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Brian A. Powell^a; Linfeng Rao^a; Kenneth L. Nash^b

^a Lawrence Berkeley National Laboratory, Berkeley, CA, USA ^b Washington State University, Pullman, WA, USA

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Examination of Uranium(VI) Leaching during Ligand Promoted Dissolution of Waste Tank Sludge Surrogates

Brian A. Powell,¹ Linfeng Rao,¹ and Kenneth. L. Nash²

¹Lawrence Berkeley National Laboratory, Berkeley, CA, USA

²Washington State University, Pullman, WA, USA

Abstract: The dissolution of synthetic boehmite (γ -AlOOH) by 1-hydroxyethane-1, 1-diphosphonic acid (HEDPA) was examined in a series of batch adsorption/dissolution experiments. Additionally, the leaching behavior of $^{233}\text{U(VI)}$ from boehmite was examined as a function of pH and HEDPA concentration. The results are discussed in terms of sludge washing procedures that may be utilized during underground tank waste remediation. In the pH range 4 to 10, complexation of Al(III) by HEDPA significantly enhanced dissolution of boehmite. This phenomenon was especially pronounced in the neutral pH region where the solubility of aluminum, in the absence of complexants, is limited by the formation of sparsely soluble aluminum hydroxides. At pH higher than 10, the dissolution of synthetic boehmite was inhibited by HEDPA, likely due to sorption of Al(III):HEDPA complexes. The addition of HEDPA to equilibrated U(VI)-synthetic boehmite suspensions yielded an increase in the aqueous phase uranium concentration. Partitioning of uranium between the solid and aqueous phase is described in terms of U(VI):HEDPA speciation and dissolution of the boehmite solid phase.

Keywords: Actimide, boehmite, Hanford, HEDPA, sludge, tank waste, uranium

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Address correspondence to Brian A. Powell, Department of Environmental Engineering and Earth Sciences, Clemson University, Anderson, SC 29625. E-mail: bpowell@clemson.edu

INTRODUCTION

The underground storage tanks at the Hanford Site in Washington State, U.S.A. contain the byproducts from a number of spent nuclear fuel reprocessing processes including the bismuth phosphate (BiPO_4), Redox and PUREX processes (1). Over time the waste has stratified into a salt cake, a supernatant phase, and an underlying sludge phase. Insoluble aluminum oxides make up a significant fraction of the sludge phase (1). Most of the transuranics have partitioned into the sludge phase, making vitrification of the sludge phase for geologic disposal a plausible treatment process (1). However, vitrification will be prohibitively expensive due to the large volume of the sludge phase. Therefore, reduction of the volume of the sludge through dissolution of the aluminum oxides presents a favorable alternative.

Various strategies of sludge leaching have been proposed and tested with sludge stimulants, including leaching with increasingly aggressive procedures ($0.01 \text{ M NaNO}_2 + 0.01 \text{ M NaOH}$, 3 M NaOH , $0.05 \text{ M glycolic acid} + 0.10 \text{ M NaOH}$, 0.10 M HNO_3 , 2.0 M HNO_3 , 0.5 M HEDPA (1-hydroxyethane-1,1-diphosphonic acid)) (1–3). It was found that the aluminosilicates cannot be removed using $0.01 \text{ M NaNO}_2 + 0.01 \text{ M NaOH}$ and no single treatment achieved complete dissolution (1–3). Data from these experiments suggest that HEDPA could be an effective leachant to reduce the volume of waste sludge. In order to develop an efficient waste treatment process, further studies are needed to understand the extent of sludge phase dissolution and the partitioning of actinides (U, Np, and Pu) during the leaching process.

As a diphosphonic acid, HEDPA is known to form strong complexes with metal ions including actinides and aluminum across a wide pH range (4–6). In the absence of complexing ligands, aluminum (hydro)oxides are only sparingly soluble. Therefore, the formation of Al(III):HEDPA complexes will promote sludge dissolution. However, partitioning of the actinides between the sludge and aqueous phase must also be understood in order to evaluate the viability of HEDPA for sludge washing. Formation of U(VI):HEDPA complexes affects the partitioning of uranium during the leaching of waste sludge with HEDPA. Previous experiments demonstrated that greater than 95% of the total uranium was leached from BiPO_4 , Redox, and PUREX sludge waste simulants by washing the sludge with 0.5 M HEDPA (3). Recently, a number of $(\text{UO}_2)_m \text{H}_h \text{L}_l$ (where L stands for the fully deprotonated HEDPA anion) complexes, ranging from cationic to anionic, were identified in the pH region from 2 to 12 using potentiometry, calorimetry, and spectroscopic techniques (4). The thermodynamic data provided by Reed et al. (4) are used below to describe the observed behavior of uranium in synthetic boehmite suspensions amended with HEDPA.

