LA-12956-MS

Radionuclide Sorption in Yucca Mountain Tuffs with J-13 Well Water: Neptunium, Uranium, and Plutonium

Yucca Mountain Site Characterization Program Milestone 3338

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Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

This work was supported by the Yucca Mountain Site Characterization Project Office as part of the Civilian Radioactive Waste Management Program of the U.S. Department of Energy. The Los Alamos data tracking number for this record package is LA000000000104.001.

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LA-12956-MS

UC-814
Issued: August 1996

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by

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ABSTRACT

We studied the retardation of actinides (neptunium, uranium, and plutonium) by sorption as a function of radionuclide concentration in water from Well J-13 and of tuffs from Yucca Mountain. Three major tuff types were examined: devitrified, vitric, and zeolitic. To identify the sorbing minerals in the tuffs, we conducted batch sorption experiments with pure mineral separates. These experiments were performed with water from Well J-13 (a sodium bicarbonate groundwater) under oxidizing conditions in the pH range from 7 to 8.5. The results indicate that all actinides studied sorb strongly to synthetic hematite and also that Np(V) and U(VI) do not sorb appreciably to devitrified or vitric tuffs, albite, or quartz. The sorption of neptunium onto clinoptilolite-rich tuffs and pure clinoptilolite can be fitted with a sorption distribution coefficient in the concentration range from 1×10^{-7} to 3×10^{-5} M. The sorption of uranium onto clinoptilolite-rich tuffs and pure clinoptilolite is not linear in the concentration range from 8×10^{-8} to 1×10^{-4} M, and it can be fitted with nonlinear isotherm models (such as the Langmuir or the Freundlich Isotherms). The sorption of neptunium and uranium onto clinoptilolite in J-13 well water increases with decreasing pH in the range from 7 to 8.5. The sorption of plutonium (initially in the Pu(V) oxidation state) onto tuffs and pure mineral separates in J-13 well water at pH 7 is significant. Plutonium sorption decreases as a function of tuff type in the order: zeolitic > vitric > devitrified; and as a function of mineralogy in the order: hematite > clinoptilolite > albite > quartz.

INTRODUCTION

The retardation of actinides by sorption onto tuffs is of major importance in assessing the performance of a potential high-level nuclear waste repository at Yucca Mountain. We have studied the sorption of actinides (neptunium, uranium, and plutonium) in Yucca Mountain tuffs and pure minerals in water from Well J-13 (under oxidizing conditions) in the pH range from 7 to 8.5. Neptunium (Nitsche et al. 1993) and uranium (Wanner and Forest 1992) have a relatively high solubility (on the order of 10^{-4} M) in a sodium bicarbonate groundwater (such as the J-13 well water) under oxidizing conditions. The high solubility of neptunium and uranium combined with their limited sorption (Thomas 1987) onto Yucca Mountain tuffs makes these radionuclides a high priority within the sorption studies of the Yucca Mountain Site Characterization Project (Meijer 1992). Our main objective in this study was to characterize the sorption of neptunium and uranium in Yucca Mountain

tuffs and J-13 water (under oxidizing conditions) as a function of radionuclide concentration.

Plutonium has been reported to sorb significantly onto Yucca Mountain tuffs in J-13 well water (Thomas 1987). The experiments reported by Thomas were performed with a plutonium solution in which the initial oxidation state was Pu(IV). Nitsche et al. (1993) reported that the dominant oxidation state of plutonium in J-13 water in the pH range from 7 to 8.5 is Pu(V). Consequently, an additional objective of our experiments was to characterize the sorption of plutonium in J-13 water with the plutonium initially in the Pu(V) oxidation state.

EXPERIMENTAL PROCEDURES FOR SORPTION EXPERIMENTS

J-13 Groundwater

We obtained the water for our experiments from Well J-13. This groundwater is predominately a sodium bicarbonate water. Other cations are calcium, potassium, and magnesium; other anions are sulfate, chloride, nitrate, and fluoride (see Fig. 1). The final major constituent is silica. Ogard and Kerrisk (1984) reported J-13 well water to be oxidizing; consequently, we performed all our batch sorption experiments under oxidizing conditions.

The J-13 water used in our sorption experiments was collected at the well site in Nevada, sent to

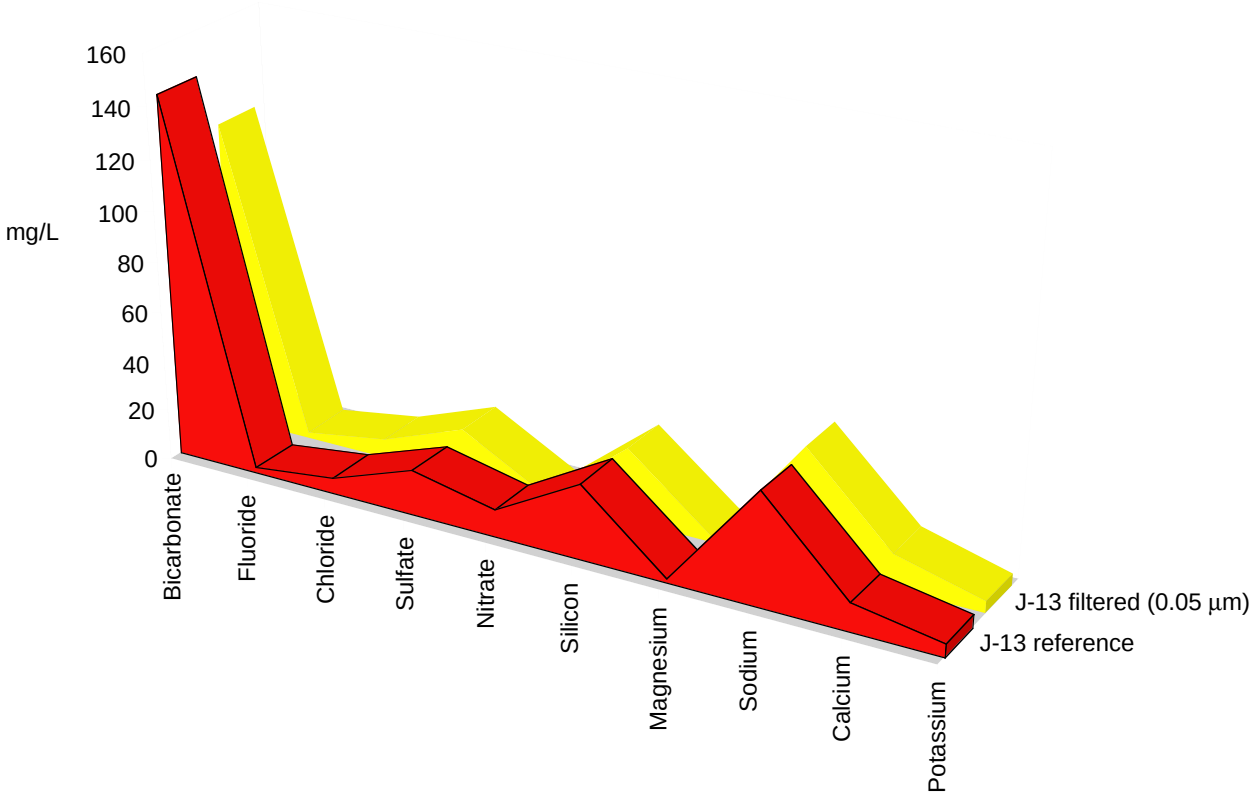

Figure 1. Water Chemistry of J-13 Well Water. "J-13 reference" is water analyzed at the Nevada well site (data recorded in Ogard et al. 1984). "J-13 filtered" is water analyzed at Los Alamos, NM, after being passed through a 0.05-μm filter (data recorded in binder TWS-INC-11-93-32, pages E24–E25).

Los Alamos, NM, and passed through a 0.05-μm filter prior to its use in the sorption experiments. The chemistry of this water is compared in Fig. 1 to the J-13 reference data obtained on site by Ogard and Kerrisk (1984). The pH of on-site J-13 water is ~7; the pH of J-13 water received at Los Alamos has increased to 8.5 because of CO_2 evolution as the water equilibrates with the higher-elevation atmosphere. No further changes seem to occur.

Minerals and Tuff Samples

The minerals used for the batch sorption experiments (the results of which are reported in Appendix A) were synthetic hematite, clinoptilolite, quartz, and albite. The synthetic hematite was commercially available Fe_2O_3 from EM Science. The origins of the natural minerals are given in Table 1.

We obtained the tuff samples used in the sorption experiments from drill holes at Yucca Mountain and labeled each sample with drill-hole number and drill depth in feet. For example, G4-268 refers to a tuff sample obtained from drill hole USW G-4 at a depth of 268 feet. Bish and Chipera (1989) have reported the location of the various drill holes.

We determined the mineralogy of the three types of tuff used in these experiments by x-ray-diffraction (XRD) analysis, and the results are shown in Fig. 2. Details of these data were reported previously (Bish and Chipera 1989; Chipera and Bish 1989 and 1994). The minerals used in the sorption experiments were more than 95% pure.

We determined the surface area of the tuff and minerals by BET (Brunauer, Emmett, and Teller) analysis, after crushing and wet-sieving all the tuffs, the quartz, and the albite samples to obtain particles in the size range from 75 to 500 μm (details of the procedures are given in LANL-CST-DP-63, *Yucca Mountain Project Detailed Procedures*, Los Alamos National Laboratory). We did not sieve or crush the synthetic hematite; we crushed and then purified the clinoptilolite samples but did not sieve them. The measured surface areas are given in Fig. 3.

The three major rock types that we used for sorption experiments were zeolitic, vitric, and devitrified. As shown in Fig. 2, the major component of zeolitic tuff, represented by samples G4-1503, G4-1510, and G4-1529, is clinoptilolite. The major component of vitric tuff, represented by sample GU3-1405, is glass. The major component of devitrified tuff, represented by samples G4-268 and G4-272, is alkali feldspar.

Neptunium, Uranium, and Plutonium Solutions

The neptunium solutions used for the batch sorption experiments were prepared by taking an aliquot of a well-characterized $^{237}Np(V)$ acidic stock and diluting it in the groundwater being studied. Nitsche et al. (1993) reported the solubility

Table 1. Natural Minerals for Sorption Experiments

Sample ID Code	Description	Origin
G	Purified clinoptilolite	Castle Creek, Idaho
G1	Purified clinoptilolite exchanged to obtain the sodium form	Castle Creek, Idaho
M	Quartz	Hot Springs, Arkansas
W	Albite	Unknown

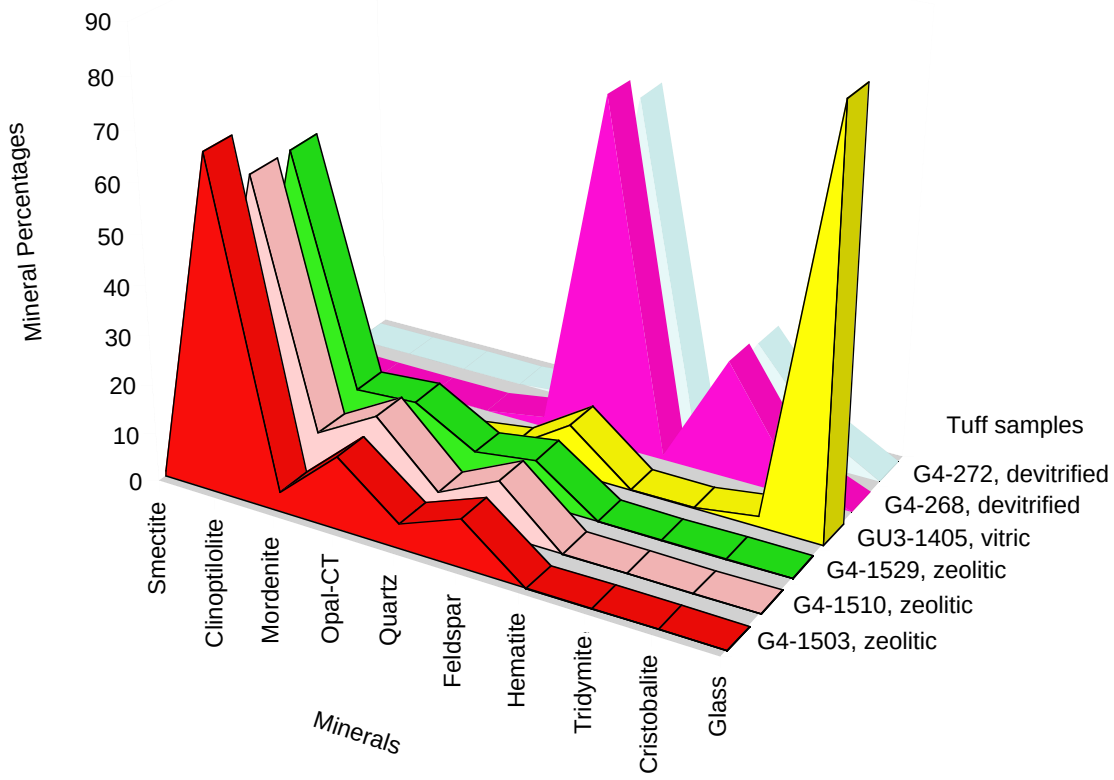

Figure 2. Mineralogy of Yucca Mountain Tuffs. Mineral percentages for the tuffs used in the sorption experiments were determined by x-ray diffraction. Each tuff, except sample GU3-1405, was wet sieved with J-13 well water to particle sizes ranging from 75 to 500 micrometers.

