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Actinide Immobilization in the Subsurface Environment by In-situ Treatment with a  
Hydrolytically Unstable Organophosphorus Complexant:

Uranyl Uptake by Calcium Phytate

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## ABSTRACT

In addition to naturally occurring uranium and thorium, actinide ions exist in the subsurface environment as a result of accidental releases and intentional disposal practices associated with nuclear weapons production. These species present a significant challenge to cost-effective remediation of contaminated environments. An attractive approach to decreasing the probability of actinide migration in the subsurface is to transform the ions into a less mobile form by remote treatment. We have under development a process which relies on a polyfunctional organophosphorus complexant to sequester the mobile metal ions by complexation/cation exchange in the near term, and to subsequently decompose, transforming the actinides into insoluble phosphate mineral forms in the long term. Studies to date include identification of a suitable organophosphorus reagent, profiling of its decomposition kinetics, verification of the formation of phosphate mineral phases upon decomposition of the reagent, and extensive comparison of the actinide uptake ability of the calcium salt of the reagent as compared with hydroxyapatite. In this report, we briefly describe the process with focus on the cation exchange behavior of the calcium salt of the organophosphorus sequestrant.

### Introduction

Fifty years of actinide production at U. S. Department of Energy facilities (and corresponding facilities in the former Soviet Union and elsewhere) have left a substantial cleanup obligation in the post-cold war era. Accidental releases, disposal of contaminated materials in landfills and soil pits, and leaking storage tanks in the subsurface have contaminated soils and waters at many DOE sites with a variety of radioactive and chemically hazardous substances [1]. For those materials possessing appreciable water solubility, transport as soluble or colloidal species in ground or surface waters represents a potential pathway for spread of the contamination to the general environment. Among the long-lived radioactive materials, the transuranic actinides are commonly acknowledged to represent the greatest hazard, though their mobility is strongly a function of conditions. Immobilization of actinide ions is the particular focus of this report.

Various technological solutions have been proposed for remediation of sites contaminated with transuranic elements. Protocols have been developed for washing of soils samples with various solvent/chelating agent mixtures. Radionuclides can be removed from groundwaters by pumping the waters to the surface, treating them (e.g., by cation exchange or membrane separations) and returning the cleaned water to the subsurface, so-called "pump and treat" methods. Installation of reactive barriers into the subsurface, injection of impermeable barriers, and even "glassification", melting the soils to convert them to glass, have been suggested as methods for minimizing the potential for migration of the contaminants to the wider environment. These treatment options achieve varying levels of success in cleaning the subsurface soils. Many suffer defects/obstacles that inhibit their deployment. Several involve movement of contaminated materials from soils, creating opportunities for increased dispersal of the radionuclides during the treatment process. Other methods are so expensive as to be impractical for implementation on a large scale. Some have proven unacceptable because they performed better in the laboratory than they did in the field.

An attractive alternative approach is to develop in-situ treatment methods that do not require extensive movement and treatment of the contaminated geomeia (soils or waters) but rather immobilize (or otherwise transform) the radioactive elements in place. Introduction of reactive or impermeable barriers into the soil are attractive from this perspective, as the need to excavate soils or pump waters is diminished. The ideal in situ treatment option would create a reactive barrier through introduction of water soluble species which would react in the soil (perhaps with soil components) to form the barrier without significant intervention from the surface. Natural processes of this type have been suggested as possible mechanisms for the formation of rich mineral veins of redox active metal ions, for example, at the Oklo mine site in Gabon [2].

The premise of our work is to immobilize the actinides in situ, transforming them into thermodynamically stable mineral phases by introducing a mineralizing agent into the subsurface environment via an aqueous solution. The mineralizing reagent is injected (or sprayed on the surface and allowed to percolate downward) as a water soluble organic compound which reacts with

naturally occurring metal ions to form insoluble compounds. These compounds readily exchange the naturally occurring metal ions for the mobile actinide cations which are thus removed from the mobile aqueous phase via a cation exchange mechanism. The complexant is designed to subsequently decompose, releasing the mineralizing agent and promoting the formation of thermodynamically stable actinide-bearing mineral phases.

