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THE USE OF CARBONATE LIXIVIANTS TO REMOVE URANIUM FROM URANIUM-CONTAMINATED SOILS

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ABSTRACT

The objective of this research was to design an extraction media and procedure that would selectively remove uranium without adversely affecting the soils' physicochemical characteristics or generating secondary waste forms difficult to manage or dispose of. Investigations centered around determining the best lixiviant and how the various factors such as pH, time, and temperature influenced extraction efficiency. Other factors investigated included the influence of attrition scrubbing, the effect of oxidants and reductants, and the recycling of lixiviants. Experimental data obtained at the bench- and pilot-scale levels indicated 80 to 95% of the uranium could be removed from the uranium-contaminated soils by using a carbonate lixiviant. The best treatment was three successive extractions with 0.25 M carbonate-bicarbonate (in presence of KMnO_4 as an oxidant) at 40 C followed with two water rinses.

INTRODUCTION

Production of enriched uranium requires a variety of chemical and metallurgical processes. As a consequence, uranium in various forms has been released to the environment at a number of U.S. Department of Energy (DOE) facilities. Uranium sources include uranium airborne particulate coming from the stacks of the facilities as well as leaks and spills of uranium-rich solvents and process effluents generated in the wide assortment of aqueous and nonaqueous extraction and treatment processes related to uranium enrichment. One of the major receptors of uranium releases has been soils. The exact quantity of uranium-contaminated soils across the DOE sites is unknown.

One DOE site known to have significant quantities of uranium-contaminated soils is the facility formerly known as the Feed Materials

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Production Center at Fernald, Ohio. Activities at the Fernald site centered around production of purified uranium metal, and it is estimated that 1 to 1.5 million m³ of soil containing unacceptable levels of uranium are present in the top surface soil layers¹. Uranium concentrations are as high as 70,000 mg/kg in soils adjacent to the processing plants. It is estimated that >50% of the surface soils (to the top 0.5 m) contain unacceptable levels of uranium. At certain locations, (e.g., within the waste pit areas) significant contamination is also present at soil depths >4.5 m. To eliminate the need for collection and disposal of the uranium-contaminated soil in engineered landfills, methods to decontaminate the soil have been investigated.

The objective of this research is to design an extraction media and procedure that would selectively remove the uranium without adversely affecting the soils' physicochemical characteristics or generating secondary waste forms difficult to manage or dispose of. The work reported here summarizes the approach as to selection of extraction media and demonstrates the feasibility of using carbonate lixivants to decontaminate uranium-contaminated soils. Data are presented on how various scientific and engineering parameters (i.e., time, temperature, pH, concentrations and characteristics of lixivants, attrition scrubbing, additions of oxidants, successive and recycled lixivants) affected uranium removal from two soils at the Fernald site.

APPROACH

Characterization of Soils

Two soils considered to be representative of the uranium source terms at the Fernald site were used for testing. One included soil adjacent to a storage pad area because it represented uranium contamination by a soluble uranium source-term (i.e., dissolved uranium contained in run-off from the storage pad). The other soil, soil adjacent to a low-temperature waste incinerator, represented an air-borne source term of uranium at the Fernald site. Characterization studies have indicated that the predominant oxidation state of uranium in both of the soils was the uranyl form. Uranium in the incinerator soil was typically found to be associated with particles in the 1 to 10 μ m diameter range as calcium uranyl phosphate (meta-autunite) while uranium in the storage pad soil was seldom observed to be in discrete particles and was thought to be predominantly schoepite (uranyl oxide hydrate) in close association with the amorphous iron oxyhydroxide coatings on soil particles^{2,3,4}. The concentrations and distribution of uranium in the Fernald soils with respect to the particle size fractions of soil are presented in Table 1. Uranium concentrations in both

soils were fairly similar (538 and 446 mg/kg, respectively for the incinerator and storage pad soils). Greater than 70% of the uranium in the incinerator soil was associated with the silt and sand fractions (particle size fractions >0.002 mm). All of the size fractions contained uranium in concentrations >52 mg/kg, the targeted cleanup level.