The objective of this study was to investigate the ability of HEDPA to accomplish dissolution of synthetic boehmite and to examine the leaching of uranium during dissolution. A companion study was also performed to examine the leaching of neptunium and plutonium under similar conditions (7). The data are expected to assist in the development of remediation strategies to be used during waste tank sludge washing.

MATERIALS AND METHODS

Solid Phase Characterization

The alumina used in this work was obtained from SASOL (trade name CATAPAL® B). Powder X-ray diffraction data, determined using a Siemens D-500 Diffractometer, indicate that the material has the crystal structure of boehmite (γ -AlOOH) although a significant amorphous character was indicated through broad peaks in the XRD spectra. A surface area of $354 \text{ m}^2 \text{ g}^{-1}$ was measured by $\text{N}_2(\text{g})$ adsorption using a Micrometrics BET Surface Area Analyzer. Potentiometric titrations were conducted with 100 g L^{-1} boehmite suspensions in 0.01, 0.1, and 1.0 M NaCl to determine the point-of-zero-salt-effect (pzse). The titration results are shown in Fig. 1. The boehmite surface has a net positive or

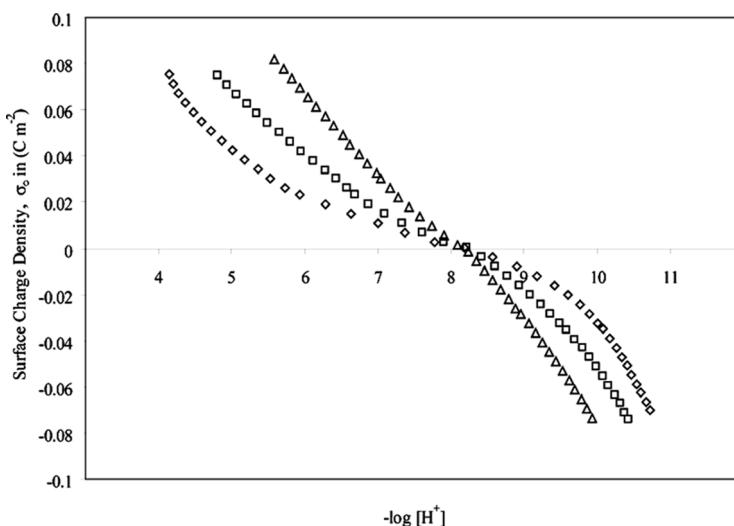


Figure 1. Potentiometric titrations of 100 g L^{-1} boehmite suspensions in 0.01 M NaCl (\diamond), 0.10 M NaCl (\square), and 1.0 M NaCl (\triangle).

negative charge due to protonation or deprotonation of surface hydroxyl groups with changing pH. The intersection of the curves for the three NaCl concentrations at pH 8.1 represents the point at which changes in the concentration of the background electrolyte have no effect on the net surface charge density (8). This value is consistent with the zero-point-of-charge measured for several synthetic aluminas (9–11). The apparent proton surface charge density was calculated assuming a surface site density of 1.7 sites per nm².

Preparation of Uranium Working Solution

A stock solution of ²³³U(VI) tracer was obtained from the inventory at the Lawrence Berkeley National Laboratory and purified by ion exchange. Analysis by α -spectroscopy indicated that the purified uranium contains 96.6% ²³³U and 3.3% ²³²U. A 1.1×10^{-4} M U(VI) working solution in 1 M NaCl at pH 3 was prepared from the stock solution and used for sorption experiments. The concentration of uranium was determined by liquid scintillation counting (LSC) using EcoLumeTM (MP Biomedicals Inc.) cocktail on a Wallac 1415 liquid scintillation counter.