As thermodynamic calculations suggest phosphate as an optimum mineral phase for actinide sequestration, our studies have focused on the natural product phytic acid (myo-inositolhexakisphosphoric acid, Figure 1) as the means of delivery of phosphate to the subsurface environment. Phytic acid is a natural product (isolated from beans and leafy vegetables) which is used commercially as a starting material in the manufacture of inositol [3], as a dietary supplement, a nutrient source for microorganisms,

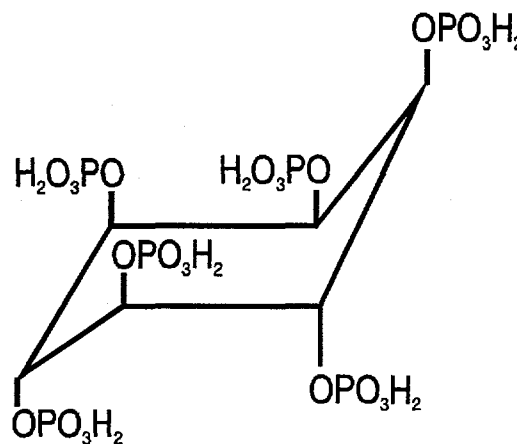


Figure 1. Phytic acid.

and a metal chelating agent and precipitating agent. It complexes polyvalent metal ions moderately and is readily hydrolyzed to release phosphate. In the U. S., phytic acid is produced at an annual rate of several hundred thousand metric tons as a byproduct of fermentation processes [4], but this material presently is not isolated for use being recycled along with the other solid fermentation residues for animal feed. Because the calcium salts of phytic acid are insoluble, we projected that these salts would function as cation exchangers for soluble actinides in the environment.

Our earlier investigations have established that phytic acid is hydrolyzed in pH 5-7 solutions of synthetic groundwaters with a half-life of about 100-150 years at 25°C [5]. Literature reports indicate that microbes can significantly promote the rate of hydrolysis of phytate, often by orders of magnitude [6]. We have also established that the hydrolysis of  $\text{Eu}^{3+}$  (as an analog for trivalent actinides) and  $\text{UO}_2^{2+}$  salts of phytic acid yield the corresponding metal phosphate solids  $\text{EuPO}_4 \cdot \frac{1}{2}$

H<sub>2</sub>O and (UO<sub>2</sub>)<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub>•H<sub>2</sub>O. Radiotracer distribution experiments established that the concentration of Eu<sup>3+</sup> was controlled in the 10<sup>-10</sup> M concentration range over the pH range of 5-8 by 10<sup>-3</sup> M phosphate. Parallel experiments with <sup>233</sup>UO<sub>2</sub><sup>2+</sup> tracer indicated less efficacious control of uranium solubility by phosphate with concentrations in solution around 10<sup>-7</sup> M at pH 6-7, and with slightly higher levels at pH 8 due to carbonate complexation. Phosphate alone appears to have little impact on the solubility of NpO<sub>2</sub><sup>+</sup> except at pH above 7 and [PO<sub>4</sub><sup>3-</sup>]<sub>t</sub> = 0.01 M. However, addition of low concentrations of Ca<sup>2+</sup> appear to reduce actinide solubility generally, presumably due to the formation of hydroxyapatite (Ca<sub>5</sub>(OH)(PO<sub>4</sub>)<sub>3</sub>•xH<sub>2</sub>O) which either coprecipitates or sorbs the actinides from the solution phase.

In the following discussion, we will present new information on the preparation, stoichiometry, and cation exchange behavior of calcium phytate with particular focus on its ability to complex/sorb uranyl.