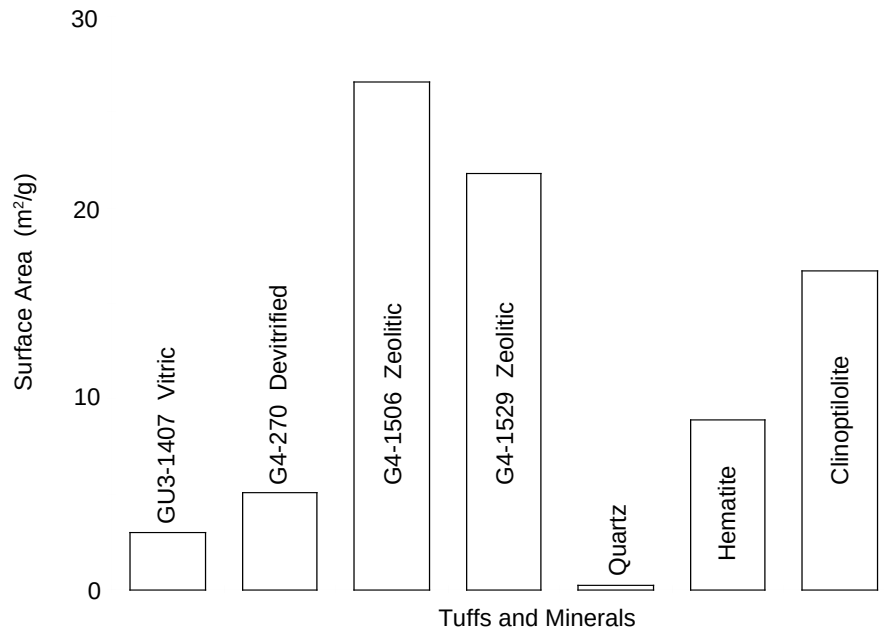

Figure 3. Surface Areas. The surface areas of tuffs and minerals used in the sorption experiments were determined by BET analysis. Each tuff or mineral, except the synthetic hematite and the clinoptilolite, was wet sieved with J-13 well water to obtain particle sizes ranging from 75 to 500 micrometers.

and speciation of neptunium in J-13 water at room temperature for pH values of 7 and 8.5. These data are summarized in Table 2.

The uranium solutions we used were prepared by taking an aliquot of an acidic natural-uranium standard (Product No. 5753 from J.T. Baker) and diluting it in J-13 groundwater. According to published data on the chemical thermodynamics of uranium (Wanner and Forest 1992), uranium exists as U(VI) in water from Well J-13 (under oxidizing conditions).

The plutonium solutions we used were prepared by taking an aliquot of a well-characterized Pu(V) acidic stock and diluting it in water from Well J-13. Pu(V) in the concentrated acidic stock disproportionates into Pu(IV) and Pu(VI); consequently, dilution of the Pu(V) acidic stock was performed immediately after preparation of the stock solution. Nitsche et al. (1993) reported the solubility and oxidation state of plutonium in J-13 water at room temperature for pH values of 7 and 8.5. These data are summarized in Table 3.

Batch Sorption Procedure

We performed all batch sorption experiments reported in Appendix A at room temperature using the following procedure. The solid phase was pretreated with J-13 groundwater in the ratio of 1 g of solid to 20 mL of solution and, after equilibration, was separated from the groundwater by centrifugation. The solid phase was then treated with a radionuclide solution of J-13 well water (again 1 g of solid to 20 mL of solution) and, after sorption and equilibration, was likewise separated from the solution by centrifugation. We measured both the initial amount of radionuclide in solution and the amount that remained in solution after sorption. The difference between these two values represents the amount of radionuclide that remained in the solid phase.

Table 2. Solubility and Speciation of Np(V) in J-13 Groundwater*

pH	Solubility (M)	NpO_2^+	$NpO_2CO_3^-$
7	1.3×10^{-4}	46%	54%
8.5	4.4×10^{-5}	38%	62%

*From Nitsche et al. 1993.

We performed the uranium analysis by inductively coupled plasma mass spectrometry (ICP-MS) and the analysis of ^{237}Np and ^{239}Pu with a liquid scintillation counter (Packard tri-carb 2550-TR/AB). The liquid-scintillation counting technique is capable of discriminating alpha from beta activity; consequently, no interference from ^{233}Pa (the daughter of ^{237}Np) is expected. The efficiency of our liquid-scintillation counter is approximately 100%, so the counts per minute (cpm) reported in Appendix A for ^{237}Np and ^{239}Pu are approximately equivalent to disintegrations per minute.

As controls, we used container tubes without any solid phase in them to monitor radionuclide precipitation and sorption onto the container walls during the experiments. The difference between the radionuclide concentration in the initial solution

Table 3. Solubility and Oxidation State of Plutonium in J-13 Groundwater*

pH	Solubility (M)	Pu(III) + Polymer	Pu(IV)	Pu(V)	Pu(VI)
7	2.3×10^{-7}	5% ± 1	6% ± 1	73% ± 7	18% ± 2
8.5	2.9×10^{-7}	3% ± 1	6% ± 1	63% ± 6	27% ± 3

*From Nitsche et al. 1993.

and in the control tubes varied for neptunium by $\pm 3\%$ and for uranium by $\pm 6\%$. Results for the plutonium solution, however, did show a small amount of sorption onto the container walls. Even here, the difference in concentration between the initial plutonium solution concentration and the plutonium solution in the control tube never exceeded 7% for any of the experiments reported in Appendix A. Nevertheless, in the case of plutonium, we calculated the amount of radionuclide sorbed in the solid phase by taking the difference of the final plutonium solution concentration both with the initial solution concentration and with the solution concentration in the control tube (see Appendix A). The latter approach is conservative because plutonium may sorb to container walls only in the absence of the geologic material.

The batch sorption distribution coefficient, K_d, for each experiment was calculated using:

$$K_d = \frac{F}{C} \quad , \qquad [1]$$

where F is the moles of radionuclide per gram of solid phase and C is the moles of radionuclide per milliliter of solution.

We performed the batch sorption experiments both under atmospheric conditions and inside glove boxes with a CO_2 overpressure. Under atmospheric conditions, the pH of J-13 water is approximately 8.5. The CO_2 overpressure in the glove-box experiments was applied to bring the pH of J-13 water down to 7. Table 4 lists references that provide details of the experimental setup and analytical techniques used in the sorption experiments.

Table 4. Procedures for Sorption Experiments

Procedure	Reference*
Batch sorption (under atmospheric conditions)	LANL-CST-DP-86
Batch sorption (within the controlled atmosphere of a glove box)	LANL-CST-DP-100
pH measurement	LANL-CST-DP-35
Liquid scintillation counting	LANL-CST-DP-79

**Yucca Mountain Project Detailed Procedures*, Los Alamos National Laboratory

Determination of very small or very large batch sorption distribution coefficients results in large uncertainties in the K_d values calculated. For the cases in which very little sorption occurs, the error is a result of subtracting two large numbers (the initial radionuclide concentration in solution and the radionuclide concentration after sorption) to obtain a small number (the amount of radionuclide in the solid phase). For the cases in which a great deal of sorption takes place, the error is a result of the uncertainty associated with measuring the small amount of radioactivity left in solution after sorption.

RESULTS AND DISCUSSION

Neptunium and Uranium Sorption

We studied the sorption of Np(V) and U(VI) onto samples of the three types of tuff in J-13 water (under oxidizing conditions) at the two pH values (7 and 8.5). However, to identify the sorbing minerals in the tuffs, we also studied sorption onto the pure minerals hematite, clinoptilolite, albite, and quartz. The results of all neptunium and uranium batch sorption experiments performed are reported in Appendix A. We found that neptunium and uranium in J-13 water do not sorb onto devitrified and vitric tuffs, albite, and quartz (see Table 5).

The initial neptunium concentrations for the data reported in Table 5 ranged from 1×10^{-7} to 3×10^{-5} M. We used wet-sieved tuffs, albite, and quartz samples with particle sizes in the range from 75 to 500 μm. The pretreatment period lasted 2 to 3 days, and the sorption period, 2 to 4 days. Initial uranium concentrations ranged from 8×10^{-8} to

Table 5. Neptunium and Uranium Sorption in J-13 Water (under oxidizing conditions)

Solid Phase	pH	K_d (mL/g)	
		Neptunium*	Uranium*
G4-268 Devitrified tuff	7	7×10^{-3}	2×10^{-1}
	8.5	-4×10^{-2}	7×10^{-1}
GU3-1405 Vitric tuff	7	2×10^{-1}	-5×10^{-1}
	8.5	3×10^{-1}	6×10^{-1}
Quartz	7	-1×10^{-1}	1×10^{-1}
	8.5	-2×10^{-1}	7×10^{-2}
Albite	7	-8×10^{-2}	-5×10^{-2}
	8.5	-1×10^{-1}	-1×10^{-1}

*The uncertainties in the data are ±0.5 for Np, ±3 for U.

1×10^{-4} M, and again, we used wet-sieved tuffs, albite, and quartz samples with particle sizes in the range from 75 to 500 μm. The pretreatment period for uranium was 2 to 4 days, and the sorption period, 3 to 4 days. The negative values reported in Table 5 are a result of the analytical error discussed earlier for the case of very little sorption (that is, a small number is obtained as the difference of two large numbers).

For the experimental conditions cited earlier, the sorption of neptunium onto zeolitic tuffs and clinoptilolite appears to be linear in the concentration range from 1×10^{-7} to 3×10^{-5} M and can be fitted using a K_d (see Figs. 4 and 5). The sorption of neptunium onto zeolites is higher at pH 7 than at pH 8.5, which might be explained by the

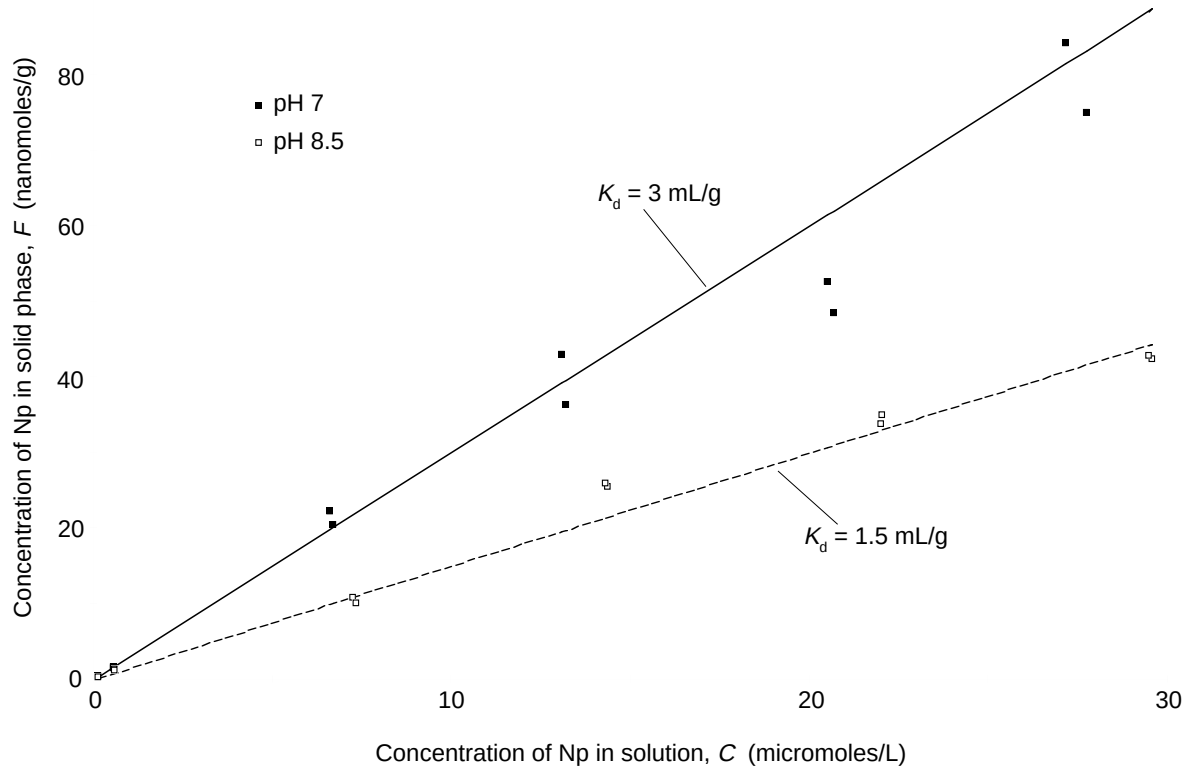

Figure 4. Neptunium Sorption onto Clinoptilolite-Rich Tuff. A plot of the concentration, *F*, of neptunium in the solid phase of the clinoptilolite-rich tuff G4-1510 versus the concentration, *C*, of neptunium in the solution phase of J-13 well water. The tuff was wet sieved to give particle sizes ranging from 75 to 500 micrometers. The period of pretreatment was 2 to 3 days; the period of sorption was 2 to 4 days.

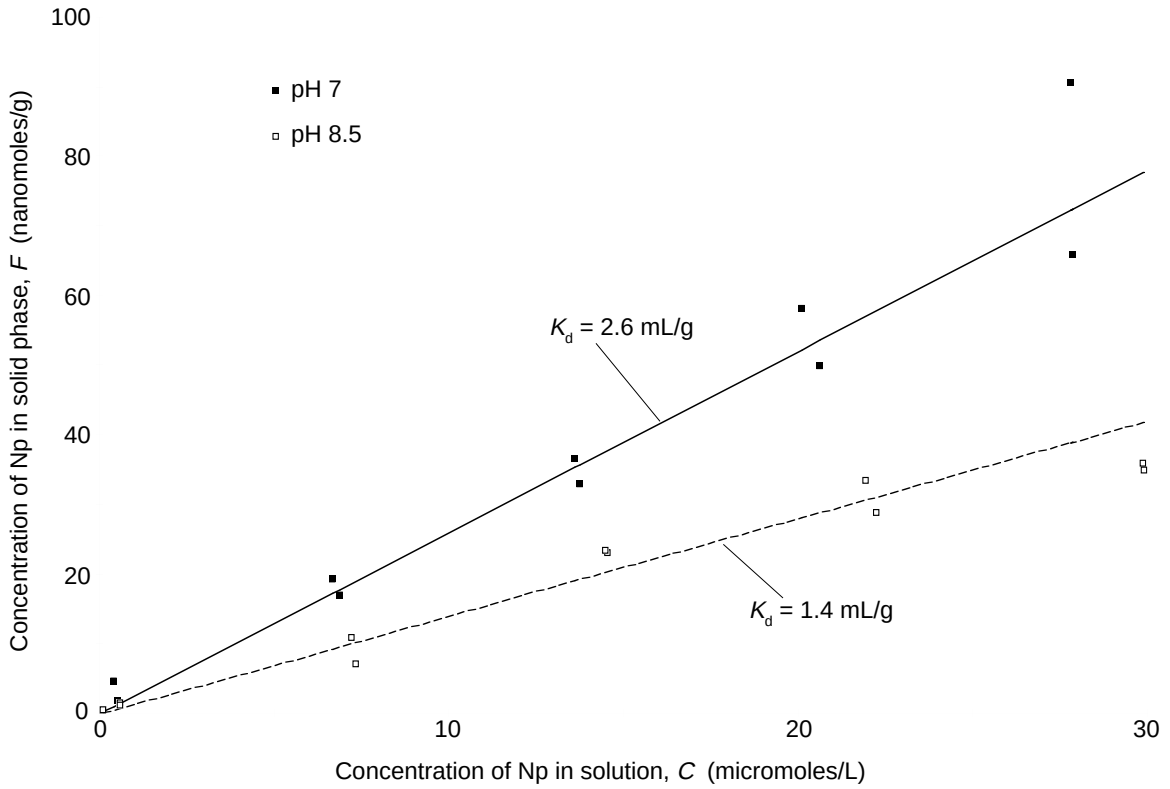

Figure 5. Neptunium Sorption onto Clinoptilolite. A plot of the concentration, *F*, of neptunium in the solid phase of clinoptilolite versus the concentration, *C*, of neptunium in the solution phase of J-13 well water. The mineral was unsieved. The period of pretreatment was 2 to 3 days; the period of sorption was 2 to 4 days.

larger amount of NpO_2^+ relative to $NpO_2CO_3^-$ in J-13 water at pH 7 than at pH 8.5 (Table 2).