Potential Removal Processes for Uranium

Soil washing in a conventional sense is based on a physical separation process. Methods of physical separation such as screening, classification (separation of soil particulate according to their settling velocities), and flotation are effective for soils in which a large amount of the contamination is concentrated in the fine-grain fraction (i.e., a small percentage of the soil volume). However, these methods are not effective in the removal of contaminants from heavy-textured soils [those soils containing high concentrations of clays (<0.002 mm) and silts ($0.053 - 0.002$ mm)]. Most soil washing operations employ a "cut" at particle size diameters >0.053 mm. For these Fernald soils a fractionation at 0.053 mm would remove 73% and 93% of the uranium from the incinerator and storage pad soil, respectively; but it would also constitute 94% and 75% of the two soils respectively. The remaining sand fractions of the two soils would contain uranium concentrations (1033 and 117 mg/kg) well in excess of the targeted 52 mg/kg cleanup level. Thus, a simple physical separation process would not be an effective cleanup technology and any successful approach would likely depend on a chemical extraction process.

Chemical extraction processes characteristically used to remove uranium from uranium ores are either acid or carbonate based extractions. For acid extractions, sulfuric acid, which is less expensive than nitric, is the most commonly used acid extractant and can remove 90 to 98% of the uranium. When ores containing uranium in the hexavalent state are leached with sulfuric acid, stable highly soluble complexes of uranyl sulfates are formed. The leaching of ores containing uranium in the tetravalent state is more difficult. Oxidizing agents such as sodium perchlorate and pyrolusite are often added to convert the uranium to the hexavalent state. Acid leaching of uranium from Fernald soils may not be applicable because these soils contain appreciable quantities of carbonate minerals [20 to 40% by weight of calcite and dolomite minerals⁵. Thus, the soils would require excessive quantities of acid and would generate copious quantities of secondary waste.

Screening tests were conducted to determine the effectiveness of extracting uranium from the Fernald soils using four chemical extraction processes⁶. These included sulfuric and citric acid extractions, a reductive dissolution process [citrate-bicarbonate-dithionite, called the CBD extraction] and extractions with carbonate. The sulfuric and citric acid extractions were conducted at pH values between 1 and 2 over extraction periods up to 18 hours. The CBD extraction is a commonly used procedure to selectively remove free iron hydrous oxides and hydroxides from soils⁷. It utilizes sodium dithionite for reduction purposes in a sodium carbonate buffer (pH 7.3) with sodium citrate for sequestering ferric and ferrous ions. Only the carbonate extractions are highly selective for uranium. Figure 1 illustrates that the carbonate extraction process removes similar levels of uranium but very little iron or calcium as compared to the CBD and the acid extractions. Thus, the CBD and acid extractants generate large volume secondary waste streams containing high levels of iron, calcium, sulfate, citrate, and uranium concentrations that are difficult to manage and dispose of. These extractions also generate wastewater streams that are difficult to treat chemically or physically.

Unlike acid and other chemical extractions, carbonate extractions are highly selective for uranium. The efficiency of the carbonate extractions is based on the formation of sodium or ammonium uranyl tri- and di-carbonates [$\text{UO}_2(\text{CO}_3)_2$ ² and $\text{UO}_2(\text{CO}_3)_3$ ⁴], highly stable, water-soluble anionic complexes. Oxidants such as potassium permanganate, hydrogen peroxide, perchlorates, and dissolved oxygen, as well as catalysts such as ferrocyanide or copper salts, may be used to increase the carbonate-bicarbonate extraction efficiency of uranium contained in primary minerals in the tetravalent form.

RESULTS AND DISCUSSION

Role of Carbonate-Bicarbonate Concentration and pH

Factorial-designed experiments were conducted on each of the contaminated soils. Three concentrations of total carbonate-bicarbonate (0.10, 0.25, and 0.50 M) were used at three initial pH levels—8, 9, and 10. Figures 2 and 3 illustrate the fractions of uranium extracted after 4 h in a rotary type extractor at a liquid-to-solid ratio of 10:1 (200 mL of extractant and 20 g of soil).