Dissolution of Boehmite using HEDPA

Working suspensions spanning the pH range 4 to 11 were prepared by equilibrating boehmite with 1.0 M NaCl then using these suspensions to prepare samples for batch experiments. The batch experiments were performed in polypropylene centrifuge tubes shaken along their longitudinal axis on an orbital shaker at 150 rpm. Batch dissolution experiments to examine the dissolution of boehmite by HEDPA were conducted in suspensions with a constant boehmite concentration of 600 mg L⁻¹ (10 mM as Al) containing 5 mM, 10 mM, and 50 mM HEDPA. At specified intervals, aliquots were removed and passed through 200 nm nylon filters (Gelman Acrodisc). The aluminum concentration in the filtrate was measured using ICP-OES (Perkin Elmer, Optima 5300 DV). The pH of each suspension was adjusted towards an initial pH during sampling intervals using 0.1 and 0.01 M HCl and NaOH. At a few intervals, the HEDPA concentration in the filtrate was measured using a standard spectrophotometric method (12) employing a Cary 5G Spectrophotometer.

A series of potentiometric titrations were attempted to examine Al(III):HEDPA complex formation in 1 M NaCl. However, the titrations were unsuccessful as a white precipitate was observed across the pH range 4 to 9. To identify the precipitate, a series of solutions were prepared in 1 M NaCl with total Al(III):HEDPA ratios varying from

1:1 to 1:5. The precipitates were isolated via centrifugation and re-dissolved in ultra pure H₂O. The aluminum and HEDPA concentrations in the resulting solution were used to calculate the Al(III):HEDPA molar ratio in the solids. Additionally, to examine the effect of the background electrolyte and electrolyte concentrations, Al(III):HEPDA precipitates were formed in 0.1 and 1.0 M NaCl, KCl, NaNO₃, and (CH₃)₄NCl as well as ultra pure water. Aliquots of each solution were passed through filters with nominal pore sizes ranging from 450 nm to 12 nm to determine the particle size range of the precipitates.

Sorption of Uranium to Boehmite

The effect of HEDPA on uranium partitioning to synthetic boehmite was examined by amending equilibrated U(VI)-boehmite suspensions with HEDPA as described below. The uranium-boehmite suspensions were prepared by adding an aliquot of the U(VI) working solution to a boehmite suspension previously adjusted to a desired pH. The initial solution conditions were 6.1 μ M U(VI), 660 mg L⁻¹ boehmite, and 1.0 M NaCl. The samples were mixed at 150 rpm for 10 days. Preliminary kinetic tests (in the absence of HEDPA) indicated steady state uranium partitioning was achieved after 7 days. At specified intervals, the soluble uranium concentration was determined in the filtrate obtained by passing a subsample through a centrifugal filter (30k molecular-weight-cut-off, Nanosep, Pall Life Sciences, estimated 12 nm pore size). The uranium concentration was measured by LSC as described above.

After 10 days, half of the uranium-boehmite suspensions were amended with a small volume of a 0.05 M HEDPA stock solution (pH 7) to yield a suspension containing 5.4 mM HEDPA, 600 mg L⁻¹ boehmite, and 1 M NaCl. The other half of the suspensions were amended with a 0.5 M HEDPA solution (pH 7) to yield a 50 mM, 600 mg L⁻¹, 1 M NaCl suspension. The final U(VI) concentration was 5.8 μ M in all suspensions. Boehmite-free control solutions were also amended to 5.4 mM HEDPA as described above. The soluble uranium concentration was determined via filtration followed by LSC as described above.

RESULTS AND DISCUSSION

HEDPA Interactions with Boehmite: Sorption and Dissolution

In the absence of complexants, the aqueous phase concentration of aluminum is limited by the formation of sparsely soluble Al (hydr)oxide