One surprise for neptunium is the relatively small amount of sorption (values of K_d ranging from 1.5 to 3 mL/g) compared to the large amount expected for a cation-exchange sorption mechanism in a zeolite with a large cation-exchange capacity (such as clinoptilolite). This result indicates that the sorption mechanism for neptunium onto clinoptilolite is a surface reaction rather than a cation exchange within the cages of the zeolite. One possible explanation is steric: the shape and large size of the neptunyl cation prevents cation exchange. This ion likely has a trans-dioxol configuration normal to a puckered equatorial ring that contains six bound water molecules.

For uranium, under the same conditions, sorption onto zeolitic tuffs and clinoptilolite is nonlinear in the concentration range from 8×10^{-8} M to 1×10^{-4} M and can be fitted with Freundlich and Langmuir isotherms (Figs. 6 and 7). Nonlinear adsorption isotherms have been reviewed by De Marsily (1986). The functional forms of the Freundlich and the Langmuir isotherms that relate radionuclide concentration in the solid phase, *F*, to the radionuclide concentration in the solution phase, *C*, are given by the following equations:

$$\text{Freundlich's Isotherm:} \quad F = KC^{1/n} \quad , \qquad [2]$$

where K is a constant > 0 and n is a constant ≥ 1.

$$\text{Langmuir's Isotherm:} \quad F = \frac{K_1 C}{1 + K_2 C} \quad , \qquad [3]$$

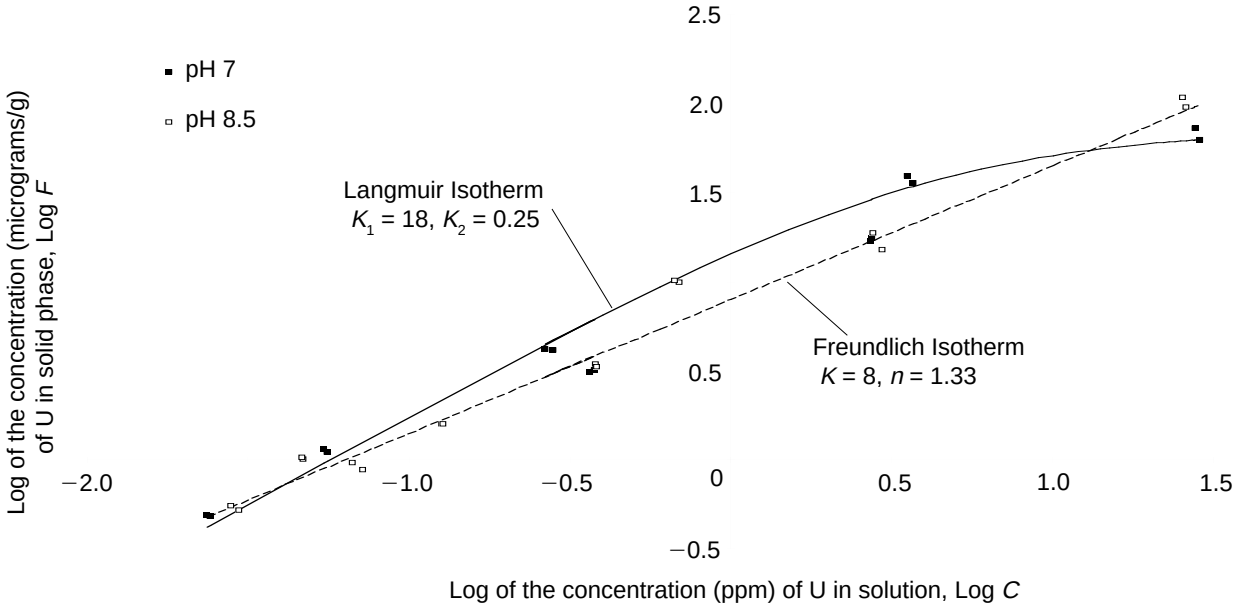

Figure 6. Uranium Sorption onto Clinoptilolite-Rich Tuff. A log-log plot of the concentration of uranium in the solid phase, *F*, of the clinoptilolite-rich tuff G4-1510 versus the concentration of uranium in the solution phase, *C*, of J-13 well water. The tuff was wet sieved to give particle that ranged in size from 75 to 500 micrometers. The period of pretreatment was 2 to 4 days; the period of sorption was 3 to 4 days. The data for a pH of 7 have been fitted with a Langmuir isotherm; the data for a pH of 8.5 have been fitted with a Freundlich isotherm.

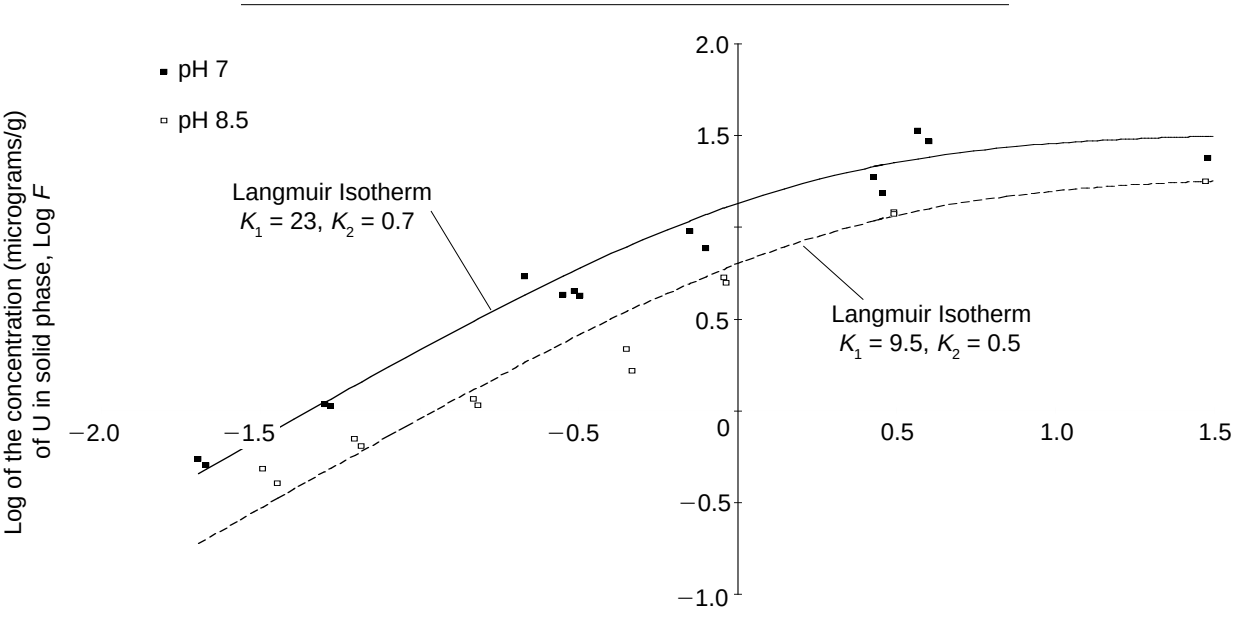

Figure 7. Uranium Sorption onto Clinoptilolite. A log-log plot of the concentration of uranium in the solid phase, *F*, of clinoptilolite versus the concentration of uranium in the solution phase, *C*, of J-13 water. The mineral was unsieved. The period of pretreatment was 2 to 4 days; the period of sorption was 3 to 4 days. The data for each pH (7 and 8.5) have been fitted with a Langmuir isotherm.

where K_1 and K_2 are constants > 0.

For the clinoptilolite-rich zeolitic tuff G4-1510, the scatter in the data makes it impossible to conclude whether there is a significant difference between the experiments performed under a CO_2 overpressure and a pH of 7 or at atmospheric conditions and a pH of 8.5 (Fig. 6). However, the experiments with pure clinoptilolite indicate that sorption increases with decreasing pH for U(VI) (Fig. 7), as is the case for Np(V). Because the major constituent of tuff G4-1510 is clinoptilolite, predictions of the K_a (K_d divided by solid-phase surface area) were made for neptunium and uranium sorption onto this tuff by assuming that clinoptilolite is the only sorbing phase. Inspection of Table 6 indicates that reasonable predictions are obtained with this assumption except in the case of uranium at pH 8.5. In all cases, predictions based on clinoptilolite sorption are conservative.

The sorption of neptunium and uranium onto pure iron oxides (such as hematite) is very large (as shown in Appendix A). For the cases in which a great deal of sorption takes place, the large uncertainties associated with a K_d measurement are the result of the uncertainty associated with measuring the small amount of radionuclide concentration left in solution after sorption. Although the measured sorption of neptunium and uranium onto pure hematite is very large, their sorption onto devitrified tuffs, which appear to have traces of hematite (1% $\pm$1), is essentially zero. This result could be due to differences in the surface of pure hematite compared to hematite in tuff. It could also be due to passivation of the hematite surfaces in the tuff by elements (such as the rare earths) that have a higher affinity for hematite than neptunium or uranium and, thus, occupy the sorption sites.

Plutonium Sorption

We studied the sorption of plutonium onto the three types of tuff in J-13 water (under oxidizing conditions) using a CO_2 overpressure (to obtain a pH of 7). To identify the sorbing minerals in the tuffs, we also studied sorption onto the pure minerals hematite, clinoptilolite, albite, and quartz. The results of the batch sorption experiments for plutonium are presented in Appendix A and summarized in Fig. 8. Because plutonium sorbs onto nongeologic media (see Experimental Procedures section), the batch sorption distribution coefficients reported in Fig. 8 are based on the concentration of plutonium in the control solutions. The affinity of tuffs for plutonium at pH 7 in decreasing order is zeolitic > vitric > devitrified. The affinity of minerals for plutonium in decreasing order is hematite > clinoptilolite > albite > quartz. Inspection of Fig. 8 indicates that plutonium sorption is nonlinear in the concentration range from 6×10^{-9} M to 2×10^{-7} M.

Table 6. Prediction of Sorption on G4-1510 Tuff in J-13 Well Water (assuming clinoptilolite is the only sorbing mineral in the tuff)

Radionuclide	Initial Concentration Range (M)	pH	$K_a = K_d$ /surface area of solid (m)	
			Measured	Predicted
^{237}Np	1×10^{-7} to 3×10^{-5}	7	1×10^{-7}	9×10^{-8}
		8.5	6×10^{-8}	5×10^{-8}
U	2×10^{-7} to 4×10^{-7}	7	8×10^{-7}	8×10^{-7}
		8.5	8×10^{-7}	4×10^{-7}

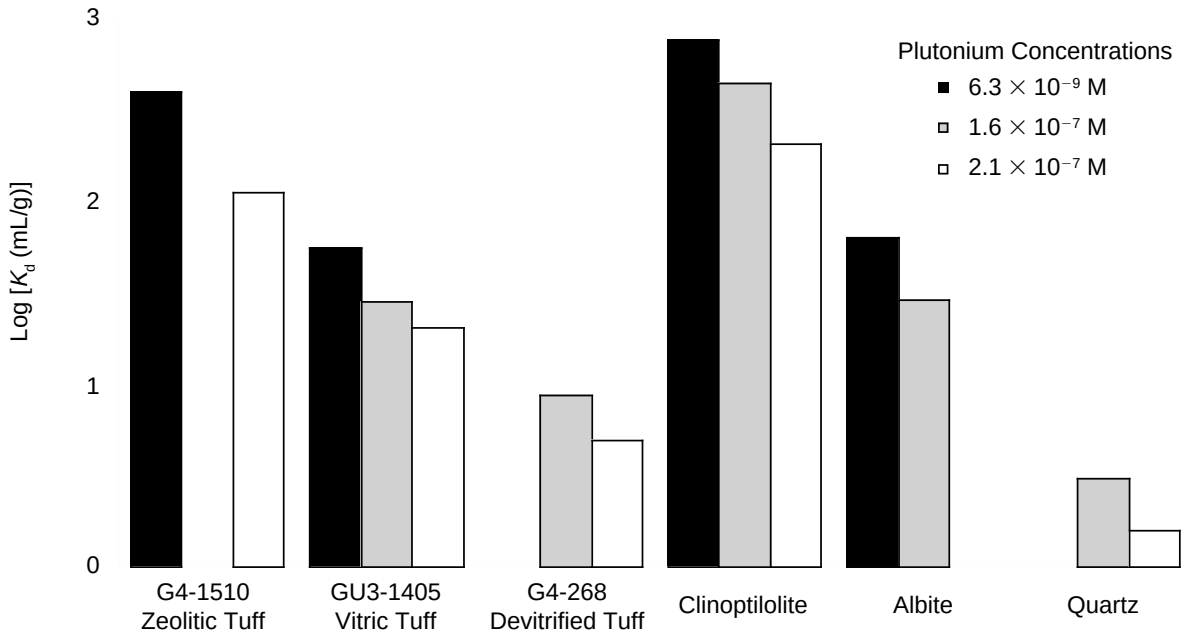

Figure 8. Plutonium Sorption. The log of the batch sorption distribution coefficient, Log K_d, is shown for the sorption of plutonium in J-13 well water at pH 7 and the specified initial plutonium concentrations. All solids, except clinoptilolite, were wet sieved to particle sizes ranging from 75 to 500 micrometers. The periods of pretreatment and sorption were each 3 days.