### Experimental

Phytic acid was purchased from Aldrich Chemical Company as the dodecasodium salt and used as received. The material was characterized by NMR spectroscopy and potentiometric titration. The phytic acid concentration was determined by potentiometric titrations of sodium phytate solutions with 1 M HClO<sub>4</sub>, quantifying the total phytate using the difference of the equivalence points at 6 and 8 equivalents of acid added, where clear breaks are observed in the titration curve [7]. All other reagents were of analytical reagent grade and used as received.

Stoichiometry of solid calcium phytate was determined as a prerequisite for actinide cation exchange studies according to the following procedure. A solution of sodium phytate (0.049 M) was adjusted to pH 7 with NaOH. A 0.994 M Ca(NO<sub>3</sub>)<sub>2</sub> solution was added to the sodium phytate solution at metal to ligand ratios of 1:1, 2:1, 3:1, 4:1, 5:1, and 6:1. A white precipitate formed immediately under all conditions. The solutions were stirred overnight then washed four times with deionized water. After the fourth washing the samples were placed in an oven and dried at 100 °C for 24 hours. After drying, the precipitates were analyzed for Ca, P, Na, C, and H.

The second phase of the cation exchange experiments was designed to test the relative affinity of phytate for  $\text{Ca}^{2+}$  and  $\text{UO}_2^{2+}$ . Solutions of calcium nitrate (0.9945 M) and uranyl nitrate (0.1123 M) were added to the vials in mole ratios of 5:1, 4:2, 3:3, 2:4, 1:5, and 0:6 respectively. A solution of sodium phytate (0.049 M) adjusted to pH 7 was then added to the  $\text{Ca}^{2+}/\text{UO}_2^{2+}$  solutions so that the ratio of the total metal concentration to ligand was 6:1. The vials were placed on a stir plate and mixed continuously for 72 hours, centrifuged, the solids washed with deionized water, dried in an oven at 110-125 °C, then analyzed for Ca, U, P, and Na.

The final phase of the cation exchange experiments was to test uranyl substitution for calcium on solid calcium phytate samples. A known amount of calcium phytate (FW=957 g/mole) of known stoichiometry ( $\text{Ca}_{4.6}\text{Na}_{0.20}\text{H}_{2.6}(\text{CH}(\text{PO}_4))_6 \cdot 6.5\text{H}_2\text{O}$ , see below) was added to six 40 ml test tubes of a predetermined weight. Ten ml of deionized water was added to the vials. A solution of uranyl nitrate was added to the mixture with a uranyl to phytate mole ratio of 1:1, 2:1, 3:1, 4:1, 5:1, and 6:1. The pH of the solutions was in the range of 6-7.5. The vials were placed on a stir plate and mixed continuously for 72 hours. After mixing, the vials were centrifuged and the mother liquor removed. The precipitate was washed once with deionized water. The vials containing the precipitate were then placed in an oven and dried at 110-125 °C to constant weight. When drying was complete the precipitates were analyzed for Ca, U, P, and Na.

### Results

Results for elemental analysis of C, H, Ca, Na, and P and the calculated stoichiometries of the calcium phytate solids are reported in Table 1. The column labeled C/P is the weight ratio of carbon:phosphorus obtained analytically, which compares favorably with the theoretical ratio of 0.387 for phytic acid. Because of the good agreement between experimental and calculated values, we elected to analyze only for phosphorus (as an indicator of phytate) in the uranyl substituted samples. The composition of calcium phytate derived from precipitation studies as a function of calcium:phytate mole ratios indicates a surprisingly constant stoichiometry for  $\text{Ca}^{2+}$  substitution onto phytate of 4.6( $\pm$ 0.2). An earlier report noted a maximum Ca:phytate stoichiometry of 4.8:1 [8]



though the authors suggest that higher substitution is possible. The present results seem to contradict that suggestion. The water content of the calcium phytate is also constant at 6.7( $\pm$ 0.6). The only variation in the composition of the solids is in the ratio of  $\text{Na}^+$  to  $\text{H}^+$ , which probably changes as a function of the pH at which the precipitate was formed. We prepared a relatively large sample of  $\text{Ca}_{4.6}(\text{H},\text{Na})_{2.8}(\text{phytate}) \cdot 6.5 \text{H}_2\text{O}$  for an investigation of actinide cation exchange.

