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Interaction of Uranium(VI) with Phthalic Acid

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Phthalic acid, a ubiquitous organic compound found in soil, water, and in domestic and nuclear wastes can affect the mobility and bioavailability of metals and radionuclides. We examined the complexation of uranium with phthalic acid by potentiometric titration, electrospray ionization-mass spectroscopy (ESI-MS), and extended X-ray absorption fine structure (EXAFS) analysis. Potentiometric titration of a 1:1 U/phthalic acid indicated uranyl ion bonding with both carboxylate groups of phthalic acid; above pH 5 the uranyl ion underwent hydrolysis with one hydroxyl group coordinated to the inner-sphere of uranium. In the presence of excess phthalic acid, ESI-MS analysis revealed the formation of both 1:1 and 1:2 U/phthalic acid complexes. EXAFS studies confirmed the mononuclear biligand 1:2 U/phthalic acid complex as the predominant form. These results show that phthalates can form soluble stable complexes with uranium and may affect its mobility.

Introduction

Phthalic acid, a dicarboxylic, aromatic compound, is widely used in the manufacture of building materials, home furnishings, plastics, transportation components, clothing, food, personal care and medical products. It is also a structural component of natural organic matter such as humic acids and fulvic acids. Plastics constitute a significant component of the transuranic (TRU) and low-level radioactive wastes, and phthalates have been found in leachates from low-level radioactive waste sites.[1,2] Degradation of plastics results in release of phthalic acid and its esters into the environment.[3] These degradation products have been linked to alteration of human male reproductive development.[4] Phthalic acid has been shown to form complexes with Ni, Co, Cu, and Zn with a variety of coordination modes ranging from monodentate to heptadentate.[5] Nuclear magnetic resonance studies revealed that beryllium forms a mononuclear biligand complex with phthalic acid, while UV−vis spectroscopic measurements have shown that vanadyl and uranyl ions form similar mononuclear and binuclear biligand complexes at pH values above 4.5.[6,7] Zinc forms an unusual one-dimensional single helix structure with phthalic acid.[8] The interactions of phthalates with mineral surfaces such as goethite, tectosilicates, quartz, albite, and various types of clays and complexation with metals has also been reported.[9−11] When phthalic acid is adsorbed onto the surface of ferrihydrite in the presence of Cu and Cd a ternary complex is formed.[12]

Although the presence of phthalic acid in the environment has been widely observed, its ability to form complexes with radionuclides has not been fully evaluated. Uranium contamination of the environment because of mining and milling operations and disposal of nuclear weapons waste along with contaminated plastic items such as gloves and containers has become a major environmental concern. The phthalic acid present in these wastes may increase the solubility of uranium in the environment by formation of uranium-phthalate

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complexes.[13−16] However there is little information available on the molecular association of phthalic acid with uranium.[7,17−19] In this study we investigated the interaction of uranyl ion with phthalic acid in solution and determined the molecular structures of the predominant U-phthalic acid complexes formed.

Experimental Section

Preparation of U/Phthalic Acid Complex. Uranyl nitrate and phthalic acid stock solutions were prepared by dissolving $UO_2(NO_3)_2 \cdot 6H_2O$ (Analar, BDH Chemicals Ltd., Poole England) or anhydrous o-phthalic acid (ACS reagent grade, Aldrich, Milwaukee, WI) in deionized water. Uranium-phthalate mixtures were prepared by slowly combining 1:1 (equimolar), 1:5 and 1:10 U/phthalic acid mixtures while slowly adjusting the pH to 5.5 with sodium hydroxide. The solution was then allowed to equilibrate in the dark for 24 h at 26 ± 1 °C prior to use after which the pH was readjusted to 5.5.

Potentiometric Titration. We determined the hydrogen ion concentration in 0.50 mM solutions of (i) phthalic acid, (ii) $UO_2(NO_3)_2$, and (iii) 1:1 U/phthalic acid complex by potentiometric titration. The change in pH because of the incremental addition of 0.01 M NaOH was obtained on duplicate samples using a Mettler-Toledo DL 53 Titrator with a DG 111 SC electrode. The ionic strength of the solutions was adjusted to 0.1 M with KCl.