Nitsche et al. (1993) report that even when a plutonium solution in J-13 water is prepared starting in the Pu(IV) oxidation state, the predominant final oxidation state is Pu(V) (see Table 3). The solution used for the plutonium-sorption experiments was prepared from a well-characterized Pu(V) acidic stock. Consequently, it would be reasonable to assume that the plutonium would have remained predominantly in the Pu(V) oxidation state in the J-13 solution used for the sorption studies. Inspection of Fig. 8 indicates that significant plutonium sorption occurred in tuffs and minerals that, on the other hand, exhibit very small sorption of Np(V) and U(VI). This result is very puzzling; if plutonium is predominantly Pu(V) and Pu(VI) in J-13 well water, it is expected that its sorption behavior would have been similar to that observed for Np(V) and U(VI). Several possible explanations (Nitsche et al. 1993) of the plutonium sorption results are 1) the data for the plutonium oxidation states are incorrect, and the predominant oxidation state of plutonium in J-13 well water at pH 7 is Pu(IV), not Pu(V) and Pu(VI); 2) plutonium sorbs as Pu(IV) from J-13 water but a re-equilibration in the solution produces more Pu(IV) (which implies that the kinetics of plutonium speciation in solution are fast); and 3) Pu(V) and Pu(VI) reduce to Pu(IV) as a result of changes in the solution redox potential in the presence of the solid phases. Future experiments will address these issues.

SUMMARY

- The sorption of neptunium, uranium, and plutonium onto tuffs and pure minerals in J-13 well water has been studied under oxidizing conditions in the pH range from 7 to 8.5. The tuff types studied were vitric, devitrified, and zeolitic. The minerals studied were clinoptilolite, albite, quartz, and synthetic hematite.

- The sorption of Np(V) and U(VI) in J-13 water onto devitrified and vitric tuffs, albite, and

quartz is essentially zero.

- The sorption of Np(V) onto clinoptilolite-rich zeolitic tuffs and pure clinoptilolite is linear in the concentration range from 1×10^{-7} to 3×10^{-5} M; it approximately doubles as the pH decreases from 8.5 to 7.

- The sorption of U(VI) onto clinoptilolite-rich zeolitic tuffs and pure clinoptilolite is nonlinear in the concentration range from 8×10^{-8} to 1×10^{-4} M, and it can be fitted with Freundlich and Langmuir isotherm models. The sorption of U(VI) onto clinoptilolite increases as the pH decreases from 8.5 to 7.

- The minimum sorption of Np(V) and U(VI) onto clinoptilolite-rich zeolitic tuffs can be predicted based on sorption data obtained with pure clinoptilolite.

- Synthetic hematite sorbs neptunium and uranium strongly; however, their sorption onto tuffs with apparent traces of hematite is zero. The iron oxides in the tuffs appear to be inactive. Future experiments will address the reason for this observation.

- Plutonium (initially, in the Pu(V) oxidation state) in J-13 well water sorbs significantly onto tuffs and minerals that exhibit minimal sorption for Np(V) and U(VI). The affinity of tuffs for plutonium in decreasing order is zeolitic > vitric > devitrified; the affinity of minerals for plutonium in decreasing order is hematite > clinoptilolite > albite > quartz.

- Future experiments will address plutonium sorption as a function of oxidation state and bicarbonate-carbonate concentrations in solution.

ACKNOWLEDGMENTS

The authors wish to thank Dr. John Husler (from the University of New Mexico) for the chemical analysis of the groundwater used in the sorption experiments, Mr. Phil Palmer (from Los Alamos National Laboratory) for providing the Np(V) and Pu(V) acidic stock used to prepare the sorption solutions, and Quantachrome Corporation for the BET analyses of the tuffs and minerals.

The work was supported by the Yucca Mountain Site Characterization Project Office of Los Alamos National Laboratory as part of the Civilian Radioactive Waste Management Program of the U.S. Department of Energy. The Los Alamos data tracking number for this record package is LA000000000104.001.

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Note on Sample IDs in Appendix A

For readability and ease of presentation, the data presented in tables in Appendix A have been consolidated from quality-program-approved and audited electronic notebooks by having repetitive information, including sample IDs, summarized in the subheadings for each series of samples. As a result, there are several minor differences in the sample IDs listed in the tables and those present in the electronic notebooks. These differences are noted below and are recorded in the appropriate binder containing the original data.

Appendix A Sample ID	Electronic notebook Sample ID	Data Binder
J-13 G4-1510-C.3243-20	J-13 G4-1510-C.324320	LA-CST-03-94-09
J-13 M-C.3264-20	J-13 M-C.326420	LA-CST-03-94-09
J-13 Gu3-1405-C.3537-20	J-13 Gu3-155-C.3537-20	LA-CST10-NBK-94-004
J-13 Gu3-1405-C.3523-20	J-13 Gu3-155-C.3523-20	LA-CST10-NBK-94-004
J-13 C-C.3740-20	J-13 C-C.374020	LA-CST10-NBK-94-004
J-13 M-C.3784-20	J-13 M-C.378420	LA-CST10-NBK-94-004

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #61

Atmosphere: CO_2 J-13 groundwater Temperature range: 22–25 ˚C Binder where data are located: LA-CST-03-94-09

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Devitrified tuff (USW G4-268), wet sieved (75-500 micrometers)													
Sample IDs: J-13 G4-268-C.XXXX-20													
3256	1.00	18.19	3	7.4	19.49	3	20.35	20.34	7.1	49.8	47.2	51.3	2×10^{-1}
3257	1.03	19.00	3	N/A	19.30	3	19.51	19.50	N/A	49.8	48.8	51.3	-4×10^{-1}
3240	1.02	18.81	3	7.3	19.43	4	19.90	19.90	7.0	2986.1	2828.9	2999.3	2×10^{-1}
3241	0.99	17.04	3	N/A	19.70	4	20.74	20.74	N/A	2986.1	2874.2	2999.3	-6×10^{-2}
3224	1.01	18.03	3	7.3	19.55	3	20.29	20.24	7.0	5989.4	5706.9	6025.8	8×10^{-2}
3225	0.97	17.92	3	N/A	19.64	3	21.25	21.21	N/A	5989.4	5772.4	6025.8	-2×10^{-1}
3192	0.97	18.17	3	7.1	19.40	3	21.09	21.08	7.1	9112.2	8603.1	9317.3	1×10^{-1}
3193	0.99	19.26	3	N/A	19.41	3	20.54	20.53	N/A	9112.2	8787.1	9317.3	-2×10^{-1}
3208	0.99	18.99	3	7.2	19.14	3	20.38	20.37	7.0	9112.2	8609.2	9317.3	9×10^{-2}
3176	0.97	19.70	3	7.0	19.54	3	21.01	21.04	7.0	12259.6	11577.5	12234.7	3×10^{-1}
3177	0.99	19.08	3	N/A	19.45	3	20.37	20.40	N/A	12259.6	11876.1	12234.7	-1×10^{-1}
Zeolitic tuff (USW G4-1510), wet sieved (75-500 micrometers)													
Sample IDs: J-13 G4-1510-C.XXXX-20													
3258	1.01	18.48	3	7.3	19.38	3	20.15	20.15	7.0	49.8	40.4	51.3	4
3259	0.98	19.49	3	N/A	19.45	3	20.77	20.77	N/A	49.8	40.7	51.3	3
3210	0.96	19.53	3	7.0	19.86	3	22.28	22.24	7.1	243.9	201.5	248.6	3
3211	0.98	19.83	3	N/A	19.83	3	21.70	21.68	N/A	243.9	199.5	248.6	3
3242	0.99	18.60	3	7.2	19.90	4	21.10	21.07	7.0	2986.1	2456.7	2999.3	3
3243	0.98	19.05	3	N/A	19.90	4	21.32	21.28	N/A	2986.1	2492.5	2999.3	3
3226	1.01	16.74	3	7.2	19.49	3	20.50	20.46	7.1	5989.4	4868.3	6025.8	3
3227	1.01	16.89	3	N/A	19.28	3	20.60	20.55	N/A	5989.4	4906.1	6025.8	3
3194	0.99	16.56	3	7.1	19.41	3	20.87	20.86	6.9	9112.2	7627.6	9317.3	3
3195	0.98	19.26	3	N/A	19.41	3	21.13	21.12	N/A	9112.2	7690.8	9317.3	2
3178	0.98	19.26	3	7.0	19.50	3	21.01	21.04	7.0	12259.6	10106.0	12234.7	3
3179	0.98	19.01	3	N/A	19.38	3	20.76	20.79	N/A	12259.6	10321.9	12234.7	3

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #61 continued

Atmosphere: CO_2 J-13 groundwater Temperature range: 22–25 ˚C Binder where data are located: LA-CST-03-94-09

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Vitric tuff (USW GU3-1405), wet sieved (75-500 micrometers)													
Sample IDs: J-13 Gu3-1405-C.XXXX-20													
3260	1.00	18.97	3	7.3	19.51	3	20.33	20.33	6.9	49.8	47.3	51.3	2×10^{-1}
3261	1.03	19.42	3	N/A	19.68	3	19.87	19.87	N/A	49.8	47.0	51.3	4×10^{-1}
3212	1.03	19.15	3	7.0	19.74	3	20.16	20.13	7.1	243.9	227.6	248.6	4×10^{-1}
3213	1.01	17.44	3	N/A	19.80	3	20.74	20.68	N/A	243.9	231.8	248.6	-6×10^{-2}
3244	1.02	18.85	3	7.1	19.66	4	20.12	20.09	6.9	2986.1	2825.2	2999.3	3×10^{-1}
3245	0.98	17.27	3	N/A	19.67	4	20.95	20.92	N/A	2986.1	2859.6	2999.3	4×10^{-2}
3228	1.02	16.12	3	7.2	19.92	4	20.48	20.43	7.1	5989.4	5686.3	6025.8	1×10^{-1}
3229	1.00	19.37	3	N/A	15.80	4	16.76	16.72	N/A	5989.4	5445.7	6025.8	7×10^{-1}
3180	0.97	18.80	3	7.0	19.49	3	21.02	21.02	7.0	12259.6	11522.1	12234.7	4×10^{-1}
3181	0.98	19.04	3	N/A	19.23	3	20.47	20.49	N/A	12259.6	11697.7	12234.7	8×10^{-2}
Synthetic Hematite, not sieved													
Sample IDs: J-13 C-C.XXXX-20													
3262	1.03	19.41	3	6.9	19.42	3	19.83	19.78	6.7	49.8	4.8	51.3	2×10^{2}
3263	0.97	19.15	3	N/A	19.54	3	20.89	20.84	N/A	49.8	1.1	51.3	9×10^{2}
3214	1.01	19.71	3	6.8	19.72	3	20.32	20.26	6.8	243.9	5.9	248.6	8×10^{2}
3215	1.03	19.59	3	N/A	19.88	3	20.24	20.17	N/A	243.9	5.6	248.6	8×10^{2}
3246	0.96	19.47	3	6.8	19.71	2	21.21	21.16	6.9	2986.1	125.1	2999.3	5×10^{2}
3247	1.00	19.27	3	N/A	19.86	2	20.48	20.43	N/A	2986.1	165.2	2999.3	3×10^{2}
3230	1.03	19.26	3	6.9	19.68	4	19.90	19.89	6.9	5989.4	273.3	6025.8	4×10^{2}
3231	0.97	18.93	3	N/A	19.87	4	21.16	21.20	N/A	5989.4	310.8	6025.8	4×10^{2}
3198	1.00	18.81	3	6.8	19.38	3	20.29	20.28	6.8	9112.2	501.4	9317.3	3×10^{2}
3199	1.00	17.86	3	N/A	19.45	3	20.35	20.33	N/A	9112.2	501.6	9317.3	3×10^{2}
3182	1.00	18.24	3	6.7	19.32	3	20.14	20.16	6.9	12259.6	931.5	12234.7	2×10^{2}
3183	1.02	15.47	3	N/A	19.17	3	19.77	19.80	N/A	12259.6	699.2	12234.7	3×10^{2}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #61 continued

Atmosphere: CO_2 J-13 groundwater Temperature range: 22–25 ˚C Binder where data are located: LA-CST-03-94-09

