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**Solvent Recycle/Contaminant Reduction Testing - Phase I
Task 3**

**Topical Report
June 1994 - December 1994**

July 1995

Work Performed Under Contract No.: DE-AC21-93MC30168

U.S. Department of Energy
Office of Environmental Management
Office of Technology Development
Washington, DC

For

U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

By
Babcock and Wilcox Company
Contract Research Division
Alliance, OH 44601

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Morgantown Energy Technology Center
P.O. Box 880
Morgantown, West Virginia 26507-0880

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EXECUTIVE SUMMARY

This work is being performed in response to DOE's request for research on methodologies for the decontamination of process equipment. Requirements of the decontamination technology being developed in this program were: (1) removal of as much contamination as possible in a cost effective manner, (2) minimization of the volume of secondary waste generated, and (3) minimization of worker exposure. The specific objective of this task of the test program was to recover or regenerate the cleaning solvent and remove the dissolved uranium from solution.

Two chemical cleaning solvents were developed in the earlier portions of the test program. The first solution contained 20 grams per liter ammonium carbonate, pH adjusted to 9.0 with ammonium hydroxide, with hydrogen peroxide added. The second solvent was similar to the first except it also had the chelating agent ethylenediaminetetraacetic acid (EDTA) added to the formulation. The addition of the EDTA improved the solvent's performance for contamination buried deeper into the components.

The ammonium carbonate solvent recycle test program resulted in five successful methods for removing uranium from the spent ammonium carbonate solvent with an efficiency of greater than 99.9%. The five successful recycle methods were:

1. acidification to pH 1.0 followed by the use of lime to a pH of 9.8 then caustic to a pH of 12.5
2. solvent pH reduction to 1.0 followed by the application of strong acid cation resin regenerated in the hydrogen form
3. application of strong base anion resin regenerated in the carbonate form at the existing solvent pH
4. application of strong base anion resin regenerated in the hydroxide form at the existing solvent pH
5. a precipitation technique involving application of trisodium phosphate after reducing the solvent pH to 2.0

The treatment process using anion resin in the carbonate form appears to have the best merit for recovery applications because it removes only uranium from solution. All other processes are acceptable and considered equal.

Only one method was found that could be used with the EDTA/ammonium carbonate decontamination solvent. This method consisted of reducing the solvent pH to less than 2.0 followed by the application of a uranium selective cation resin (Purolite S-940) in the hydrogen form. No precipitation techniques provided acceptable results with the EDTA - carbonate solvent system.

1.0 INTRODUCTION

The Department of Energy (DOE) is now faced with the task of meeting decontamination and decommissioning obligations at numerous facilities by the year 2019. Due to the tremendous volume of material involved, innovative decontamination technologies are being sought that can reduce the volumes of contaminated waste materials and secondary wastes requiring disposal. With sufficient decontamination, some of the material from DOE facilities could be released as scrap into the commercial sector for recycle, thereby reducing the volume of radioactive waste requiring disposal. Although recycling may initially prove to be more costly than current disposal practices, rapidly increasing disposal costs are expected to make recycling more and more cost effective (Reference 1). Additionally, recycling is now perceived as the ethical choice in a world where the consequences of replacing resources and throwing away reusable materials are impacting the well-being of the environment.

Current approaches to the decontamination of metals most often involve one of four basic process types: (1) chemical, (2) manual and mechanical, (3) electrochemical, and (4) ultrasonic (Reference 2). "Hard" chemical decontamination solutions, capable of achieving decontamination factors (Df's) of 50 to 100, generally involve reagent concentrations in excess of 5%, tend to physically degrade the surface treated, and generate relatively large volumes of secondary waste. "Soft" chemical decontamination solutions, capable of achieving Df's of 5 to 10, normally consist of reagents at concentrations of 0.1 to 1%, generally leave treated surfaces in a usable condition, and generate relatively low secondary waste volumes. Under contract to the Department of Energy, the Babcock & Wilcox Company is developing a chemical decontamination process using chelating agents to remove uranium compounds and other actinide species from process equipment. This report describes the technical approach being taken to achieve project objectives, and presents the results of Task 3 of the program, "Solvent Recycle/Contaminant Reduction Testing."

2.0 BACKGROUND/OBJECTIVES

2.1 Overall Program Background/Goals

This work is being performed in response to DOE's request for research on methodologies for the decontamination of process equipment. At the time of project initiation, DOE offered equipment from the gaseous diffusion plant at Oak Ridge, Tennessee, for use in the full scale demonstration of the developed technology. For this reason, the selected technical approach was to develop a process suitable for application to process equipment contaminated in this manner.

Internal components of equipment utilized in the uranium enrichment process were in contact with uranium hexafluoride (UF_6) gas at elevated temperatures over many years of operation. It is suspected that inleakage of air during operations and after plant shutdown have resulted in deposits of other uranium compounds, possibly UO_2F_6 and UF_4 . Additionally, occasional outleakage of process gas has left small radioactive deposits on external surfaces.