The removal of uranium from either of the two soils was statistically independent of extraction pH and concentration of carbonate-bicarbonate (at the 5% level). Removal of uranium from the incinerator soil (Fig. 2)

was more difficult than removal from the storage pad soil (Fig. 3). These experiments as well as experiments conducted in latter stages of the bench-scale studies indicated that concentrations of carbonate-bicarbonate in the range of 0.25 and 0.50 M were equivalent and that the pH of the extraction, if between 8.0 and 9.5, was not a major variable in extraction of uranium from the Fernald soils. Tests evaluating the recovery of uranium from the carbonate-bicarbonate lixiviants showed that more than 90% of the uranium could be removed from the lixiviant containing ≤ 0.25 M total carbonate-bicarbonate (pH 9.3) in a single contact with commercially available resins. At carbonate-bicarbonate concentrations of 0.5 M, uranium removal from the lixiviant was very poor. Thus, the pilot-scale testing was conducted at 0.25 M.

Influence of Time and Temperature

Two other variables investigated were temperature and time of extraction (when a 0.5 M carbonate-bicarbonate concentration was used). The influence of each appeared to be dependent on the particular uranium-contaminated soil being tested. For example, increased extraction time increased uranium extraction from the Fernald incinerator soil but not from the storage pad soil (Fig. 4). Similarly, increasing temperature increased removal of uranium from the incinerator soil but not from the storage pad soil (Fig. 5). The likely reason for such observations was that the composition of the storage pad soil made extraction easier under any circumstances and, therefore less dependent on the variables being investigated. The Fernald incinerator soil contained nearly equal concentrations of uranium in the sand and clay fractions whereas the clay fraction of the storage pad soil contained nearly ten times more uranium than the sand fraction (see Table 1). In addition to the simple influence of larger available surface area of uranium in the clay fraction of the storage pad soil, characterization data indicated the presence of recalcitrant low-fired uranium metaphosphate particles in the incinerator soil whereas the uranium form in the storage pad soil appear to be dominated by schoepite (uranyl oxide hydrate) in close association with amorphous iron oxyhydroxide coatings⁴.

Role of Attrition Scrubbing

Attrition scrubbing is a physical process intended to dislodge and remove or scarify finer size particulate from sand size particles. It is commonly used in the uranium mining and milling industry to enhance extraction efficiencies. Bench-scale studies revealed 10 to 15% better

extraction effectiveness for uranium from the Fernald incinerator soil when a Denver Equipment attrition scrubber was used (Model 533) instead of the simple end-to-end rotary extraction technique (liquid-to-solid ratio of 10:1). Interestingly, attrition scrubbing at 55% solids was not as effective as attrition scrubbing at 33% solids. No difference was noted in the extractability of uranium from the Fernald storage pad soil.

A pilot-scale study was also conducted to evaluate the influence of attrition scrubbing. The Fernald incinerator soil was fed to the pilot-scale attrition scrubber (a two-cell unit design with a capacity of 4 ft³). Samples were removed after 15, 30, 45, and 60 min of attrition and then used in a factorial designed experiment (four replicates) to investigate the removal of uranium as a function of carbonate-bicarbonate concentration (0.25 and 0.50 M) and temperature (25 and 40 C). This experiment indicated little advantage in the use of attrition scrubbing. Attrition scrubbing at 15 min released approximately 5% more uranium (across both temperatures and carbonate-bicarbonate concentrations) than no attrition scrubbing (Fig. 6). Attrition scrubbing at 30, 45, and 60 min yielded extraction efficiencies of uranium similar to those of samples not scrubbed. More detailed analysis indicated that prolonged attrition scrubbing (>30 min) followed with extraction at elevated temperatures actually decreased removal rates of uranium from the Fernald incinerator soil. Follow-up attrition studies in the laboratory with soil fractions of >20 μ m also showed little advantage in the use of an attrition scrubber.