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Clinoptilolite, not sieved													
Sample IDs: J-13 G-C.XXXX-20													
3266	1.01	19.63	3	7.2	19.34	3	20.32	20.28	7.1	49.8	37.4	51.3	5
3267	1.01	19.70	3	N/A	19.45	3	20.64	20.59	N/A	49.8	37.9	51.3	5
3218	1.01	18.86	3	7.1	19.68	3	20.94	20.88	7.1	243.9	146.5	248.6	10
3219	1.00	18.21	3	N/A	19.65	3	21.48	21.41	N/A	243.9	192.4	248.6	3
3250	1.04	19.12	3	7.4	19.56	2	19.83	19.80	7.1	2986.1	2473.0	2999.3	3
3251	0.99	18.93	3	N/A	19.79	2	20.96	20.91	N/A	2986.1	2553.9	2999.3	2
3234	1.01	19.06	3	7.3	19.83	4	20.59	20.55	7.1	5989.4	5059.5	6025.8	3
3235	1.01	18.72	3	N/A	19.91	4	20.74	20.69	N/A	5989.4	5113.7	6025.8	2
3202	1.00	18.46	3	7.2	18.07	3	19.15	19.14	7.0	9112.2	7473.6	9317.3	3
3203	1.00	16.56	3	N/A	19.27	3	20.46	20.46	N/A	9112.2	7675.5	9317.3	2
3186	0.99	19.39	3	7.1	19.31	3	20.68	20.71	6.6	12259.6	10363.3	12234.7	2
3187	1.00	15.64	3	N/A	20.06	3	20.48	20.51	N/A	12259.6	10347.1	12234.7	3
Quartz, wet sieved (75-500 micrometers)													
Sample IDs: J-13 M-C.XXXX-20													
3264	1.03	19.63	3	7.3	19.52	3	19.58	19.53	6.9	49.8	49.4	51.3	-4×10^{-1}
3265	0.98	19.62	3	N/A	19.65	3	20.84	20.77	N/A	49.8	49.7	51.3	-7×10^{-1}
3216	1.00	19.50	3	7.0	19.69	3	20.59	20.56	7.0	243.9	232.6	248.6	8×10^{-2}
3217	1.03	19.66	3	N/A	19.74	3	20.10	20.06	N/A	243.9	235.8	248.6	-2×10^{-1}
3248	1.01	19.19	3	7.2	19.67	2	20.13	20.09	7.0	2986.1	2897.2	2999.3	-2×10^{-2}
3249	1.01	19.19	3	N/A	19.60	2	20.24	20.17	N/A	2986.1	2940.7	2999.3	-5×10^{-1}
3232	1.00	19.21	3	7.1	19.90	4	20.59	20.34	7.0	5989.4	5774.4	6025.8	3×10^{-1}
3233	1.00	19.35	3	N/A	19.90	4	20.59	20.57	N/A	5989.4	5874.3	6025.8	-3×10^{-1}
3200	0.99	17.47	3	7.0	19.33	3	20.31	20.30	7.0	9112.2	8755.4	9317.3	2×10^{-2}
3201	1.03	18.29	3	N/A	19.59	3	19.66	19.65	N/A	9112.2	8878.5	9317.3	-1×10^{-1}
3184	0.98	18.48	3	6.9	19.46	3	20.57	20.60	6.9	12259.6	11752.5	12234.7	1×10^{-1}
3185	1.02	19.14	3	N/A	19.31	3	19.56	19.59	N/A	12259.6	11837.9	12234.7	2×10^{-2}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #61 continued

Atmosphere: CO_2 J-13 groundwater Temperature range: 22–25 ˚C Binder where data are located: LA-CST-03-94-09

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Albite, wet sieved (75-500 micrometers)													
Sample IDs: J-13 W-C.XXXX-20													
3268	0.97	19.58	3	7.1	19.48	3	20.86	20.81	7.1	49.8	46.4	51.3	7×10^{-1}
3269	0.98	20.60	3	N/A	19.62	3	20.78	20.72	N/A	49.8	48.9	51.3	-4×10^{-1}
3220	1.02	18.17	3	7.5	19.00	3	19.37	19.32	7.1	243.9	236.8	248.6	-1×10^{-1}
3221	1.03	19.42	3	N/A	19.70	3	19.80	19.76	N/A	243.9	239.2	248.6	-3×10^{-1}
3252	0.99	18.38	3	7.4	19.62	2	20.62	20.58	7.0	2986.1	2890.1	2999.3	-1×10^{-1}
3253	0.99	17.85	3	N/A	19.60	2	20.64	20.60	N/A	2986.1	2925.3	2999.3	-4×10^{-1}
3236	1.00	18.24	3	7.2	19.43	4	20.21	20.22	7.0	5989.4	5726.8	6025.8	1×10^{-1}
3237	1.00	19.37	3	N/A	19.82	4	20.74	20.71	N/A	5989.4	5799.7	6025.8	-2×10^{-1}
3204	1.02	19.65	3	7.2	18.87	3	19.28	19.27	7.0	9112.2	8709.7	9317.3	8×10^{-2}
3205	0.99	19.44	3	N/A	19.35	3	20.56	20.55	N/A	9112.2	8725.8	9317.3	-1×10^{-1}
3188	1.02	19.31	3	7.1	19.27	3	19.86	19.90	6.9	12259.6	11639.7	12234.7	-4×10^{-3}
3189	1.01	18.97	3	N/A	19.35	3	20.02	20.05	N/A	12259.6	11870.8	12234.7	-3×10^{-1}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #64

Atmosphere: air | J-13 groundwater | Temperature range: 22–25 ˚C | Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Devitrified tuff (USW G4-268), wet sieved (75-500 micrometers)													
Sample IDs: J-13 G4-268-C.XXXX-20													
3533	1.00	19.90	3	N/A	20.08	3	20.88	20.72	N/A	49.6	48.6	50.5	-2×10^{-1}
3534	0.98	19.85	3	8.3	19.98	3	21.17	21.15	8.4	49.6	48.7	50.5	-4×10^{-1}
3519	1.02	19.67	2	N/A	20.12	4	20.43	20.33	N/A	248.0	239.4	250.8	1×10^{-1}
3520	1.00	19.70	2	8.4	19.95	4	20.79	20.77	8.4	248.0	241.5	250.8	-3×10^{-1}
3505	1.00	19.89	2	N/A	20.25	4	20.91	20.73	N/A	2997.6	2919.8	3041.4	6×10^{-2}
3506	1.00	19.79	2	8.3	20.08	4	20.73	20.64	8.4	2997.6	2914.8	3041.4	8×10^{-3}
3491	0.99	19.81	2	N/A	19.99	3	20.82	20.80	N/A	6042.7	5836.1	6168.7	1×10^{-1}
3492	1.00	19.81	2	8.5	19.93	3	20.60	20.63	8.3	6042.7	5814.1	6168.7	8×10^{-2}
3477	0.97	19.72	2	N/A	20.14	3	21.45	21.57	N/A	9143.0	8772.4	9246.2	7×10^{-2}
3478	1.00	19.77	2	8.4	20.07	3	20.78	20.87	8.4	9143.0	8830.2	9246.2	-9×10^{-2}
3461	1.01	20.03	2	N/A	20.07	3	20.53	20.52	N/A	12220.2	11825.3	12296.6	1×10^{-2}
3462	1.01	20.01	2	8.5	20.05	3	20.49	20.52	8.7	12220.2	11794.3	12296.6	5×10^{-2}
Zeolitic tuff (USW G4-1510), wet sieved (75-500 micrometers)													
Sample IDs: J-13 G4-1510-C.XXXX-20													
3535	0.99	19.85	3	N/A	20.02	3	21.11	21.03	N/A	49.6	41.6	50.5	3
3536	1.00	19.85	3	8.2	20.04	3	20.83	20.79	8.4	49.6	42.4	50.5	3
3521	0.99	19.62	2	N/A	20.33	4	21.50	21.26	N/A	248.0	217.1	250.8	2
3522	1.00	19.63	2	8.1	20.24	4	21.23	21.07	8.4	248.0	217.9	250.8	2
3507	1.00	19.81	2	N/A	20.05	4	20.84	20.81	N/A	2997.6	2694.8	3041.4	1
3508	1.00	19.76	2	8.1	20.21	4	20.96	20.86	8.4	2997.6	2724.2	3041.4	1
3493	1.00	19.80	2	N/A	20.05	3	20.83	20.91	N/A	6042.7	5340.2	6168.7	2
3494	1.01	19.84	2	8.3	20.01	3	20.59	20.67	8.3	6042.7	5322.5	6168.7	2
3479	1.02	19.69	2	N/A	20.10	3	20.46	20.47	N/A	9143.0	8185.1	9246.2	2
3480	1.00	19.50	2	8.4	20.08	3	20.79	20.83	8.3	9143.0	8189.7	9246.2	2
3463	1.03	19.98	2	N/A	20.04	3	20.15	20.17	N/A	12220.2	11004.0	12296.6	1
3464	1.01	19.97	2	8.4	20.07	3	20.63	20.70	8.3	12220.2	10962.0	12296.6	1

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #64 continued

Atmosphere: air | J-13 groundwater | Temperature range: 22–25 ˚C | Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Vitric tuff (USW GU3-1405), wet sieved (75-500 micrometers)													
Sample IDs: J-13 Gu3-1405-C.XXXX-20													
3537	1.00	19.81	3	N/A	19.91	3	20.82	20.73	N/A	49.6	46.6	50.5	4×10^{-1}
3538	1.01	19.75	3	8.4	19.98	3	20.69	20.62	8.5	49.6	46.3	50.5	6×10^{-1}
3523	1.01	19.60	2	N/A	20.28	4	21.01	20.89	N/A	248.0	230.0	250.8	8×10^{-1}
3524	0.99	19.61	2	8.4	20.24	4	21.30	21.13	8.4	248.0	232.2	250.8	7×10^{-1}
3509	1.00	19.81	2	N/A	20.00	4	20.67	20.58	N/A	2997.6	2887.0	3041.4	2×10^{-1}
3510	1.01	19.73	2	8.4	20.29	4	20.80	20.61	8.4	2997.6	2909.0	3041.4	9×10^{-2}
3495	1.00	19.79	2	N/A	19.99	3	20.69	20.76	N/A	6042.7	5752.1	6168.7	2×10^{-1}
3496	1.00	19.80	2	8.5	20.05	3	20.77	20.86	8.4	6042.7	5725.6	6168.7	3×10^{-1}
3481	1.01	19.70	2	N/A	20.06	3	20.53	20.61	N/A	9143.0	8788.3	9246.2	5×10^{-2}
3482	1.02	19.66	2	8.5	20.08	3	20.34	20.44	8.4	9143.0	8746.8	9246.2	1×10^{-1}
3465	1.00	19.95	2	N/A	20.10	3	20.82	20.91	N/A	12220.2	11624.6	12296.6	2×10^{-1}
3466	1.00	19.92	2	8.5	19.13	3	19.82	19.92	8.4	12220.2	11606.3	12296.6	2×10^{-1}
Synthetic Hematite, not sieved													
Sample IDs: J-13 C-C.XXXX-20													
3539	0.99	19.77	3	N/A	20.00	3	20.85	20.84	N/A	49.6	0.7	50.5	1×10^{3}
3540	1.00	19.72	3	7.6	19.93	3	20.56	20.49	8.3	49.6	0.4	50.5	2×10^{3}
3525	1.00	19.59	2	N/A	20.39	4	20.83	20.69	N/A	248.0	3.8	250.8	1×10^{3}
3526	0.99	19.61	2	7.5	20.32	4	21.06	20.98	8.3	248.0	3.6	250.8	1×10^{3}
3511	0.99	19.79	2	N/A	20.36	4	21.14	21.01	N/A	2997.6	53.1	3041.4	1×10^{3}
3512	1.02	19.74	2	7.5	20.50	4	20.65	20.30	8.2	2997.6	52.8	3041.4	1×10^{3}
3497	0.99	19.79	2	N/A	20.00	3	20.68	20.73	N/A	6042.7	290.9	6168.7	4×10^{2}
3498	1.01	19.72	2	7.5	20.06	3	20.35	20.42	8.2	6042.7	274.5	6168.7	4×10^{2}
3483	1.00	19.82	2	N/A	20.07	3	20.55	20.63	N/A	9143.0	1093.3	9246.2	1×10^{2}
3484	1.00	19.87	2	7.4	20.04	3	20.59	20.66	8.2	9143.0	749.7	9246.2	2×10^{2}
3467	1.00	19.93	2	N/A	20.13	3	20.69	20.83	N/A	12220.2	1029.9	12296.6	2×10^{2}
3468	1.01	19.83	2	7.4	20.10	3	20.38	20.49	8.2	12220.2	853.1	12296.6	3×10^{2}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #64 continued

Atmosphere: air | J-13 groundwater | Temperature range: 22–25 ˚C | Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Clinoptilolite, not sieved													
Sample IDs: J-13 G-C.XXXX-20													
3543	1.01	19.89	3	N/A	19.85	3	20.67	20.68	N/A	49.6	39.2	50.5	4
3544	0.99	19.92	3	8.4	19.81	3	20.95	20.91	8.5	49.6	39.7	50.5	4
3529	1.00	19.75	2	N/A	20.28	4	21.13	20.98	N/A	248.0	214.4	250.8	2
3530	0.99	19.71	2	8.5	20.08	4	21.23	21.19	8.4	248.0	216.0	250.8	2
3515	1.02	19.77	2	N/A	20.12	4	20.61	20.52	N/A	2997.6	2684.5	3041.4	2
3516	1.00	19.76	2	8.5	20.06	4	21.03	21.05	8.4	2997.6	2730.7	3041.4	1
3501	0.99	19.75	2	N/A	19.93	3	20.94	20.93	N/A	6042.7	5404.3	6168.7	2
3502	1.00	19.70	2	8.2	20.04	3	20.85	20.85	8.3	6042.7	5391.5	6168.7	2
3487	1.02	19.87	2	N/A	20.06	3	20.41	20.45	N/A	9143.0	8270.3	9246.2	1
3488	1.00	19.91	2	8.5	20.06	3	20.89	20.95	8.3	9143.0	8161.4	9246.2	2
3471	0.98	19.87	2	N/A	20.06	3	21.23	21.29	N/A	12220.2	11118.2	12296.6	1
3472	1.01	19.80	2	8.5	20.13	3	20.67	20.72	8.3	12220.2	11123.9	12296.6	1
Quartz, wet sieved (75-500 micrometers)													
Sample IDs: J-13 M-C.XXXX-20													
3541	0.99	19.71	3	N/A	19.91	3	20.73	20.67	N/A	49.6	49.4	50.5	-5×10^{-1}
3542	0.99	19.76	3	8.3	19.95	3	20.76	20.69	8.4	49.6	49.9	50.5	-7×10^{-1}
3527	0.50	20.13	2	N/A	20.67	4	43.47	42.59	N/A	248.0	246.4	250.8	-1
3528	0.99	19.82	2	8.4	20.30	4	21.08	20.94	8.4	248.0	244.6	250.8	-1×10^{-1}
3513	1.00	19.80	2	N/A	20.27	4	20.84	20.40	N/A	2997.6	2937.2	3041.4	3×10^{-1}
3514	1.01	19.73	2	8.4	20.18	4	20.63	20.58	8.4	2997.6	2930.9	3041.4	-1×10^{-1}
3499	1.01	19.78	2	N/A	20.04	3	20.32	20.36	N/A	6042.7	5906.3	6168.7	-6×10^{-2}
3500	0.99	19.75	2	8.5	20.08	3	20.84	20.88	8.3	6042.7	5839.1	6168.7	1×10^{-1}
3485	1.00	19.89	2	N/A	20.01	3	20.58	20.63	N/A	9143.0	8866.1	9246.2	9×10^{-3}
3486	1.00	19.94	2	8.5	20.10	3	20.55	20.60	8.3	9143.0	8887.9	9246.2	7×10^{-2}
3469	1.00	19.84	2	N/A	20.11	3	20.60	20.69	N/A	12220.2	11959.0	12296.6	-1×10^{-1}
3470	1.00	19.85	2	8.5	20.19	3	20.68	20.76	8.4	12220.2	11867.6	12296.6	3×10^{-2}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Neptunium-237 Sorption Experiment #64 continued