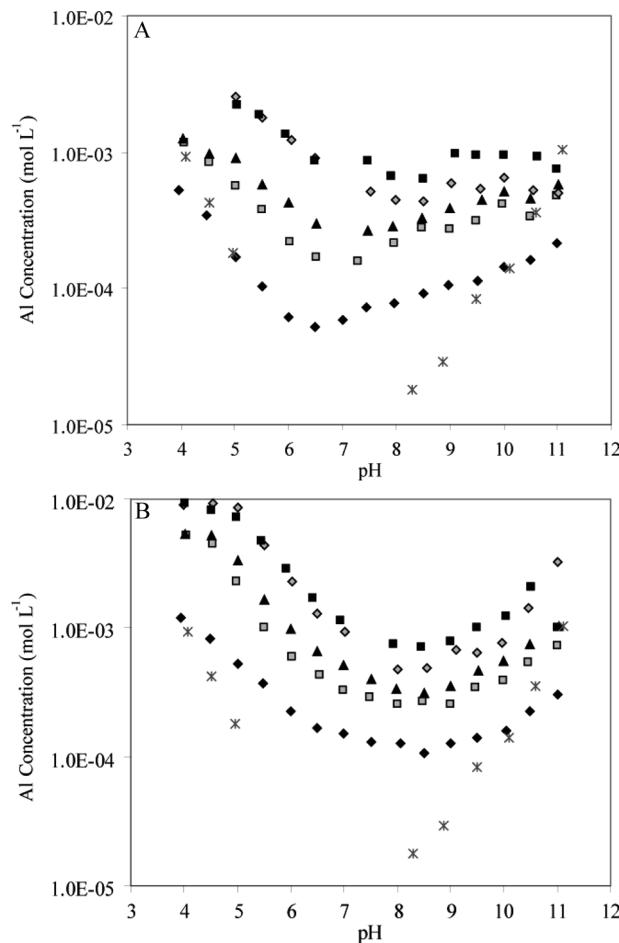


Figure 2. Effect of HEDPA on synthetic boehmite (γ -AlOOH) dissolution, $[\gamma\text{-AlOOH}] = 600 \text{ mg L}^{-1}$ (10 mM as Al^{3+}), $[\text{NaCl}] = 1.0 \text{ M}$. (A) $[\text{HEDPA}] = 5.4 \text{ mM}$; (B) $[\text{HEDPA}] = 50 \text{ mM}$. Symbols: (◆) 2 days, (□) 16 days, (▲) 36 days, (◇) 86 days, and (■) 135 days. (*) Control system contains no HEDPA and was measured after 135 days. Error bars, typically contained within area of symbol at 95% certainty, have been removed for clarity.

solids, especially at circum-neutral pH values. As shown in Fig. 2, addition of HEDPA clearly enhances the dissolution of boehmite. Across the pH range 5 to 9, samples amended with HEDPA have aluminum concentrations significantly higher than the HEDPA-free control system. The enhanced dissolution of boehmite in the presence of HEDPA is likely the result of formation of $\text{Al}_m\text{H}_h\text{L}_l$ complexes (where L represents

completely deprotonated HEDPA and m , h , and l are stoichiometric coefficients). The exact identity of specific complexes is uncertain at this point as the data available in the current literature are not in agreement (5,6) and our potentiometric experiments to obtain such data were unsuccessful due to the formation of a white precipitate across the neutral pH range (see Section 2.3). The nature and composition of the white precipitate was not fully determined, but analysis of the precipitate isolated from 1.0 M NaCl solutions with various Al(III):HEDPA molar ratios (from 1:1 to 1:2.5) indicated that the precipitate always had a 1:1 Al(III):HEDPA molar ratio. The observation of a 1:1 Al(III):HEDPA precipitate suggests that the low aluminum concentration across the neutral pH range (Fig. 2) may be controlled by the solubility of an Al(III):HEDPA solid phase. It was worth noting that the constitution and concentration of the background electrolyte significantly affected the physical characteristics of the precipitate. Precipitates prepared in 1.0 M NaCl and 1.0 M NaNO₃ tended to form clearly distinct particles while precipitates from 1.0 M (CH₃)₄NCl and 1.0 M KCl tended to have a gel-like character, suggesting that sodium may cause significant aggregation of the Al(III):HEDPA particles. In addition, the concentration of NaCl was also found to have a significant effect on the precipitate. Solutions containing aluminum and HEDPA in a 2:5 molar ratio in ultra pure H₂O, 0.1 M NaCl, or 1.0 M NaCl were passed through filters with pore sizes ranging from 450 nm to 12 nm. The fraction of soluble aluminum was found to be inversely related to the ionic strength of the solution.