Solubility of U in the Presence of Phthalic Acid. The solubility of uranium added as uranyl nitrate in the presence of 1:1, 1:5, and 1:10 U/phthalic acid was determined by addition of uranyl nitrate to a solution containing the appropriate concentration of phthalic acid as described above. The pH was adjusted to 5.5, and the mixture was allowed to equilibrate in the dark for 24 h at 26 ± 1 °C on a rotary shaker at 125 rpm. An aliquot was withdrawn, and the solution was filtered through a 0.45 μm Durapore (Millipore, MA) filter.

The concentration of uranium was determined using a kinetic phosphorescence analyzer KPA-11 (Chemchek, WA). Uraplex (1.5 mL) was added to the appropriately diluted sample to complex the uranyl ion and reduce interferences due to quenching. The concentration of uranium in the samples was then determined by measurement of the phosphorescence intensity due to excitation by a nitrogen laser at 337 nm. This excites the dye laser (stilbene-420) to produce an excitation wavelength of 420 nm. A 515 nm band-pass filter was used to filter the emission signal and pass the 515 nm uranium peak.

Electrospray Ionization-Mass Spectrometry (ESI-MS) Analysis. The molecular weight of the U/phthalic acid complex formed at pH 5.5 in the presence of excess phthalic acid (1:5 U/phthalic acid) was determined by ESI-MS. An aliquot of the complex was introduced into an LCQ Advantage ion trap mass spectrometer (ThermoFinnigan, San Jose, CA) system at a rate of 10 μL min^{-1} using a microsyringe pump. Operating parameters for the ESI including gas flow rate, operating voltage, and current were optimized using an equimolar 10^{-3} M Cu(II)-citrate complex.[6] The capillary temperature and spray voltage were set at 230 °C and 3.42 kV, respectively. Spectra for the molecular ion peaks were obtained in both the positive and negative modes. Data acquisition for both the full scan mass spectra and MS/MS fragmentation were obtained using the Xcalibur software. Full scan mass spectra were collected in the *m*/*z* range of 50 to 2000 and were averaged for 1 min. Fragments resulting from the MS/MS spectra scans were similarly obtained using a collision energy of 30% with nitrogen gas.

X-ray Absorption Spectroscopy (XAS). The 1:5 U:phthalate complex at pH 5.5 was freeze-dried using a Labconco Freeze-Dry System/Freezone 4.5 (Kansas City, MO). It was then ground to a fine powder and mixed with boron nitride. The uranyl acetate was obtained from Atomergic Chemicals (Farmingdale, NY). About 5 mg of the powder was mounted on an Al sample holder having a cut-out geometry of 2 mm (H) × 20 mm (L) × 1.5 mm thickness, and the holder was sealed with Kapton tape. The sample was analyzed at the U L_{III} edge (17.166 keV) on the X-10C beam line at the National Synchrotron Light Source (NSLS). The fluorescence spectrum was collected using a 13 element Ge detector. Information on the type and number of atoms surrounding the absorbing atom was determined by extended X-ray absorption fine structure (EXAFS) analysis.[20−23] The theoretical EXAFS modeling code FEFF6 was used to calculate the back-scattering phase and amplitude information for individual neighboring atoms. Four scans were collected per sample, and the data were averaged. The amplitude reduction factor (S_0^2) was fixed at 1.0 for all of the fits. Background was subtracted from the spectra, and fitting of the data was performed in *R* space by taking the Fourier transform of the $k^3X(k)$ (2.0 to 12.0 Å^{-1}) data. The $U \rightarrow O \rightarrow U \rightarrow O \rightarrow U$ multiple scattering path was taken into consideration for the fitting procedure.[24,25]

Results

Potentiometric Titration. Figure 1 shows the number of hydrogens released by potentiometric titration from phthalic acid, $UO_2(NO_3)_2$, and 1:1 U/phthalic acid complex using 0.01 mM NaOH. Phthalic acid has two titratable hydrogen ions which are removed from the carboxylic acid groups in overlapping steps. A sharp inflection point around 2.0 mM OH^-/mM phthalic acid (pH 7.5) confirmed that the carboxylate groups were completely neutralized. Titration of uranyl nitrate shows the presence of two inflection points, a broad inflection point at 1.2 mM OH^-/mM U at pH 4 and a sharper one at 2.2 mM OH^-/mM U at pH 7.5. These indicate a stepwise hydrolysis of the uranium with formation of $UO_2(OH)_2$. The titration of the 1:1 U/phthalic acid complex also shows two inflection points. The first one is a broad inflection point extending from 1.5 to 2.5 mM OH^-/mM U at pH 5 to 6 corresponding to dissociation of the two carboxylic acid hydrogens of the organic ligand as a result