Atmosphere: air | J-13 groundwater | Temperature range: 22–25 ˚C | Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Neptunium-237 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			
							Initial	Final		Initial	Final	Control	
Albite, wet sieved (75-500 micrometers)													
Sample IDs: J-13 W-C.XXXX-20													
3545	0.98	20.05	3	N/A	19.88	3	20.78	20.78	N/A	49.6	48.7	50.5	-1×10^{-1}
3546	1.00	19.96	3	8.3	20.56	3	20.47	20.36	8.5	49.6	48.8	50.5	5×10^{-1}
3531	0.99	19.65	2	N/A	19.90	4	20.78	20.99	N/A	248.0	241.3	250.8	-3×10^{-1}
3532	1.01	19.69	2	8.4	18.72	4	19.14	20.52	8.3	248.0	241.0	250.8	-1
3517	0.99	19.71	2	N/A	20.22	4	21.03	20.89	N/A	2997.6	2965.2	3041.4	-2×10^{-1}
3518	1.00	19.68	2	8.3	20.21	4	20.84	20.82	8.4	2997.6	2933.7	3041.4	-2×10^{-1}
3503	0.99	19.73	2	N/A	20.02	3	20.72	20.78	N/A	6042.7	5881.9	6168.7	-5×10^{-3}
3504	1.00	19.74	2	8.3	19.98	3	20.46	20.51	8.3	6042.7	5834.7	6168.7	2×10^{-1}
3489	1.01	19.85	2	N/A	20.07	3	20.35	20.39	N/A	9143.0	8865.1	9246.2	1×10^{-1}
3490	1.01	19.85	2	8.4	20.10	3	20.37	20.61	8.4	9143.0	8895.4	9246.2	-2×10^{-1}
3473	1.01	19.83	2	N/A	20.05	3	20.36	20.44	N/A	12220.2	11902.8	12296.6	-6×10^{-2}
3474	0.99	19.77	2	8.4	20.14	3	20.81	20.88	8.3	12220.2	11849.7	12296.6	1×10^{-1}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Uranium Sorption Experiment #66

Atmosphere: CO_2 J-13 groundwater Temperature: 21 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid) Initial	Soln/solid ratio (mL/g of solid) Final	Final pH	Concentration of Uranium in solution (ppm) Initial	Concentration of Uranium in solution (ppm) Final	Concentration of Uranium in solution (ppm) Control	
Devitrified tuff (USW G4-268), wet sieved (75-500 micrometers); Sample IDs: J-13 G4-268-C.XXXX-20													
3804	0.99	19.45	3	7.3	19.87	3	21.04	20.91	7.1	0.049	0.043	0.048	2
3805	0.97	19.13	3	N/A	19.82	3	21.35	21.35	N/A	0.049	0.044	0.048	1
3790	1.00	19.51	3	7.3	14.60	3	18.42	18.45	7.1	0.12	0.08	0.11	3
3791	0.99	19.42	3	N/A	17.57	3	18.57	18.53	N/A	0.12	0.11	0.11	8×10^{-2}
3762	1.01	17.46	4	7.1	19.95	3	20.67	20.73	N/A	0.55	0.59	0.55	−2
3763	1.02	17.52	4	N/A	19.92	3	20.36	20.42	N/A	0.55	0.59	0.55	−2
3776	0.98	19.58	3	7.5	11.60	3	16.02	16.15	7.0	0.59	0.50	N/A	−2
3777	1.02	19.62	3	N/A	14.56	3	15.12	15.12	N/A	0.59	0.54	N/A	6×10^{-1}
3748	1.02	17.49	4	7.3	19.70	3	20.13	20.14	N/A	1.2	1.1	1.2	1
3749	0.97	17.48	4	N/A	19.93	3	21.37	21.40	N/A	1.2	1.1	1.2	1
3734	1.00	17.67	4	7.1	19.78	3	20.56	20.57	N/A	3.8	4.3	3.8	−3
3735	0.98	17.54	4	N/A	19.79	3	21.30	21.32	N/A	3.8	4.3	3.8	−4
3704	1.01	19.59	3	7.3	19.28	3	20.00	19.97	7.4	33	29	33	2
3705	1.00	19.58	3	N/A	19.80	3	20.83	20.84	N/A	33	32	33	-4×10^{-1}
Zeolitic tuff (USW G4-1510), wet sieved (75-500 micrometers); Sample IDs: J-13 G4-1510-C.XXXX-20													
3806	1.00	19.46	3	7.3	19.78	3	20.90	20.83	7.1	0.049	0.023	0.048	20
3807	0.98	19.37	3	N/A	19.85	3	21.47	21.47	N/A	0.049	0.024	0.048	20
3792	1.00	19.49	3	7.3	18.70	3	18.87	18.84	7.1	0.12	0.054	0.11	20
3793	0.98	19.63	3	N/A	18.52	3	19.43	19.42	N/A	0.12	0.056	0.11	20
3764	1.03	17.29	4	7.3	19.93	3	20.71	20.73	N/A	0.55	0.36	0.55	9
3765	0.97	17.30	4	N/A	19.96	3	21.67	21.70	N/A	0.55	0.38	0.55	9
3778	1.00	19.61	3	7.3	13.64	3	14.67	14.71	7.1	0.59	0.27	N/A	20
3779	1.03	19.60	3	N/A	14.68	3	15.44	15.43	N/A	0.59	0.28	N/A	10
3736	1.02	17.62	4	7.1	19.68	3	20.79	20.80	N/A	3.8	2.7	3.8	6
3737	1.00	17.75	4	N/A	19.67	3	20.87	20.90	N/A	3.8	2.7	3.8	6
3722	0.99	17.70	4	7.1	19.86	3	21.07	21.09	N/A	5.7	3.5	5.7	10
3723	0.98	17.64	4	N/A	19.89	3	21.52	21.54	N/A	5.7	3.7	5.7	10
3706	1.02	19.87	3	7.3	19.71	3	20.42	20.43	7.4	33	28	33	3
3707	0.82	19.78	3	N/A	19.49	3	25.29	25.30	N/A	33	29	33	2

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Uranium Sorption Experiment #66 continued

Atmosphere: CO_2 J-13 groundwater Temperature: 21 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid) Initial	Soln/solid ratio (mL/g of solid) Final	Final pH	Concentration of Uranium in solution (ppm) Initial	Concentration of Uranium in solution (ppm) Final	Concentration of Uranium in solution (ppm) Control	
Zeolitic tuff (USW G4-1529), wet sieved (75-500 micrometers); Sample IDs: J-13 G4-1529-C.XXXX.X-20													
3709.3	0.98	19.67	3	7.3	19.42	3	20.78	20.77	7.3	33	29	33	2
3709.4	0.98	19.79	3	N/A	19.40	3	20.75	20.76	N/A	33	25	33	5
Vitric tuff (USW GU3-1405), wet sieved (75-500 micrometers); Sample IDs: J-13 Gu3-1405-C.XXXX-20													
3808	1.00	19.47	3	7.3	19.80	3	20.79	20.81	7.1	0.049	0.041	0.048	3
3809	1.00	19.39	3	N/A	19.79	3	20.75	20.74	N/A	0.049	0.043	0.048	2
3794	1.01	19.60	3	7.3	16.07	3	17.20	17.14	7.1	0.12	0.13	0.11	−3
3795	1.04	19.27	3	N/A	17.54	3	18.01	18.01	N/A	0.12	0.10	0.11	7×10^{-1}
3766	1.02	17.34	4	7.2	19.93	3	20.58	20.61	N/A	0.55	0.57	0.55	−2
3767	1.02	17.32	4	N/A	19.88	3	20.62	20.64	N/A	0.55	0.56	0.55	−1
3752	1.01	17.58	4	7.2	20.02	3	20.88	20.94	N/A	1.2	1.5	1.2	−4
3753	1.01	17.36	4	N/A	19.94	3	20.60	20.67	N/A	1.2	1.2	1.2	-3×10^{-3}
3738	1.00	17.52	4	7.1	19.75	3	20.85	20.92	N/A	3.8	4.1	3.8	−3
3739	1.03	17.45	4	N/A	19.66	3	20.07	20.11	N/A	3.8	4.5	3.8	−4
3724	1.03	17.65	4	7.0	19.86	3	20.24	20.26	N/A	5.7	5.4	5.7	1×10^{-1}
3725	1.00	17.64	4	N/A	19.88	3	21.11	21.13	N/A	5.7	5.1	5.7	9×10^{-1}
3708	0.97	19.67	3	7.3	19.73	3	21.25	21.25	7.3	33	27	33	3
3709	0.98	19.87	3	N/A	19.18	3	20.42	20.43	N/A	33	32	33	-2×10^{-1}
Synthetic Hematite, not sieved; Sample IDs: J-13 C-C.XXXX-20													
3810	0.99	19.22	3	7.1	19.76	3	20.76	20.82	6.9	0.049	0.0006	0.048	2×10^{3}
3811	1.00	19.37	3	N/A	19.79	3	20.72	20.71	N/A	0.049	0.0	0.048	
3796	1.01	19.27	3	7.0	17.64	3	18.16	18.09	7.1	0.12	0.0005	0.11	4×10^{3}
3797	0.98	19.15	3	N/A	17.73	3	18.86	18.85	N/A	0.12	0.0001	0.11	2×10^{4}
3768	0.98	17.28	4	7.0	19.95	3	21.19	21.22	N/A	0.55	0.037	0.55	3×10^{2}
3769	0.99	17.34	4	N/A	19.87	3	21.12	21.16	N/A	0.55	0.037	0.55	3×10^{2}
3740	0.99	17.49	4	6.8	19.56	3	20.61	20.63	N/A	3.8	0.45	3.8	1×10^{2}
3741	0.99	17.39	4	N/A	19.72	3	20.69	20.70	N/A	3.8	0.40	3.8	2×10^{2}
3726	0.99	17.76	4	6.8	19.91	3	20.78	20.79	N/A	5.7	0.21	5.7	5×10^{2}
3727	1.01	17.78	4	N/A	19.93	3	20.51	20.52	N/A	5.7	0.29	5.7	4×10^{2}

Uranium Sorption Experiment #66 continued

Atmosphere: CO_2 J-13 groundwater Temperature: 21 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Concentration of Uranium in solution (ppm)			
							Initial	Final		Initial	Final	Control	
Quartz, wet sieved (75-500 micrometers); Sample IDs: J-13 M-C.XXXX-20													
3812	0.99	19.24	3	7.3	19.79	3	20.76	20.77	7.1	0.049	0.046	0.048	7×10^{-1}
3813	1.02	19.38	3	N/A	19.82	3	20.29	20.29	N/A	0.049	0.051	0.048	-2
3798	1.01	19.30	3	7.2	17.73	3	18.27	18.32	7.1	0.12	0.11	0.11	9×10^{-1}
3799	1.01	20.11	3	N/A	17.68	3	18.21	18.21	N/A	0.12	0.11	0.11	1×10^{-1}
3771	1.02	17.37	4	N/A	19.78	3	20.28	20.30	N/A	0.55	0.51	0.55	6×10^{-1}
3784	1.03	19.36	3	7.3	14.57	3	14.99	15.09	7.1	0.59	0.54	N/A	5×10^{-1}
3756	1.02	17.55	4	7.1	19.91	3	20.29	20.31	N/A	1.2	1.1	1.2	1
3757	0.99	17.45	4	N/A	19.86	3	20.99	21.00	N/A	1.2	1.2	1.2	2×10^{-1}
3742	1.03	17.48	4	7.1	19.61	3	19.88	19.90	N/A	3.8	4.0	3.8	-2
3743	0.99	17.56	4	N/A	19.52	3	20.36	20.38	N/A	3.8	3.8	3.8	-9×10^{-1}
3728	1.02	17.80	4	7.0	19.89	3	20.25	20.26	N/A	5.7	5.1	5.7	2
3729	1.02	17.57	4	N/A	19.82	3	20.04	20.05	N/A	5.7	5.1	5.7	1
3712	1.00	19.64	3	7.3	19.52	3	20.23	20.23	7.4	33	32	33	-3×10^{-1}
3713	0.98	19.65	3	N/A	19.43	3	20.53	20.54	N/A	33	34	33	-1
Sodium form of Clinoptilolite, not sieved; Sample IDs: J-13 G1-C.XXXX-20													
3814	0.98	19.45	3	7.4	18.87	3	20.21	20.14	7.2	0.049	0.020	0.048	30
3815	1.02	18.55	3	N/A	19.15	3	19.90	19.89	N/A	0.049	0.021	0.048	20
3800	0.99	19.28	3	7.4	17.44	3	18.86	18.99	7.2	0.12	0.050	0.11	20
3801	0.99	19.16	3	N/A	17.38	3	18.65	18.64	N/A	0.12	0.052	0.11	20
3772	1.00	17.39	4	7.4	19.85	3	21.03	21.09	N/A	0.55	0.31	0.55	10
3773	1.00	17.44	4	N/A	19.76	3	20.85	20.89	N/A	0.55	0.32	0.55	10
3786	0.99	19.40	3	7.4	14.62	3	15.89	15.90	7.1	0.59	0.21	N/A	30
3787	0.99	19.25	3	N/A	14.57	3	15.76	15.75	N/A	0.59	0.28	N/A	20
3758	1.00	17.44	4	7.3	19.70	3	20.86	20.87	N/A	1.2	0.71	1.2	10
3759	0.99	17.49	4	N/A	19.77	3	21.19	21.21	N/A	1.2	0.79	1.2	10
3744	0.98	17.54	4	7.1	19.48	3	21.04	21.07	N/A	3.8	2.9	3.8	5
3745	1.00	17.55	4	N/A	19.59	3	20.74	20.78	N/A	3.8	2.7	3.8	7
3730	1.01	17.68	4	7.2	19.90	3	20.68	20.71	N/A	5.7	4.0	5.7	7
3731	1.01	17.65	4	N/A	19.71	3	20.88	20.90	N/A	5.7	3.7	5.7	9
3714	1.00	19.55	3	7.5	19.58	3	20.75	20.76	7.4	33	30	33	8×10^{-1}
3715	1.01	19.65	3	N/A	19.39	3	20.32	20.29	N/A	33	31	33	2×10^{-1}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Uranium Sorption Experiment #66 continued