The concentrations of aluminum in more acidic (pH 4 to 5) and basic (pH > 10) regions were higher than those in the neutral pH region (Fig. 2). After 2 days at pH 4 to 5, the concentrations of aluminum were comparable in systems with or without HEDPA. This is due to the formation of Al(OH)²⁺ and Al(OH)₂⁺ species yielding macromolar concentrations of aluminum in the absence of HEDPA. At pH > 10, the dominant aqueous species is Al(OH)₄⁻, yielding high aluminum concentrations in systems without HEDPA. At pH 11, the presence of HEDPA resulted in a lower aluminum concentration than that in the control system up to 16 days (Fig. 2). This difference may be due to precipitation of an Al(III):HEDPA solid as described above or slow kinetics of dissociation of Al(III):HEDPA complexes from the mineral surface. Data in the literature indicate that phosphate may form multi-nuclear surface complexes which could inhibit detachment of the metal-ligand complex from the surface or even inhibit dissolution (13). Therefore, as HEDPA surface complexation would be expected to occur through the phosphate group, the inhibition of boehmite dissolution may be due to formation of a multi-dentate surface complex.

Sorption of U(VI) to Synthetic Boehmite in the Absence of HEDPA

Data describing the sorption of uranium in the absence of HEDPA are shown in Fig. 3. In the absence of HEDPA, approximately 15% of the uranium was sorbed at pH 4. As the pH increased, the fraction of uranium sorbed increased until relatively steady partitioning was reached at pH > 5. Above pH 5, approximately 90% of the total uranium was sorbed. The sorption edge occurring between pH 4 and 5 has been previously observed by a number of researchers examining uranium sorption to aluminum (hydr)oxides (10,14–15). This behavior is consistent with electrostatic attraction/repulsion between uranium and the boehmite surface. As shown in Fig. 1, the boehmite surface charge transitions from a net positive to a net negative charge as the pH increases. This is due to the protonation and deprotonation of aluminol surface sites. At low pH values the positively charged uranyl dioxycation is repelled by the positively charged boehmite surface. As the pH was increased, both the net positive surface charge and the positive charge on the U(VI) species decreased (the latter due to hydrolysis), resulting in stronger sorption of U(VI) onto boehmite. The slight decrease in the fraction of uranium sorbed observed at higher pH values is presumed to be due to formation of soluble uranyl-carbonates. Although experiments were run in sealed centrifuge tubes, no effort was made to exclude carbonate from these experiments.

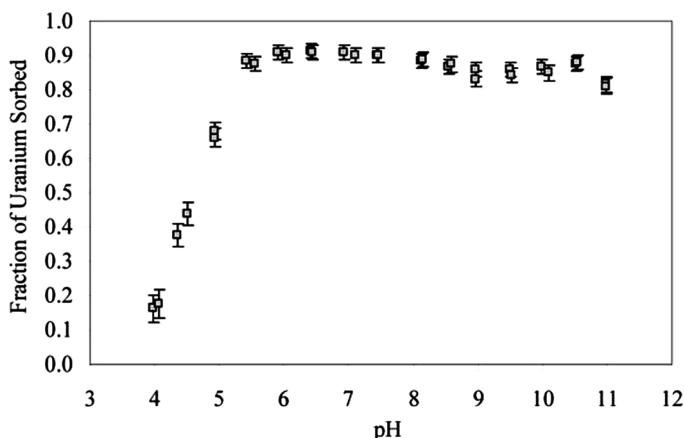


Figure 3. Fraction of uranium sorbed to boehmite versus pH after 10 day equilibration. System parameters: $[\gamma\text{-AlOOH}] = 0.66 \text{ g L}^{-1}$; $[\text{U(VI)}] = 6.1 \mu\text{M}$; $[\text{NaCl}] = 1.0 \text{ M}$.

Effect of HEDPA Concentration on U(VI) Sorption to Boehmite

Significant leaching of uranium from boehmite occurred rapidly following the amendment of the equilibrated uranium-boehmite suspensions to either 5 mM HEDPA or 50 mM HEDPA (Figs. 4a and 4b). At all pH values above 4.5, the addition of HEDPA resulted in rapid leaching of uranium from boehmite. Four hours after amendment of the uranyl-boehmite suspensions with 5 mM HEDPA, the fraction of uranium sorbed within the neutral pH region dropped from approximately 90% to approximately 50% (Fig. 4a). Over the next 181 days, the concentration of uranium in the aqueous phase continued to increase. This slow step could be due to a rate limitation of the dissociation of a U(VI):HEDPA complex from the boehmite surface or due to continued dissolution of the boehmite phase. A similar effect was observed for the 50 mM HEDPA system although a larger fraction of the total uranium was leached into the aqueous phase within the first 4 hours (Fig. 4b). Furthermore, the concentration of uranium in the aqueous phase after 181 days was significantly greater in the 50 mM HEDPA suspension relative to the 5 mM HEDPA suspension. The increased partitioning of uranium to the aqueous phase in the 50 mM HEDPA system is presumably due to enhanced dissolution of the boehmite solid phase, saturation of the remaining boehmite surface sites with HEDPA, and/or formation of U(VI):HEDPA complexes. As shown in Fig. 2, after 135 days, at least 10% of the aluminum in boehmite suspensions with 50 mM HEDPA is soluble at all pH values examined (Al concentration greater than 1.0×10^{-3} M). At pH values less than 5, the boehmite was almost completely dissolved. Therefore the total boehmite surface available for sorption of uranium or U(VI):HEDPA complexes was significantly diminished in the 50 mM HEDPA system.

