200	1074	2000	9890	200	8049
Sn(IV)	4000	3900	2000	3360	4000	71,762	2000	5520
Sr(II)	0.2 – 0.5 ^(a)	28.1	0.8	39.1	0.2 – 0.5 ^(a)	2.9	0.8	54.6
Tc(-VII) ^(d)	0	0.8	0	1.4	5000	NA	5000	NA
U(VI) ^(d)	1000	259	70	165	5000	2489	5000	3182
Y(III)	5000	4830	500	4738	5000	64,821	500	5336

^(a) Ranges of K_d values exist where the proposed K_d values in Stage 1 differ from those in Stage 2. All single K_d entries indicate that the K_d values for Stages 1 and 2 are the same.

^(b) K_d values for Oxidizing Cementitious Solids are the "Best" estimates for Stage 3 taken from Table 13 in Kaplan (2006). K_d values for Reducing Cementitious Solids are the "Best" estimates for Stages 1&2 taken from Table 14 in Kaplan (2006).

^(c) For these experiments, negative K_d values should be assumed to be equal to 0 mL/g, and are likely attributed to analytical error.

^(d) Np(V), Pa(V), Tc(VII) and U(VI) were added as NpO_2^+ , PaO_2^+ , TcO_4^- , and UO_2^{2+} respectively.

Table 4. Comparison of new (this report) and old (Kaplan 2006) “Best Estimate” values (units = mL/g).

	Oxidizing Cementitious Solids				Reducing Cementitious Solids			
	Stages 1 & 2		Stage 3		Stages 1 & 2		Stage 3	
	Kaplan (2006) ^(b)	Recommended “Best” Estimate	Kaplan (2006) ^(b)	Recommended “Best” Estimate	Kaplan (2006) ^(b)	Recommended “Best” Estimate	Kaplan (2006) ^(b)	Recommended “Best” Estimate
Am(III)	5000	6000	500	600	5000	5000	500	1000
Cd(II)	500	5000	250	500	500	5000	250	1000
Ce(III)	5000	6000	500	600	5000	5000	500	1000
Co(II)	1000	4000	500	1000	1000	5000	500	1000
Cs(I)	2 – 4 ^(a)	2 – 20	2	10	2 – 4	0 – 2	2	10
Hg(II)	500	300	250	300	500	1000	250	300
I(-I)	8-20	8 – 15	0	4	8 – 20	2 – 10	0	4
Np(V) ^(c)	2000	1600	200	250	2000	3000	200	300
Pa(V) ^(c)	2000	1600	200	250	2000	5000	200	500
Sn(IV)	4000	4000	2000	2000	4000	5000	2000	2000
Sr(II)	0.2 – 0.5	3 – 30	0.8	15	0.2 – 0.5	0.5 – 3.0	0.8	20
Tc(-VII) ^(c)	0	0.8	0	0.5	5000	NA	5000	NA
U(VI) ^(c)	1000	250	70	70	5000	2500	5000	2500
Y(III)	5000	5000	500	500	5000	5000	500	1000

^(a) Ranges of K_d values exist where the proposed K_d values in Stage 1 differ from those in Stage 2. All single K_d entries indicate that the K_d values for Stages 1 and 2 are the same.

^(b) K_d values for Oxidizing Cementitious Solids are the "Best" estimates for Stage 3 taken from Table 13 in Kaplan (2006). K_d values for Reducing Cementitious Solids are the "Best" estimates for Stages 1&2 taken from Table 14 in Kaplan (2006).

^(c) Np(V), Pa(V), Tc(VII) and U(VI) were added as NpO_2^+ , PaO_2^+ , TcO_4^- , and UO_2^{2+} respectively.