Atmosphere: CO_2 J-13 groundwater Temperature: 21 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid) Initial	Soln/solid ratio (mL/g of solid) Final	Final pH	Concentration of Uranium in solution (ppm) Initial	Concentration of Uranium in solution (ppm) Final	Concentration of Uranium in solution (ppm) Control	
Albite, wet sieved (75-500 micrometers)													
Sample IDs: J-13 W-C.XXXX-20													
3802	1.00	19.00	3	7.4	16.73	3	17.58	17.13	7.1	0.12	0.11	0.11	1
3803	1.01	18.99	3	N/A	17.57	3	18.34	18.35	N/A	0.12	0.11	0.11	-4×10^{-1}
3774	0.99	17.38	4	7.2	19.76	3	20.96	20.99	N/A	0.55	0.49	0.55	2
3775	1.02	17.41	4	N/A	19.65	3	19.99	20.03	N/A	0.55	0.53	0.55	3×10^{-2}
3788	1.01	19.49	3	7.3	14.57	3	15.26	15.35	6.9	0.59	0.50	N/A	2
3789	0.98	19.26	3	N/A	14.63	3	15.82	15.81	N/A	0.59	0.53	N/A	1
3760	1.03	17.48	4	7.1	19.68	3	19.97	20.02	N/A	1.2	1.1	1.2	4×10^{-1}
3761	1.03	17.35	4	N/A	19.70	3	19.90	19.92	N/A	1.2	1.2	1.2	2×10^{-1}
3746	1.04	17.50	4	7.3	19.57	3	19.61	19.62	N/A	3.8	4.0	3.8	-2
3747	1.01	17.55	4	N/A	19.47	3	20.06	20.07	N/A	3.8	3.7	3.8	-4×10^{-1}
3732	0.99	17.43	4	7.1	19.77	3	20.64	20.67	N/A	5.7	5.3	5.7	8×10^{-1}
3733	1.02	17.57	4	N/A	19.75	3	19.91	19.96	N/A	5.7	5.6	5.7	-4×10^{-1}
3716	0.99	19.39	3	7.4	19.48	3	20.57	20.53	7.3	33	33	33	-7×10^{-1}
3717	1.01	19.55	3	N/A	19.39	3	20.10	20.11	N/A	33	39	33	-4

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Uranium Sorption Experiment #65

Atmosphere: air | J-13 groundwater | Temperature range: 22–25 ˚C | Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Concentration of Uranium in solution (ppm)			
							Initial	Final		Initial	Final	Control	
Devitrified tuff (USW G4-268), wet sieved (75-500 micrometers)													
Sample IDs: J-13 G4-268-C.XXXX-20													
3733	1.00	19.73	4	8.2	20.07	4	20.75	20.60	8.3	0.056	0.047	0.055	3
3734	1.01	19.75	4	N/A	20.10	4	20.56	20.35	N/A	0.056	0.049	0.055	2
3719	0.99	19.76	4	N/A	19.96	4	20.79	20.69	N/A	0.099	0.088	0.10	2
3720	1.00	19.70	4	8.3	20.08	4	20.70	20.46	8.1	0.099	0.090	0.10	2
3677	0.99	19.61	2	N/A	20.31	3	21.12	21.12	N/A	0.12	0.13	0.12	–2
3678	1.02	19.70	2	8.3	20.21	3	20.56	20.50	8.2	0.12	0.11	0.12	6×10^{-1}
3705	1.02	19.77	3	8.4	20.13	4	20.42	20.31	N/A	0.21	0.20	0.22	6×10^{-1}
3706	1.01	19.74	3	N/A	20.20	4	20.62	20.46	8.2	0.21	0.20	0.22	7×10^{-1}
3691	0.99	19.75	2	N/A	20.00	3	20.78	20.68	N/A	0.57	0.62	0.57	–2
3692	1.02	19.72	2	8.3	20.09	3	20.30	20.14	8.2	0.57	0.61	0.57	–2
3649	1.01	20.00	2	N/A	20.13	3	20.65	20.53	N/A	1.2	1.0	1.2	3
3650	0.99	20.03	2	8.4	20.25	3	21.21	21.05	8.4	1.2	1.0	1.2	4
3663	1.00	19.80	2	N/A	20.08	3	20.75	20.63	N/A	3.8	4.1	N/A	–2
3664	0.99	19.81	2	8.4	20.06	3	20.98	20.91	8.3	3.8	4.1	N/A	–2
3633	0.99	19.73	3	N/A	20.19	3	21.02	21.03	N/A	32	28	30	2
3634	1.01	19.70	3	8.3	20.27	3	20.71	20.65	8.6	32	28	30	2
Devitrified tuff (USW G4-272), wet sieved (75-500 micrometers)													
Sample IDs: J-13 G4-272X													
A	1.00	19.90	4	8.3	20.10	4	20.74	20.59	8.3	0.056	0.048	0.055	3
B	1.02	19.98	4	N/A	20.17	4	20.41	20.27	N/A	0.056	0.044	0.055	5
A	1.01	20.03	3	8.3	20.19	3	20.61	20.51	8.5	32	32	30	-7×10^{-1}
B	1.01	19.96	3	N/A	20.16	3	20.65	20.58	N/A	32	29	30	9×10^{-1}
Zeolitic tuff (USW G4-1503), wet sieved (75-500 micrometers)													
Sample IDs: J13 G4-1503X													
A	1.00	20.05	4	8.2	20.13	4	20.95	20.75	8.3	0.056	0.026	0.055	20
B	1.01	20.06	4	N/A	20.25	4	21.01	20.83	N/A	0.056	0.025	0.055	20
A	1.02	19.83	3	8.2	20.21	3	20.65	20.54	8.5	32	28	30	2
B	1.01	19.90	3	N/A	20.22	3	20.83	20.72	N/A	32	27	30	2

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Uranium Sorption Experiment #65 continued

Atmosphere: air J-13 groundwater Temperature range: 22–25 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid) Initial	Soln/solid ratio (mL/g of solid) Final	Final pH	Concentration of Uranium in solution (ppm) Initial	Concentration of Uranium in solution (ppm) Final	Concentration of Uranium in solution (ppm) Control	
Zeolitic tuff (USW G4-1510), wet sieved (75-500 micrometers)													
Sample IDs: J-13 G4-1510-C.XXXX-20													
3735	1.00	19.76	4	8.1	20.16	4	20.96	20.74	8.3	0.056	0.030	0.055	20
3736	1.00	19.71	4	N/A	20.07	4	20.89	20.71	N/A	0.056	0.028	0.055	20
3721	1.02	19.75	4	N/A	20.08	4	20.48	20.31	N/A	0.099	0.047	0.10	20
3722	1.01	19.73	4	8.3	20.25	4	20.81	20.54	8.0	0.099	0.047	0.10	20
3679	0.99	19.71	2	N/A	20.25	3	21.25	21.16	N/A	0.12	0.071	0.12	10
3680	1.00	19.76	2	8.2	20.26	3	21.06	21.05	8.3	0.12	0.067	0.12	10
3707	1.00	19.73	3	8.3	20.01	4	20.81	20.71	N/A	0.21	0.13	0.22	10
3708	1.00	19.71	3	N/A	20.19	4	20.98	20.84	8.2	0.21	0.13	0.22	10
3693	0.99	19.74	2	N/A	19.98	3	21.01	20.90	N/A	0.57	0.38	0.57	9
3694	1.00	19.84	2	8.1	19.92	3	20.74	20.67	8.3	0.57	0.38	0.57	9
3651	1.00	19.88	2	N/A	20.21	3	20.91	20.85	N/A	1.2	0.69	1.2	10
3652	1.00	19.87	2	8.2	20.01	3	20.76	20.75	8.3	1.2	0.67	1.2	20
3665	1.00	19.82	2	8.3	20.11	3	20.91	20.87	N/A	3.8	2.8	N/A	7
3666	0.99	19.80	2	N/A	20.14	3	21.15	21.08	8.2	3.8	3.0	N/A	5
3635	1.01	19.68	3	N/A	20.30	3	20.87	20.80	N/A	32	26	30	4
3636	1.01	19.76	3	8.2	20.21	3	20.81	20.78	8.5	32	25	30	4
Zeolitic tuff (USW G4-1529), wet sieved (75-500 micrometers)													
Sample IDs: J13 G4-1529X													
A	1.00	19.91	4	8.2	20.22	4	20.97	20.76	8.3	0.056	0.028	0.055	20
B	1.01	19.93	4	N/A	20.20	4	20.84	20.64	N/A	0.056	0.026	0.055	20
A	0.98	19.94	3	8.3	20.19	3	21.28	21.17	8.5	32	28	30	3
B	0.42	19.97	3	N/A	20.21	3	49.09	48.85	N/A	32	30	30	2

Uranium Sorption Experiment #65 continued

Atmosphere: air | J-13 groundwater | Temperature range: 22–25 ˚C | Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Concentration of Uranium in solution (ppm)			
							Initial	Final		Initial	Final	Control	
Vitric tuff (USW GU3-1405), wet sieved (75-500 micrometers); Sample IDs: J-13 Gu3-1405-C.XXXX-20													
3737	1.00	19.80	4	8.3	20.12	4	20.93	20.81	8.3	0.056	0.054	0.055	2×10^{-1}
3738	1.01	19.82	4	N/A	20.18	4	20.72	20.49	N/A	0.056	0.053	0.055	6×10^{-1}
3723	1.01	19.75	4	N/A	20.10	4	20.71	20.59	N/A	0.099	0.078	0.10	5
3724	1.00	20.76	4	8.3	20.13	4	20.93	19.77	8.1	0.099	0.080	0.10	5
3681	0.99	19.76	2	N/A	20.30	3	21.31	21.29	N/A	0.12	0.12	0.12	−1
3682	1.01	19.79	2	8.3	20.29	3	20.88	20.79	8.3	0.12	0.13	0.12	−2
3709	1.00	19.62	3	8.4	20.10	4	20.80	20.72	N/A	0.21	0.20	0.22	2×10^{-1}
3710	0.99	19.74	3	N/A	20.15	4	21.07	20.94	8.3	0.21	0.21	0.22	-2×10^{-1}
3695	1.00	19.72	2	N/A	19.89	3	20.65	20.55	N/A	0.57	0.65	0.57	−3
3696	1.01	19.70	2	8.3	20.03	3	20.49	20.30	8.2	0.57	0.57	0.57	-7×10^{-1}
3653	1.00	19.89	2	N/A	20.12	3	20.91	20.85	N/A	1.2	1.0	1.2	3
3654	0.98	19.88	2	8.3	20.14	3	21.41	21.32	8.3	1.2	1.1	1.2	2
3667	0.99	19.88	2	8.4	20.06	3	21.15	21.09	8.2	3.8	4.1	N/A	−2
6769	0.99	19.90	2	N/A	20.12	3	21.07	21.02	N/A	3.8	4.1	N/A	−2
3637	0.99	19.71	3	N/A	20.24	3	21.13	21.06	N/A	32	28	30	2
3638	1.00	19.70	3	8.4	19.92	3	20.64	20.62	8.6	32	26	30	4
Synthetic Hematite, not sieved; Sample IDs: J-13 C-C.XXXX-20													
3739	1.01	19.89	4	7.5	19.98	4	20.32	20.26	7.8	0.056	0.0031	0.055	3×10^{2}
3740	1.00	19.93	4	N/A	20.04	4	20.60	20.40	N/A	0.056	0.0020	0.055	5×10^{2}
3725	1.01	19.90	4	N/A	20.11	4	20.43	20.24	N/A	0.099	0.0	0.10	
3726	1.02	19.84	4	8.0	20.00	4	20.09	19.90	7.7	0.099	0.0	0.10	
3711	0.99	19.81	3	8.0	20.09	4	20.78	20.78	N/A	0.21	0.0050	0.22	8×10^{2}
3712	0.99	19.86	3	N/A	20.05	4	20.79	20.76	7.7	0.21	0.0050	0.22	8×10^{2}
3655	0.99	19.89	2	N/A	20.09	3	20.80	20.78	N/A	1.2	0.0092	1.2	3×10^{3}
3656	1.00	19.89	2	7.5	20.07	3	20.59	20.55	8.2	1.2	0.0082	1.2	3×10^{3}
3669	0.99	19.78	2	N/A	20.16	3	20.85	20.74	8.1	3.8	0.051	N/A	2×10^{3}
3670	0.99	19.90	2	8.3	20.04	3	20.68	20.58	N/A	3.8	0.053	N/A	1×10^{3}
3639	1.01	19.59	3	N/A	20.22	3	20.65	20.60	N/A	32	10	30	4×10^{1}
3640	1.01	19.61	3	7.6	20.20	3	20.55	20.49	8.3	32	10	30	4×10^{1}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Uranium Sorption Experiment #65 continued