The elemental analysis and calculated stoichiometry of the solid samples prepared from soluble calcium and uranyl phytate are presented in Table 2. As noted above, the phosphorus analysis was used to calculate the phytate content of the solid. Hydrogen ions ( $\text{H}^+$ ) were added to achieve charge balance in the solid and the excess weight not accounted for by  $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{phytate}^{12-}$ , and  $\text{UO}_2^{2+}$  was assumed to represent waters of hydration. Corresponding results for samples prepared by contacting soluble uranyl nitrate with solid calcium phytate are given in Table 3. The solid species precipitated from aqueous solution suggest that, as expected, phytate has a much higher affinity for uranyl than calcium, as evidenced by the elevated ratio of  $\text{UO}_2^{2+}/\text{Ca}^{2+}$  in the solids prepared with low ratios of uranyl:calcium. The highest stoichiometry for  $\text{UO}_2^{2+}$  is 5.4 equivalents/phytate, 91% of the theoretical maximum of 6 equivalents. In the two phase experiments, the trend of uranyl substitution for calcium is less regular for 1-3 equivalents of uranyl, but ultimately reaches a maximum substitution of 96% at the 6:1 mole ratio.

### Discussion

The requirements for a viable cation exchanger based on Ca-phytate are 1) the calcium phytate should preferably have consistent, well-defined stoichiometry, 2)  $\text{Ca}^{2+}$  should be readily displaced by the target metal ions, 3) the calcium salt, the target metal ion salt and mixed cation salts of the phytate must all be insoluble, and 4) the capacity of the solid phytate for the metal ion should be known. Assuming electroneutrality of the insoluble metal phytate, the theoretical maximum capacity of phytic acid for divalent cations is  $6 \text{ eq } (\text{R-PO}_4^-) / (660.04 \text{ g/mole}) = 9.1 \text{ meq/g}$ . The acid ( $\text{H}^+$ ) capacity is 18.2 meq/g. By comparison, the total acid capacity of strongly acidic sulfonic acid cation exchangers (e.g. Dowex 50) is typically 4-6 meq/g, that for  $\text{Ca}^{2+}$  is 2-3 meq/g. For the

intended application, the kinetics of cation exchange are probably of little consequence. Nevertheless, kinetics of cation exchange onto calcium phytate should be rapid. The experiments conducted in this study had apparently achieved a steady-state within 72 hours.

The calcium/uranyl phytate solids formed as a result of precipitation from mixed calcium/uranyl phytate solution indicate that the insoluble material has a higher affinity for uranyl than calcium, as expected, and tend to be more strongly hydrated. Overall hydration of the solid increases more-or-less in direct proportion to the degree of uranyl substitution. Though these experiments were designed primarily to determine only the stoichiometry of the precipitated solids, in all cases, most of the uranium was removed from the solution phase. The high degree of loading of uranyl onto phytate (91% at 6:1 mole ratio) indicates that the sequestering ability of phytate is not reduced as uranyl loading increases to the theoretical maximum. In the two phase uptake experiments involving soluble  $\text{UO}_2^{2+}$  and solid  $\text{Ca}_{4.6}\text{H}_{2.8}\text{phytate}$ , uranyl is taken up to a slightly greater degree, though the calcium residue is somewhat higher in these experiments than in the parallel aqueous mixing experiment. The quality of the agreement between the initially homogeneous mixing and the solution solid mixing experiments with uranyl also argue that the system is at a steady state within the 72 hour period of mixing