Data in Fig. 4a (for the 5 mM HEDPA system) indicate a loss of uranium from the aqueous phase at low pH values following the addition of HEDPA. Such loss appears to be consistent with the precipitation of U(VI):HEDPA complexes that was observed at low pH and low HEDPA:U(VI) ratios (<2) by Reed et al. (4). However, precipitation of a U(VI):HEDPA complex in these sorption experiments is not expected, because the HEDPA concentration is between 3 and 4 orders of magnitude higher than the uranium concentration and uranium was found to remain in the aqueous phase in all control (with HEDPA but without boehmite) solutions spanning the pH range 4 to 11 for the duration of these experiments. Possible reasons for the observed loss of U(VI) in the low pH regions of the 5 mM HEDPA system are discussed in the following section.

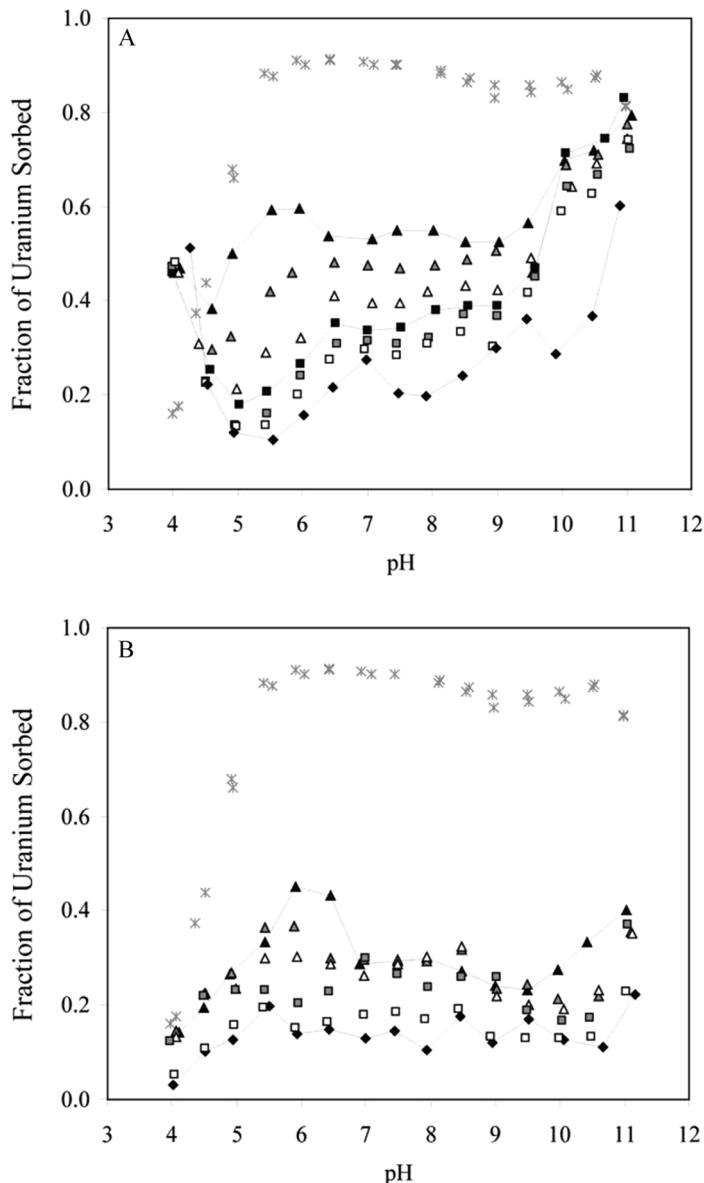


Figure 4. Effect of 5 mM (A) or 50 mM HEDPA (B) on Uranium Sorption to Boehmite: Symbols (▲) 0.2 days, (△) 1 day, (△) 6 days, (■) 14 days, (□) 30 days, (□) 59 days, and (◆) 180 days. For comparison, data from Fig. 3 showing steady state distribution (10 day equilibrium) of U(VI) without HEDPA present is shown (*). System parameters: $[\gamma\text{-AlOOH}] = 600 \text{ mg L}^{-1}$; $[\text{NaCl}] = 1.0 \text{ M}$; $[\text{U(VI)}] = 5.8 \mu\text{M}$. Error bars removed for clarity, average $2\sigma = 3\%$ propagated from counting statistics.

Effect of pH and HEDPA on U(VI) Sorption to Boehmite

Uranium Partitioning in the Acidic pH Region

Unless specified, the effects of pH on uranium partitioning will be discussed below in terms of the 5.4 mM HEDPA system (Fig. 4a). At pH 4, the addition of HEDPA increased the fraction of uranium sorbed from 15% to 47% within 4 hours (the fraction remained unchanged for the remainder of the experiment up to 181 days). This is the only sample in which the addition of HEDPA caused an increase in the sorption of uranium. The uranium partitioning at low pH observed in this system is similar to the observations of Cheng et al. (16) who found that phosphate caused increased uranium sorption to goethite-coated sands at low pH values, relative to phosphate free suspensions. A similar pH effect was also observed by Guo et al. (17) and Guo et al. (18) when examining thorium and uranium sorption in alumina suspensions amended with phosphate. Guo et al. (18) proposed that the enhanced sorption of uranium was due to formation of ternary surface complexes with U(VI) and phosphate.