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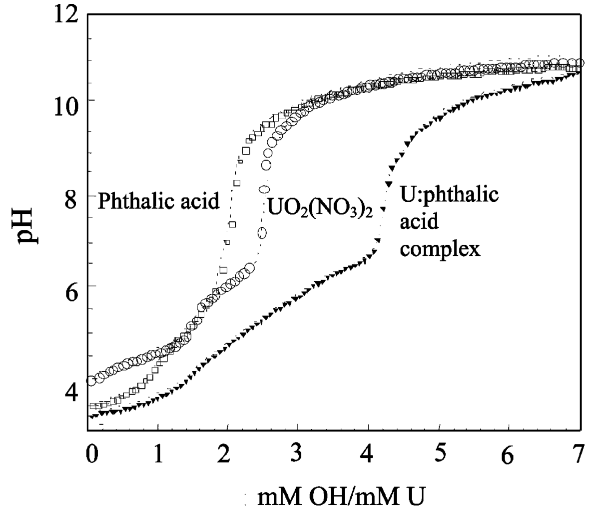

Figure 1. Potentiometric titration curves for 0.5 mM phthalic acid, 0.5 mM $UO_2(NO_3)_2$, and 0.5 mM 1:1 U/phthalic acid complex.

Table 1. Solubility of 0.5 mM Uranyl Nitrate in the Presence of Phthalic Acid at pH 5.5 after 24 hours[a]

complex	U in filtrate (mM)
uranyl nitrate	0.24 ± 0.01
1:1 U/phthalic acid	0.41 ± 0.01
1:5 U/phthalic acid	$0.49 + 0.01$
1:10 U/phthalic acid	0.48 ± 0.02

[a] ±1SEM.

of complex formation with uranium, as well as some hydrolysis of the uranium. The second inflection point at 4.2 mM OH^-/mM U at pH 7.5 is a result of uranium hydrolysis.

Solubility of U in the Presence of Phthalic Acid. Table 1 shows the relative solubility of 0.5 mM U in a solution containing various concentrations of phthalic acid at pH 5.5 after 24 hours. In the absence of phthalic acid 48% of the uranium was in solution. In the presence of 1:1 U/phthalic acid about 82% of the uranium remained in solution; in the presence of 1:5 and 1:10 U/phthalic acid >96% of the uranium remained in solution. These results indicate that excess phthalic acid is necessary to maintain the uranium in solution as complexed form.

ESI-MS Analysis. The spectra showing the molecular weight of the 1:5 U/phthalic acid complex in solution is presented in Figure 2. There are three main *m/z* peaks 165.0, 452.2, and 599.0 detected in the negative ion spectra. These values are usually accurate to within one mass unit. The peak at 165.0 *m/z* is due to the presence of the uncomplexed singly charged phthalic acid molecule $[C_8H_5O_4]^-$. The peak at *m/z* 452.2 corresponds to an equimolar 1:1 U/phthalic acid complex with one hydroxyl group coordinating with the uranyl ion $[(UO_2)C_8O_4H_4(OH)]^-$. The peak at *m/z* 599.0 is due to the presence of two phthalic acid molecules coordinating to the uranyl ion due to formation of a 1:2 U/phthalic acid complex $[(UO_2)(C_8O_4H_4)_2(H)]^-$. ESI-MS spectra for the 1:10 U/phthalic acid complex gave similar spectral information.

Figure 3 shows the ESI-MS spectrum for the 1:5 U/phthalic acid complex obtained in the MS/MS mode. Specific ion fragmentation during MS/MS analysis gives the ability

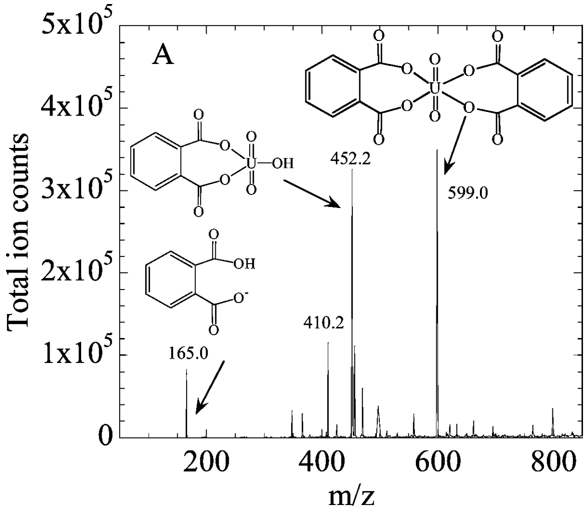

Figure 2. ESI-MS spectrum obtained in the negative mode for 1:5 U/phthalic acid complex.