Atmosphere: air J-13 groundwater Temperature range: 22–25 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid) Initial	Soln/solid ratio (mL/g of solid) Final	Final pH	Concentration of Uranium in solution (ppm) Initial	Concentration of Uranium in solution (ppm) Final	Concentration of Uranium in solution (ppm) Control	
Quartz, wet sieved (75-500 micrometers)													
Sample IDs: J-13 M-C.XXXX-20													
3741	1.00	19.78	4	8.3	20.04	4	20.54	20.38	8.3	0.056	0.051	0.055	2
3742	1.00	19.78	4	N/A	20.18	4	20.65	20.50	N/A	0.056	0.050	0.055	2
3727	1.01	19.78	4	N/A	20.16	4	20.40	20.27	N/A	0.099	0.097	0.10	2×10^{-1}
3728	1.00	19.82	4	8.3	20.15	4	20.72	20.57	8.1	0.099	0.093	0.10	9×10^{-1}
3713	0.99	19.87	3	8.4	20.17	4	20.86	20.72	N/A	0.21	0.21	0.22	-4×10^{-1}
3714	0.99	19.81	3	N/A	19.96	4	20.62	20.53	8.3	0.21	0.21	0.22	-4×10^{-1}
3657	1.02	19.89	2	N/A	20.10	3	20.18	20.07	N/A	1.2	1.2	1.2	-3×10^{-1}
3658	0.99	19.92	2	8.4	20.16	3	20.87	20.74	8.3	1.2	1.2	1.2	-3×10^{-1}
3671	0.99	19.88	2	N/A	20.10	3	20.82	20.75	8.3	3.8	4.1	N/A	-2
3672	1.01	19.85	2	8.2	20.14	3	20.46	20.32	N/A	3.8	3.9	N/A	-1
3641	1.00	19.61	3	N/A	20.21	3	20.75	20.69	N/A	32	30	30	4×10^{-1}
3642	1.00	19.66	3	8.5	20.22	3	20.68	20.62	8.6	32	31	30	-1×10^{-1}
Sodium form of Clinoptilolite, not sieved													
Sample IDs: J-13 G1-C.XXXX-20													
3743	0.99	19.67	4	8.2	20.29	4	21.18	21.03	8.4	0.056	0.036	0.055	10
3744	0.99	19.68	4	N/A	20.23	4	21.19	20.99	N/A	0.056	0.032	0.055	20
3729	1.02	19.71	4	N/A	20.19	4	20.64	20.43	N/A	0.099	0.062	0.10	10
3730	0.99	19.69	4	8.3	20.20	4	21.30	21.16	8.1	0.099	0.065	0.10	10
3715	0.99	19.68	3	N/A	20.04	4	20.99	20.92	N/A	0.21	0.15	0.22	7
3716	1.01	19.70	3	8.3	20.09	4	20.65	20.47	8.3	0.21	0.15	0.22	8
3701	0.99	19.79	2	N/A	19.83	3	20.81	20.74	N/A	0.57	0.47	0.57	4
3702	0.99	19.80	2	8.4	20.06	3	21.05	20.88	8.3	0.57	0.45	0.57	5
3659	0.99	19.98	2	N/A	19.98	3	21.01	20.98	N/A	1.2	0.90	1.2	6
3660	1.00	19.97	2	8.5	20.08	3	20.90	20.78	8.3	1.2	0.92	1.2	5
3673	0.99	19.93	2	N/A	20.05	3	21.05	21.04	8.3	3.8	3.1	N/A	4
3674	0.99	19.90	2	8.5	20.18	3	21.32	21.15	N/A	3.8	3.1	N/A	4
3643	1.02	19.74	3	N/A	20.18	3	20.60	20.51	N/A	32	30	30	6×10^{-1}
3644	1.01	19.78	3	8.5	20.14	3	20.99	20.94	8.6	32	31	30	-6×10^{-1}

Uranium Sorption Experiment #65 continued

Atmosphere: air | J-13 groundwater | Temperature range: 22–25 ˚C | Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Uranium Solution SorptionTreatment								Distr. Coeff., K_d (mL/g)
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Concentration of Uranium in solution (ppm)			
							Initial	Final		Initial	Final	Control	
Albite, wet sieved (75-500 micrometers)													
Sample IDs: J-13 W-C.XXXX-20													
3745	1.00	19.67	4	8.4	20.24	4	20.70	20.48	8.4	0.056	0.052	0.055	1
3746	1.00	19.72	4	N/A	20.19	4	20.69	20.50	N/A	0.056	0.054	0.055	6×10^{-1}
3731	1.01	19.68	4	N/A	20.14	4	20.43	20.27	N/A	0.099	0.099	0.1	-2×10^{-1}
3732	0.99	19.72	4	8.2	20.20	4	20.84	20.73	8.1	0.099	0.097	0.1	2×10^{-1}
3689	1.00	19.87	2	N/A	20.25	3	20.72	20.71	N/A	0.12	0.13	0.12	-2
3690	0.98	19.90	2	8.3	20.29	3	21.18	21.12	8.4	0.12	0.12	0.12	-4×10^{-1}
3717	0.99	19.65	3	N/A	20.08	4	20.75	20.58	N/A	0.21	0.21	0.22	6×10^{-2}
3718	1.00	19.67	3	8.3	19.98	4	20.47	20.39	8.3	0.21	0.22	0.22	-1
3703	0.99	19.78	2	N/A	19.92	3	20.63	20.50	N/A	0.57	0.53	0.57	1
3704	1.01	19.85	2	8.3	19.93	3	20.22	20.11	8.2	0.57	0.53	0.57	8×10^{-1}
3661	0.96	19.85	2	N/A	19.27	3	20.66	21.44	N/A	1.2	1.2	1.2	-1
3662	1.02	19.87	2	8.4	19.98	3	20.08	20.05	8.2	1.2	1.2	1.2	8×10^{-2}
3675	0.99	19.86	2	N/A	20.07	3	20.83	20.81	8.3	3.8	3.8	N/A	-4×10^{-1}
3676	1.00	19.94	2	8.5	18.60	3	19.02	19.00	N/A	3.8	3.8	N/A	-5×10^{-1}
3645	1.00	19.77	3	N/A	20.14	3	20.59	20.56	N/A	32	33	30	-1
3646	1.00	19.77	3	8.4	20.10	3	20.60	20.54	8.6	32	30	30	7×10^{-1}

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Plutonium-239 Sorption Experiment #68

Atmosphere: CO_2 J-13 groundwater Temperature: 20 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Plutonium-239 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g), based on:	
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			Control soln	Initial soln
							Initial	Final		Initial	Final	Control		
Devitrified tuff (USW G4-268), wet sieved (75-500 micrometers); Sample IDs: J-13 G-14-268-C.XXXX-20														
3240	0.99	18.70	3	7.2	19.58	3	20.72	20.73	7.1	45.6	30.1	44.0	8	9
3241	0.98	19.40	3	N/A	19.73	3	21.19	21.18	N/A	45.6	28.0	44.0	10	10
3192	1.03	19.64	3	7.2	19.58	3	20.24	20.25	7.1	5920.8	3470.3	5525.0	10	10
3193	1.03	19.65	3	N/A	19.49	3	19.91	19.92	N/A	5920.8	3820.7	5525.0	7	9
3176	1.00	19.71	3	6.9	19.87	3	20.82	20.84	7.1	7833.7	5530.7	7335.0	6	7
3177	1.00	19.65	3	N/A	19.94	3	21.04	21.06	N/A	7833.7	5774.9	7335.0	4	6
Zeolitic tuff (USW G4-1510), wet sieved (75-500 micrometers); Sample IDs: J-13 G-14-1510-C.XXXX-20														
3226	1.01	19.42	3	7.3	19.84	3	20.90	20.92	7.2	232.1	10.2	224.3	4×10^2	4×10^2
3227	1.02	19.39	3	N/A	19.93	3	20.74	20.76	N/A	232.1	11.3	224.3	4×10^2	4×10^2
3178	0.97	19.68	3	7.2	20.01	3	21.73	21.74	6.9	7833.7	1321.2	7335.0	90	1×10^2
3179	1.00	19.73	3	N/A	19.95	3	21.07	21.09	N/A	7833.7	989.7	7335.0	1×10^2	1×10^2
Vitric tuff (USW GU3-1405), wet sieved (75-500 micrometers); Sample IDs: J-13 G-1u3-1405-C.XXXX-20														
3244	1.03	19.35	3	7.2	19.80	3	20.21	20.23	7.1	45.6	9.0	44.0	70	80
3245	1.02	17.25	3	N/A	19.74	3	20.42	20.41	N/A	45.6	10.3	44.0	60	70
3228	1.00	19.39	3	7.2	19.74	3	20.69	20.68	7.1	232.1	54.2	224.3	60	60
3229	0.97	19.52	3	N/A	19.71	3	21.46	21.48	N/A	232.1	63.7	224.3	50	50
3196	0.96	19.58	3	7.3	19.45	3	21.39	21.41	7.0	5920.8	2272.9	5525.0	30	30
3197	0.99	19.60	3	N/A	19.25	3	20.53	20.54	N/A	5920.8	2205.7	5525.0	30	30
3180	0.99	19.80	3	7.1	19.93	3	21.12	21.13	7.0	7833.7	3518.9	7335.0	20	20
3181	1.01	19.86	3	N/A	19.88	3	20.68	20.70	N/A	7833.7	3562.5	7335.0	20	20
Synthetic Hematite, not sieved; Sample IDs: J-13 C-C.XXXX-20														
3246	0.99	19.26	3	7.1	19.67	3	20.74	20.75	7.0	45.6	0.3	44.0	3×10^3	4×10^3
3247	0.98	18.46	3	N/A	19.68	3	21.01	21.02	N/A	45.6	0.3	44.0	3×10^3	3×10^3
3230	1.00	18.95	3	7.1	19.68	3	20.53	20.55	7.0	232.1	0.1	224.3	6×10^4	6×10^4
3231	0.99	18.10	3	N/A	19.75	3	20.84	20.86	N/A	232.1	0.1	224.3	5×10^4	6×10^4
3198	0.97	19.65	3	7.1	18.84	3	20.30	20.33	7.0	5920.8	0.6	5525.0	2×10^5	2×10^5
3199	1.02	19.63	3	N/A	19.40	3	19.89	19.90	N/A	5920.8	0.9	5525.0	1×10^5	1×10^5
3182	1.02	19.84	3	7.1	19.86	3	20.18	20.19	6.9	7833.7	2.9	7335.0	5×10^4	5×10^4
3183	1.02	19.75	3	N/A	19.87	3	20.41	20.41	N/A	7833.7	2.1	7335.0	7×10^4	7×10^4

Appendix A: Batch Sorption Results for Neptunium, Uranium, and Plutonium

Plutonium-239 Sorption Experiment #68 continued

Atmosphere: CO_2 J-13 groundwater Temperature: 20 ˚C Binder where data are located: LA-CST10-NBK-94-004

XXXX Part of Sample ID	Mass of mineral (g)	J-13 Pretreatment			Plutonium-239 Solution Sorption Treatment								Distr. Coeff., K_d (mL/g), based on:	
		Mass of added water (g)	Period (days)	Final pH	Mass of added soln (g)	Period (days)	Soln/solid ratio (mL/g of solid)		Final pH	Alpha activity in solution (cpm/g of soln)			Control soln	Initial soln
							Initial	Final		Initial	Final	Control		
Sodium form of Clinoptilolite, not sieved														
Sample IDs: J-13 G-1-C.XXXX-20														
3234	1.00	18.28	3	7.3	19.70	3	20.87	20.87	7.2	232.1	5.9	224.3	7×10^2	8×10^2
3235	1.00	19.33	3	N/A	19.68	3	21.01	21.04	N/A	232.1	5.5	224.3	8×10^2	8×10^2
3202	1.02	19.53	3	7.4	19.52	3	20.40	20.42	6.7	5920.8	244.5	5525.0	4×10^2	4×10^2
3203	1.00	19.45	3	N/A	19.38	3	20.36	20.37	N/A	5920.8	220.1	5525.0	5×10^2	5×10^2
3186	1.00	19.51	3	7.3	19.72	3	21.05	21.06	7.2	7833.7	616.0	7335.0	2×10^2	2×10^2
3187	1.01	19.71	3	N/A	19.62	3	20.88	20.89	N/A	7833.7	672.2	7335.0	2×10^2	2×10^2
Quartz, wet sieved (75-500 micrometers)														
Sample IDs: J-13 M-C.XXXX-20														
3248	1.04	19.06	3	7.2	19.63	3	19.70	19.71	7.0	45.6	39.4	44.0	1	2
3249	1.03	19.20	3	N/A	19.64	3	19.83	19.84	N/A	45.6	41.1	44.0	6×10^{-1}	1
3232	0.99	18.39	3	7.2	19.71	3	20.72	20.73	7.0	232.1	204.0	224.3	1	2
3233	0.99	16.50	3	N/A	19.72	3	20.75	20.76	N/A	232.1	210.2	224.3	5×10^{-1}	1
3200	1.03	19.69	3	7.2	19.45	3	19.84	19.85	7.0	5920.8	4595.1	5525.0	3	4
3201	0.97	19.66	3	N/A	19.39	3	20.88	20.89	N/A	5920.8	4568.1	5525.0	3	5
3184	1.06	19.77	3	7.1	19.83	3	19.44	19.42	7.0	7833.7	6475.3	7335.0	2	3
3185	0.95	19.76	3	N/A	19.75	3	21.55	21.54	N/A	7833.7	6659.3	7335.0	1	3
Albite, wet sieved (75-500 micrometers)														
Sample IDs: J-13 W-C.XXXX-20														
3252	1.00	19.18	3	7.2	19.62	3	20.46	20.47	7.1	45.6	9.1	44.0	70	80
3253	1.02	18.80	3	N/A	19.42	3	19.99	19.99	N/A	45.6	5.4	44.0	100	100
3236	0.97	19.45	3	7.2	19.57	3	20.98	21.00	7.0	232.1	47.4	224.3	70	80
3237	0.95	19.50	3	N/A	19.48	3	21.43	21.45	N/A	232.1	63.3	224.3	50	50
3204	0.95	19.57	3	7.2	18.67	3	20.72	20.73	7.1	5920.8	2028.1	5525.0	30	40
3205	1.02	19.59	3	N/A	19.47	3	19.89	19.90	N/A	5920.8	2364.1	5525.0	20	30

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