### Conclusions

In this report, we have reported the dominant stoichiometry of the calcium salt of phytic acid and summarized the binding characteristics of  $\text{UO}_2^{2+}$  to the dominant calcium phytate salt. This series of experiments describes one aspect of our investigation of a two-step process for the removal of actinide ions from groundwaters. We have demonstrated that  $\text{Ca}_{4.6}\text{H}_{2.8}(\text{phytate}) \cdot 6.7 \text{H}_2\text{O}$  is readily and consistently formed upon contact between soluble phytic acid and  $\text{Ca}^{2+}$ . This material completely exchanges  $\text{Ca}^{2+}$  for  $\text{UO}_2^{2+}$  when contacted with solutions containing macroscopic concentrations of  $\text{UO}_2(\text{NO}_3)_2$ . Similar quantitative substitution of  $\text{UO}_2^{2+}$  for  $\text{Ca}^{2+}$  is observed when the solids are prepared from aqueous solution. Other aspects of the overall actinide isolation process have either been previously reported or are under continuing study.

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Table 1. Elemental analysis of calcium phytate samples ( $\pm 10\%$ ) and calculated stoichiometries of the solids.

<u>Ca:Phytate</u>						
<u>mole ratio</u>	<u>C, Wt. %</u>	<u>H, Wt. %</u>	<u>Ca, Wt. %</u>	<u>Na, Wt. %</u>	<u>P, Wt. %</u>	<u>C/P</u>
1:1	7.65	2.19	20.3	0.76	18.9	0.405
2:1	7.57	2.20	19.7	0.57	19.3	0.392
3:1	7.71	2.17	19.5	0.29	19.6	0.394
4:1	7.74	2.23	19.2	0.27	20.3	0.381
5:1	7.68	2.31	18.8	0.24	19.3	0.398
6:1	7.85	2.26	18.7	0.22	19.4	0.405

<u>Starting Ca:Phytate mole ratio</u>	<u>Calculated Stoichiometry of Solid Calcium Phytate</u>
1:1	$\text{Ca}_{4.88}\text{Na}_{0.32}\text{H}_{1.92}(\text{CH}(\text{PO}_4))_6 \cdot 7.0 \text{H}_2\text{O}$
2:1	$\text{Ca}_{4.71}\text{Na}_{0.24}\text{H}_{2.34}(\text{CH}(\text{PO}_4))_6 \cdot 6.8 \text{H}_2\text{O}$
3:1	$\text{Ca}_{4.54}\text{Na}_{0.10}\text{H}_{2.82}(\text{CH}(\text{PO}_4))_6 \cdot 6.5 \text{H}_2\text{O}$
4:1	$\text{Ca}_{4.43}\text{Na}_{0.11}\text{H}_{3.03}(\text{CH}(\text{PO}_4))_6 \cdot 5.6 \text{H}_2\text{O}$
5:1	$\text{Ca}_{4.47}\text{Na}_{0.10}\text{H}_{2.96}(\text{CH}(\text{PO}_4))_6 \cdot 7.4 \text{H}_2\text{O}$
6:1	$\text{Ca}_{4.41}\text{Na}_{0.09}\text{H}_{3.09}(\text{CH}(\text{PO}_4))_6 \cdot 7.2 \text{H}_2\text{O}$

Table 2. Elemental analysis and solid stoichiometry of calcium/uranyl phytate samples ( $\pm 10\%$ ) prepared from homogeneous aqueous solution.