Assuming that the behavior of the phosphate groups of HEDPA may be similar to that of phosphate, the increased uranium sorption observed in this work was also likely caused by the formation of a ternary U-HEDPA-AlOH surface complex. As the speciation plot of U(VI) in a 5.4 mM HEDPA solution shows, a number of anionic U(VI):HEDPA species exist in low pH regions (Fig. 5). On the

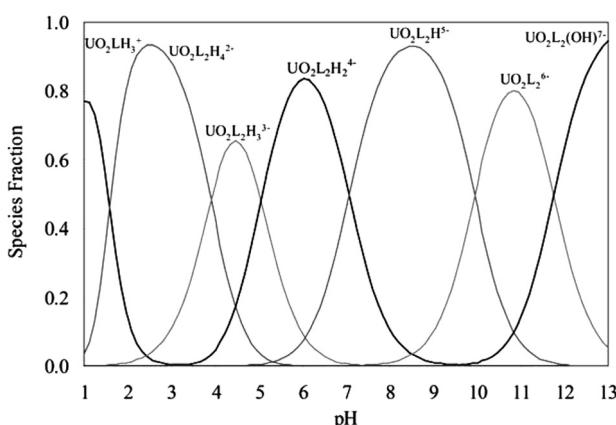


Figure 5. Speciation of U(VI) in 5.4 mM HEDPA and 1.0 M NaCl, calculated with the thermodynamic constants reported by Reed et al. (4). L stands for the fully deprotonated HEDPA anion.

other hand, the boehmite surface carries a net positive charge at low pH values. Therefore, the observed increase in sorption could be due to sorption of anionic species such as $\text{UO}_2\text{L}_2\text{H}_4^{2-}$ or $\text{UO}_2\text{L}_2\text{H}_3^{3-}$. Similar ternary surface complexes have been proposed to explain increased metal sorption at low pH values in the presence of anionic metal-ligand complexes (19–21). In this case, the sorption of the metal is proposed to be due to the formation of a ternary complex where the ligand bridges the mineral surface and the metal. Such geometry could be conceptualized utilizing the two phosphate groups of HEDPA. Within this pH region ($\text{pH} \sim 4$), HEDPA will be present as H_2L^{2-} (based upon the pK_a values reported by Reed et al. (4) and will be attracted to the positively charged boehmite surface. Nowack and Stone (22) examined HEDPA sorption to goethite and found that at low pH values, deprotonated anionic HEDPA species were attracted to the positively charged goethite surface. As the pH increased, and the surface developed a net negative charge, the sorption of HEDPA decreased (22). He et al. (23) observed similar pH effects when examining phosphate sorption to alumina.