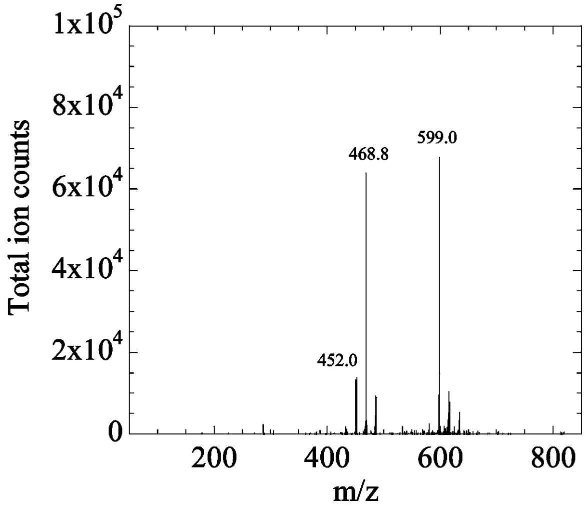

Figure 3. ESI-MS/MS spectrum obtained in the negative mode for *m/z* 599 peak from 1:5 U/phthalic acid complex. Collision energy was 30%.

to identify distinctive structural features that help to determine the composition of the molecule. Therefore, to establish the presence of two phthalates in the *m/z* 599 complex we subjected the molecule to bombardment with N_2 gas at a collision energy of 30%. The results in Figure 3 indicate that the primary mode of fragmentation is the removal of one phthalic acid molecule from the biligand complex resulting in formation of the *m/z* 452 peak $[(UO_2)C_8O_4H_4(OH)]^-$ and a primary fragment peak at *m/z* 468.8 due to the formation of the monoligand $[(UO_2)C_8O_4H_5(OH)_2]^-$ or $[(UO_2)C_8O_4H_4(H_2O)(OH)]^-$ complex species. These results confirm the biligand nature of the complex as well as the presence of associated hydroxyl groups with the uranyl ion.

X-ray Absorption Spectroscopy. EXAFS analysis was performed to determine the molecular structure of 1:5 U/phthalic acid complex. Figure 4A,B shows the k^3-weighted EXAFS spectra (2.0 to 12.5 $Å^{-1}$) and Fourier-transformed spectra of uranyl nitrate and uranyl acetate standards and the 1:5 U/phthalic acid complex. The fitting parameters are shown in Table 2. The EXAFS fitting for uranyl nitrate