Influence of a Chemical Oxidant

Chemical oxidants are used extensively in the industry to enhance oxidation of highly resistant uranite mineral forms (those minerals containing predominately tetravalent uranium) to the uranyl forms, which form soluble complexes with sulfuric acid and carbonate extracts. Potassium permanganate (KMnO₄), though expensive on a commercial scale, is a good oxidant for carbonate extractants because of its high oxidation capacity in mildly alkaline solutions. Potassium permanganate amendments (0.02 g of KMnO₄ per g of soil) increased extraction effectiveness of uranium by approximately 10% for the Fernald incinerator soil but had little influence on the ability of carbonates to extract uranium from the storage pad soil (Fig. 7).

Successive Extractions

To determine if successive extractions of carbonates would remove significantly more uranium than an initial single extraction, the Fernald

incinerator soil was extracted five times in succession with 0.1, 0.25, and 0.5 M carbonate-bicarbonate (pH 9.5, without KMnO_4 amendments). These extractions involved 16-h extraction times in a rotary extractor at a liquid-to-solid ratio of 2:1 (replicated in triplicate). At the higher carbonate-bicarbonate concentrations (0.25 and 0.50 M), approximately 90% of the extractable uranium was removed in the initial extraction. The remaining 8% and 2% were extracted in the second and third extractions. Uranium removal in the initial extraction was statistically the same for the 0.25 and 0.50 M concentrations and approximately 10% more effective than the 0.10 M carbonate-bicarbonate concentration.

The Potential to Recycle Lixiviant

An important aspect in determining the applicability of an extraction technology is the ability to recycle the extractant for additional extractions of uranium from soil. To test the applicability of recycling a sodium carbonate extractant, extractant from a previous carbonate extraction was used to extract uranium from soil. This process was repeated through seven recycles (Fig. 8). Fresh carbonate extract was added only to make up for entrained losses during the liquid-to-solid separation process (keeping the liquid-to-solid ratio equal to 2:1 during the extractant stage).

Extraction effectiveness was lowered to approximately 200 to 250 mg of uranium per kg of soil in the sixth and seventh recycles as compared to 300 to 350 mg of uranium per kg of soil in the initial two extractions. These data imply a slight decline in extraction effectiveness of uranium on continued recycle. However, uranium concentrations of 100 to 250 mg/L in the first and second recycle certainly do not significantly affect extraction efficiency and indicate that any recycled carbonate extractant containing these levels of uranium will not affect overall extraction effectiveness.

Pilot-Scale Studies

The pilot-scale study conducted at the Fernald site used carbonate extractants to remove uranium from the Fernald storage pad and incinerator soils. The facility operated in a batch-mode and treated between 350 and 400 lbs. of soil at a time. The facility contained trommel and vibrating screens, an attrition scrubber, a centrifuge, and several reaction vessels (one jacketed for temperature-related studies). Detailed operating procedures and treatments conducted are available⁸.

Several parameters were investigated, the main ones being the influence of attrition scrubbing and elevated temperatures on rates of uranium removal. The basic extraction process was carried out over a 2-h extraction period and used 0.25 *M* sodium carbonate-bicarbonate (pH 9.5) containing 0.02 g of KMnO_4 per g of treated soil. Uranium concentration in the extraction was measured at various extraction intervals and revealed that uranium removal was very rapid; 85% of what was extracted in 2 h was removed in the first 15 min. Heating to 40 C removed greater levels of uranium from the incinerator soil, and equilibrium levels of uranium in the reactor were attained more quickly, indicating that heating to 40 C should be a practical application in a commercial-scale operation.

The process removed approximately $90 \pm 5\%$ of the uranium from the storage pad soil and $80 \pm 5\%$ of the uranium from the incinerator soil. Starting concentrations of uranium in the storage pad soil ranged from approximately 1700 to 2000 mg/kg. Uranium concentrations in the treated storage pad soils were usually <150 mg/kg. Initial uranium concentrations in the untreated incinerator soil were not as high (ranging from approximately 500 to 1000 mg/kg), but treated concentrations were always <200 mg/kg. The best treatment was three successive extractions at 40 C with 0.25 *M* carbonate-bicarbonate followed with two water rinses. For the Fernald incinerator soil, this treatment produced a final product containing approximately 85 mg/kg uranium.