U:Ca:Phytate mole ratio

<u>(initial)</u>	<u>Ca, Wt. %</u>	<u>P, Wt. %</u>	<u>U, Wt. %</u>	<u>Na, Wt. %</u>
1:5:1	5.43	11.3	35.2	1.99
2:4:1	2.65	10.0	37.4	1.21
3:3:1	1.83	10.5	43.4	0.81
4:2:1	0.72	9.11	48.4	0.35
5:1:1	0.10	7.86	52.4	0.36
6:0:1	0.01	7.71	53.6	0.08

Starting U:Ca:Phytate mole ratio

Calculated Stoichiometry of Solid Calcium Phytate

1:5:1	$(\text{UO}_2)_{2.43}\text{Ca}_{2.24}\text{Na}_{1.42}\text{H}_{1.24}\text{Phytate} \cdot 12.1\text{H}_2\text{O}$
2:4:1	$(\text{UO}_2)_{2.92}\text{Ca}_{1.23}\text{Na}_{0.98}\text{H}_{2.72}\text{Phytate} \cdot 19.6\text{H}_2\text{O}$
3:3:1	$(\text{UO}_2)_{3.23}\text{Ca}_{0.81}\text{Na}_{0.63}\text{H}_{3.29}\text{Phytate} \cdot 11.4\text{H}_2\text{O}$
4:2:1	$(\text{UO}_2)_{4.15}\text{Ca}_{0.37}\text{Na}_{0.32}\text{H}_{2.64}\text{Phytate} \cdot 13.9\text{H}_2\text{O}$
5:1:1	$(\text{UO}_2)_{5.21}\text{Ca}_{0.06}\text{Na}_{0.38}\text{H}_{1.08}\text{Phytate} \cdot 16.7\text{H}_2\text{O}$
6:0:1	$(\text{UO}_2)_{5.43}\text{Ca}_{0.00}\text{Na}_{0.07}\text{H}_{1.07}\text{Phytate} \cdot 16.4\text{H}_2\text{O}$

Table 3. Elemental analysis and solid stoichiometry of calcium/uranyl phytate samples ( $\pm 10\%$ ) prepared by contact of soluble uranyl nitrate with solid calcium phytate.

Solid calcium phytate and  $\text{UO}_2^{2+}$

<u>(initial)</u>	<u>Ca, Wt. %</u>	<u>P, Wt. %</u>	<u>U, Wt. %</u>	<u>Na, Wt. %</u>
1:4.6:1	13.6	14.4	20.3	<0.05
2:4.6:1	8.55	11.3	34.3	0.13
3:4.6:1	11.1	13.0	28.4	<0.05
4:4.6:1	2.36	8.76	49.3	<0.05
5:4.6:1	0.99	7.97	52.7	<0.05
6:4.6:1	0.29	7.50	55.4	<0.05

Starting U:Ca:Phytate mole ratio

Calculated Stoichiometry of Solid

1:4.6:1	$(\text{UO}_2)_{1.10}\text{Ca}_{4.38}\text{Na}_{0.07}\text{H}_{0.97}\text{Phytate} \cdot 9.4 \text{ H}_2\text{O}$
2:4.6:1	$(\text{UO}_2)_{2.37}\text{Ca}_{3.51}\text{Na}_{0.00}\text{H}_{0.24}\text{Phytate} \cdot 12.0 \text{ H}_2\text{O}$
3:4.6:1	$(\text{UO}_2)_{1.71}\text{Ca}_{3.96}\text{Na}_{0.00}\text{H}_{0.66}\text{Phytate} \cdot 9.0 \text{ H}_2\text{O}$
4:4.6:1	$(\text{UO}_2)_{4.39}\text{Ca}_{1.25}\text{Na}_{0.00}\text{H}_{0.72}\text{Phytate} \cdot 17.2 \text{ H}_2\text{O}$
5:4.6:1	$(\text{UO}_2)_{5.16}\text{Ca}_{0.58}\text{Na}_{0.00}\text{H}_{0.52}\text{Phytate} \cdot 14.8 \text{ H}_2\text{O}$
6:4.6:1	$(\text{UO}_2)_{5.77}\text{Ca}_{0.18}\text{Na}_{0.00}\text{H}_{0.10}\text{Phytate} \cdot 14.9 \text{ H}_2\text{O}$