Uranium Partitioning at Circumneutral pH

Across the pH region from 5 to 8, the fraction of sorbed uranium decreased significantly following the addition of HEDPA (Fig. 4a). After 1 and 6 days, the fraction of uranium sorbed increased as the pH was increased from approximately 5 to 6.5. Across the pH range 6.5 to 9, a relatively steady partitioning of uranium was observed for the sampling intervals at 0.2, 1, and 6 days. After 6 days, a more general trend appeared in the data where the fraction of uranium sorbed was found to increase as the pH increased from pH 5. It is noteworthy that coprecipitation of uranium with an Al(III):HEDPA complex as discussed above is also a possibility. However, the increase in uranium sorption with increasing pH is not consistent with coprecipitation as the primary mechanism for loss of uranium from the aqueous phase. Across this circumneutral pH region (5 to 9), the anionic $\text{U}(\text{VI})$:HEDPA species $\text{UO}_2\text{L}_2\text{H}_4^{2-}$, $\text{UO}_2\text{L}_2\text{H}_5^{3-}$, and $\text{UO}_2\text{L}_2\text{H}_6^{4-}$ are predominant (Fig. 5) and the boehmite surface maintains a net positive surface charge (Fig. 1). This will allow for an attractive force between the anionic $\text{U}(\text{VI})$:HEDPA complexes and the positively charged sites on the boehmite surface. Since both free HEDPA and free $\text{U}(\text{VI})$ are known to sorb within this pH region, a ternary surface complex could form through either uranium or HEDPA.

Uranium Partitioning above pH 9

Above pH 9, the fraction of sorbed uranium decreased significantly following the addition of HEDPA (Fig. 4a). There was a slight decrease in the fraction of uranium sorbed over the first 59 days followed by a significant decrease after 180 days. The rate at which uranium was leached into the aqueous phase was similar to the rate of boehmite dissolution (Fig. 2), indicating that dissolution was a primary factor controlling uranium partitioning within this pH region. At earlier time intervals, the fraction of uranium sorbed across the pH region 9 to 11 was higher relative to the circumneutral pH region. This difference is proposed to be due to a change in the partitioning of HEDPA within this pH region. Nowack and Stone (22) observed a significant decrease in the fraction of HEDPA sorbed to goethite above pH 9. This is consistent with the full deprotonation of HEDPA in 1.0 M NaCl ($pK_{a,4} = 9.5$, measured in this work, data not shown). Therefore, boehmite sorption sites which are occupied by HEDPA at neutral and acidic pH regions may be available for sorption of uranyl either as a uranyl hydroxide or as a U(VI):HEDPA complex. In the absence of a complexing ligand, uranium strongly sorbs to aluminum (oxy)hydroxides at high pH values (Fig. 3 and (10,14,15)). Therefore the formation of a U(VI):hydroxide or U(VI):HEDPA (whereby surface complexation would occur through uranium rather than through the phosphate groups of HEDPA) surface complex is feasible. However, verification of such complexes through spectroscopy would be desirable, if not necessary, prior to the incorporation of such species into modeling efforts.

CONCLUSIONS

Results from this work show that HEDPA is capable of significantly enhancing the solubility of aluminum hydroxides. The degree of enhancement is dependent upon the bulk solution pH and the concentration of HEDPA. HEDPA is capable of leaching uranium from synthetic boehmite through solid phase dissolution and/or the formation of U(VI):HEDPA complexes. Across the pH range examined, the speciation of U(VI):HEDPA complexes affects the partitioning of uranium between the solid and aqueous phase. Partitioning of uranium was observed to vary with pH and correlated with the partitioning of HEDPA and HEDPA-promoted boehmite dissolution. The sorption of uranium to boehmite in the presence of HEDPA is proposed to be through ternary U(VI):HEDPA surface complexes. However, no spectroscopic evidence is provided to indicate formation of such complexes. These results

indicate that HEDPA could be used to reduce the volume of the aluminum component of sludge within the Hanford waste tanks. However, at the circumneutral pH values examined in this work, the aluminum concentrations achieved during HEDPA leaching are several orders of magnitude too low to be of practical use. Furthermore, because a significant fraction of uranium (and presumably other actinides) could also be leached from boehmite by the addition of HEDPA, careful consideration of the partitioning of the actinides must be made if a strong complexant such as HEDPA is selected for sludge washing.

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