CONCLUSIONS

Experimental data obtained at the bench- and pilot-scale levels indicated 80 to 95% of the uranium could be removed from the Fernald soils by using a carbonate lixiviant. Uranium concentrations in the Fernald storage pad soil ranging from 1700 to 2000 mg/kg could be lowered to concentrations in the range of 100 to 150 mg/kg. Uranium in the Fernald incinerator soil was more difficult to extract; however, initial uranium concentrations were lower, ranging from 500 to 1000 mg/kg. Final concentrations in the carbonate-bicarbonate-treated Fernald incinerator soil ranged from 85 to 150 mg/kg. The best treatment was three successive extractions with 0.25 *M* carbonate-bicarbonate (in presence of KMnO_4 as an oxidant) at 40 C followed with two water rinses.

The influence of various factors on carbonate extraction of uranium from the Fernald soils can be summarized as follows: (1) initial extraction is rapid (<30 min) for 90% of extractable uranium, (2) increased extraction occurs when temperature is increased from 25 C to 40 C, but little effect is shown at >40 C., (3) equal extraction efficiency at concentrations of 0.25 and 0.50 *M* (especially at 40 to 45 C) appear to be equivalent, and both

concentrations have greater extraction efficiency than the 0.10 M concentration, (4) effect of pH does not vary between 8 and 10, (5) attrition scrubbing shows little effect, (5) data indicate a possible >10% but <20% increase in extraction efficiency when an oxidant is used (only for the incinerator soil), (6) approximately 85 to 90% of extractable uranium is removed in the initial extraction, and the remaining uranium is removed in the second and third extractions, (7) carbonate extractant can be easily recycled two and maybe three times with little effect on extraction efficiency of uranium.

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Table 1. Concentration and distribution of uranium in the Fernald soils

Soil	Size fraction (mm)	Particle size distribution (%)	Uranium concentration (mg/kg)	Uranium burden by size fraction (%)
Incinerator	Total soil	na ¹	538	na
	Sand (2.0 – 0.053)	12.5	1033	27
	Silt (0.053 – 0.002)	73.9	286	44
	Clay (<0.002)	13.6	1019	29
Storage Pad	Total soil	na	446	na
	Sand (2.0 – 0.053)	22.6	117	7
	Silt (0.053 – 0.002)	56.5	239	37
	Clay (<0.002)	20.9	983	56

¹na = not applicable

Figure 1. Quantities of calcium, iron, and uranium removed from the Fernald incinerator soil by the four extractants (CBD is a reductive dissolution process using a sodium citrate, bicarbonate, and dithionate extractant).

Figure 2. Fraction of uranium extracted from the Fernald incinerator soil as a function of final extraction pH and carbonate/bicarbonate concentration (0.10, 0.25, and 0.50 M).

Figure 3. Fraction of uranium extracted from the Fernald storage pad soil as a function of final extraction pH and carbonate/bicarbonate concentration (0.10, 0.25, and 0.50 M).

Figure 4. The influence of time on the removal of uranium from the Fernald soils by a 0.50 M carbonate-bicarbonate extractant.

Figure 5. The influence of temperature on the removal of uranium from the Fernald soils by 0.50 M carbonate-bicarbonate extractant.

Figure 6. The influence of attrition scrubbing on the removal of uranium from the Fernald incinerator soil (main effects across 0.25 and 0.50 M carbonate-bicarbonate concentrations and 25 and 40°C extraction temperatures).

Figure 7. The influence of adding potassium permanganate to carbonate-bicarbonate extractions for uranium removal.

Figure 8. The effect of recycling carbonate-bicarbonate extractant (0.50 M, 40°C, 15 h) on extraction of uranium from the Fernald incinerator soil.

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