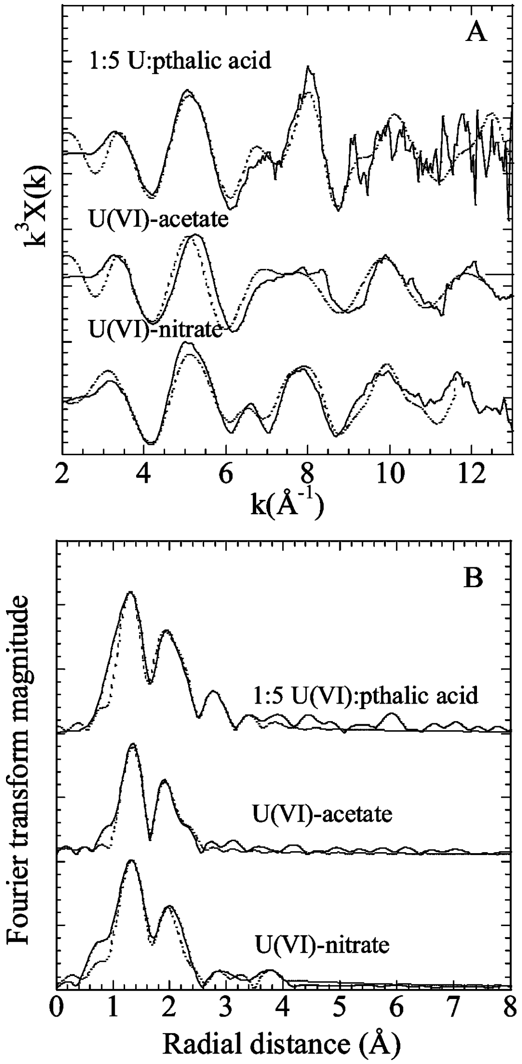

Figure 4. Uranium L$_{III}$ k^3-weighted EXAFS spectra (2.0−12.5 Å^{-1}) (A) and corresponding Fourier transform EXAFS spectra (B) for uranyl nitrate, uranyl acetate, and 1:5 U/phthalic acid complexes. Experimental data (solid line); theoretical fit (dashed line).

Table 2. EXAFS Fitting Parameters for Uranyl Nitrate, Uranyl Acetate, and 1:5 U/phthalic Acid Complex[a]

sample	atom pair	CN	R (Å)	σ^2 ($\times 10^{-3}$ Å^{-2})	ΔE_0 (eV)
$UO_2(NO_3)_2$	U−O$_{ax}$	2.0	1.77 ± 0.02	2.0 ± 0.1	8.4 ± 1.0
	U−O$_{eq}$	5.1 ± 0.6	2.47 ± 0.03	1.1 ± 0.3	7.4 ± 2.9
$UO_2(CH_3COO)_2$	U−O$_{ax}$	2.0	1.76 ± 0.01	3.0 ± 1.0	7.9 ± 1.7
	U−O$_{eq}$	6.0 ± 1.0	2.35 ± 0.02	8.1 ± 3.3	1.6 ± 0.7
	U−C	2.0 ± 1.0	2.91 ± 0.02	3.5 ± 2.1	1.6 ± 0.3
1:5 U/phthalic acid complex	U−O$_{ax}$	2.0	1.73 ± 0.01	1.3 ± 0.2	5.0 ± 1.3
	U−O$_{eq}$	6.1 ± 1.0	2.48 ± 0.03	8.6 ± 0.2	14.0 ± 5.6
	U−C$_1$	4.3 ± 2.1	2.92 ± 0.04	2.7 ± 0.4	3.5 ± 1.0
	U−O	3.0 ± 1.8	3.27 ± 0.01	3.1 ± 0.6	8.2 ± 1.0
	U−C$_2$	4.1 ± 0.5	4.31 ± 0.02	7.6 ± 0.1	6.7 ± 2.0

[a] CN, number of atoms in each coordination shell; R, radial distance (Å) between two atoms of each atomic pair; σ^2, relative mean square displacement from the average path length (Debye−Waller factor); E_0, photoelectron energy shift. The CN for U−O$_{ax}$ was set at 2.0 in all fits.

indicates that the structure consists of 2.0 axial O's at 1.77 ± 0.02 Å which is typical for uranium in the hexavalent state, and an equatorial O shell with 5.1 ± 0.6 O's at 2.47 ± 0.03 Å. The uranium in the uranyl acetate standard is also present in the hexavalent state and has 2.0 axial O's at 1.76 ± 0.01 Å, 6.0 ± 1.0 U−O$_{eq}$ interactions at 2.35 ± 0.02 Å and a third shell with 2.0 ± 1.0 U−C interaction at 2.91 ± 0.02 Å indicating the presence of carboxylate bonding to the uranium. The U/phthalic acid complex structure consists of 2.0 axial O's at 1.73 ± 0.01 Å, and an equatorial O shell with 6.1 ± 1.0 O's at 2.48 ± 0.03 Å. There are also two U−C interactions, the first one appears at 2.92 ± 0.04 Å corresponding to the presence of 4.3 ± 2.1 C's carboxylate groups indicating there are four carboxylate groups bonding to the central U atom. The second interaction at 4.31 ± 0.02 Å has 4.1 ± 0.5 C's which are associated with the aromatic group of the phthalic acid. Finally a U−O interaction is present with 3.0 ± 1.8 O's at 3.27 ± 0.01 Å which corresponds to the double bonded O atoms of the carboxylate group. No U−U interaction was observed. These results are supported by the ESI-MS analysis and confirm the presence of a biligand complex with two phthalic acids surrounding the U atom. The proposed structures for the 1:1 and 1:2 U/phthalic acid complexes are shown in Figure 5A,B, respectively.

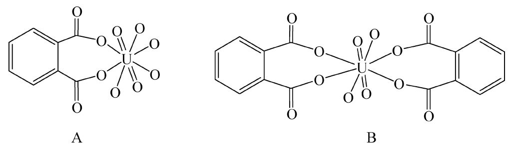

Figure 5. Proposed structures for the 1:1 (A) and the 1:2 U/phthalic acid (B) complexes. Atoms: carbon, black; oxygen, gray.

Discussion

Phthalic acid has been shown to form novel chelate structures with metals and actinides. Potentiometric titration of U/phthalic acid complex suggested the involvement of the carboxylic acid groups of the phthalic acid in the complexation of uranium. In addition, the displacement of the titration curve above pH 5 compared to the uranyl ion indicated that uranium underwent hydrolysis reaction. Potentiometric and spectrophotometric methods were used by Goncalves et al. [7] and Rajan and Martell[17] to establish the presence of a hydrolyzed complex species of uranium. The formation of a uranium bidentate complex involving the two carboxylates of phthalic acid by potentiometric titration has been observed.[17,28] The stability constant (log K) for the equimolar U-phthalate complex was calculated to be 4.38 and 4.81 in 1 M KNO_3 and in 0.1 M $NaClO_4$, respectively. This is greater than that obtained for succinic acid (log K = 3.68), another dicarboxylic acid but is similar to maleic acid (log K = 4.46) which has a rigid structure because of the presence of a double bonded carbon backbone. The relative greater stability of the phthalic acid complex compared to linear dicarboxylic acids such as succinate has been attributed to the cis arrangement of the carboxylate groups maintained by the aromatic ring.[17]

Analysis of the U/phthalic acid complex by ESI-MS showed two types of complexes are formed, one with a hydrolyzed equimolar 1:1 U/phthalic acid and a second mononuclear biligand 1:2 U/phthalic acid complex. In the presence of excess phthalic acid the latter is presumed to be the predominant species in solution because it has the highest peak intensities obtained in the ESI-MS spectrum. The MS/MS analysis confirmed the presence of two phthalic acids associated with uranyl ion in the complex. EXAFS analysis showed the organic ligand is linked to uranium through the carboxylate O's confirming the formation of the 1:2 U/phthalic acid complex.

Phthalic acid has been shown to form a variety of structures (mononuclear, multinuclear, polymeric) with metals under differing conditions of pH and concentration.[5,17] X-ray crystallography studies have shown that Zn forms a helical structure by coordination to two phthalic acid molecules through alternating monodentate and bidentate bonding of the carboxylate groups. One water molecule was involved in the coordination plane.[7] Three unique polymers of phthalic acid and N-donor ligands like pyridine and imidazole with Cu(II), Zn(II), and Co(II) have been identified by X-ray crystallography.[5] The complexes contained extended hydrogen bonding networks which stabilized the polymeric network. Double salts of phthalic acid with U(VI) and NH_4^+, K^+, and Cs^+ were prepared which showed the presence of a tetrameric anion $[(UO_2)_4O_2(C_6H_4C_2O_4)_4]^{4-}$ with the carboxylate groups of phthalic acid coordinated to uranium in both monodentate and bidentate fashion.[19] X-ray diffraction studies have shown that Np(VI) formed a monoligand complex which was similar in structure to the U(VI)-phthalate complex.[26] Double salts of Np(VI) and Pu(VI) of the type $M_2AnO_2(C_8H_4O_4)_2 \cdot nH_2O$ isolated from neutral solutions in the presence of excess phthalate formed similar biphthalate structures, while U(VI) formed a complex anion of the form $[(UO_2)_4O_2L_4]^{4-}$.[27] The inability to synthesize a uranyl salt with a complex ion was attributed to its increased tendency to hydrolyze in comparison with Np(VI) and Pu(VI).

In this study we have shown that uranyl ion forms mono- and biligand complexes with phthalic acid in aqueous solution. However, based upon the ability of phthalic acid to form a number of different complexes, the presence of minor concentrations for other U-phthalate coordination modes cannot be ruled out. The results of this work suggest that in a repository environment containing a high concentration of plastics, the mobility of U(VI) may be enhanced by formation of soluble U-phthalate complexes. Thus, any mitigation strategy for uranium should take into account the presence of such complexing organic compounds.

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