The compressor internals are expected to not only have surface contamination, but, due to long exposures at high temperatures, it is anticipated that the radioactive contaminants have penetrated the grain boundaries of the base metal. The Allis Chalmers centrifugal compressors in the K-27 building typically have housings constructed of carbon steel with less than 0.001 inch of internal nickel plating. The rotors are typically made of Monel; the shafts, of carbon steel.

DOE's preliminary evaluation of the potential for recycling the compressor materials was that some equipment parts, such as the compressor base and rotor shaft, might be surface contaminated only, and, as such, releasable for unrestricted use under existing guidelines (eg. NRC Regulatory Guide 1.86, DOE Order 5400.5). Other parts, such as the compressor housing, rotor, and piping, were expected to be bulk contaminated with radionuclides, preventing unrestricted release and making the equipment attractive for reuse within the DOE complex. Requirements of the decontamination technology applied to the compressors were: (1) removal of as much contamination as possible in a cost effective manner, (2) minimization of the volume of secondary waste generated, and (3) minimization of worker exposure. The technical approach and test plan for this program were developed in accordance with this information.

2.2 Results - Solvent Screening/Refinement Tests

During Task 2 of this program, a large number of candidate solvents were evaluated for uranium dioxide dissolution and base metal corrosion response (Reference 3). The combination of chelant, ammonium carbonate, and hydrogen peroxide at a pH of 9.0 (adjusted with ammonium hydroxide) was found to effect virtually 100% dissolution of the uranium dioxide placed in the test flask, within the first two hours of exposure. The solvent was effective in uranium dissolution at room temperature. Based on the results of the screening and refinement

test program, two solvent systems were concluded to provide optimum uranium dioxide dissolution:

- (1) EDTA, ammonium carbonate, and hydrogen peroxide: pH 9.0 with NH_4OH
- (2) ammonium carbonate and hydrogen peroxide: pH 9.0 with NH_4OH .

Testing during Task 2 of this program indicated that the basic solvent systems were effective over a wide range of concentrations. This can allow for tailoring the solvent for differing levels of contamination and to minimize chemical usage.

Based on the Task 2 testing, the recommended application scenario for the process was as follows:

1. 2-Hours at room temperature with either solvent
2. Heatup to 66°C or 93°C and hold for approximately 4-hours
3. Cooldown to room temperature
4. Inject peroxide to remove additional uranium contamination uncovered by the high temperature exposure.
5. Hold 2 hours at room temperature.

By starting with an initial low temperature application, all readily available uranium contamination is easily removed. Heatup to 93°C (200°F) appears to dissolve the remaining readily available uranium contamination. The 93°C application results in higher corrosion rates than the 66°C (150°F) application. This is necessary to expose contamination buried more deeply into the component. Even at 200°F, the corrosion is uniform in nature.

Note that at the time of the Task 3 testing, both the 150°F and 200°F application scenarios were under consideration. Data with spent solvents using both application scenarios is presented in this report. The ultimate process recommendation was to use the 200°F application scenario.

2.3 Specific Objectives - Recycle Testing

As mentioned previously, one requirement of the developed process was to minimize the secondary waste generation from the decontamination process. The objective of this task in the program was to accomplish waste minimization by means of recycling of the spent solvent. After the decontamination process has been performed, removed radionuclides will be present as dissolved species in the chelating solution. The objective of the tests described in this report was to develop a process to break the chelant-contaminant bond, remove the dissolved contaminants from solution, and to regenerate the chelant solution.

When dealing with chelating agents, the extent to which complex formation occurs is defined as K (a stability constant). For the metal/chelate reaction

K is defined as follows:

$$K = \frac{[MY^{(n-m)}]}{[M^{+n}][Y^{-m}]}$$

where, K = constant defining the ratio of the complexed metal ion in equilibrium to the free metal ion in solution

M = molar concentration of the metal ion being complexed

Y = molar concentration of the chelant.

For a chelant to be effective at dissolving a metal species and holding it in solution, the K value, or stability, of the complex must be high. Therefore, once metals are chelated, their removal is difficult because of the high stability of the chelate complexes. To separate the chelant from the metal, it is necessary to alter chemical conditions to make the chelate less stable and to promote the formation of an insoluble compound with the metal. For example, it was anticipated that a high pH solution could promote uranium hydroxide formation.

B&W has performed laboratory research on the removal of iron from EDTA chemical cleaning solutions. Two approaches were applied. The first technique, in which solution pH was increased to 11, resulted in a filter cake of precipitated contaminant and a regenerated solvent solution. A second method broke the chelant-contaminant bond by reducing the solution pH to <1.5, resulting in formation of the insoluble tetra-acid EDTA. It was demonstrated that the precipitated EDTA could be filtered and stored for reuse.

Another technique which was investigated during this study was ion exchange. Ion exchange is the process of removing ions from solution in an equivalent exchange for other preferred ions supplied by a solid material having a special structure to do this. This solid ion exchange material (resin) must either be recharged periodically with the preferred ions or discarded. For this application, the likely preferred approach would be to discard the resin as a solid radioactive waste.

Tests were run to evaluate whether these techniques could be used on the selected solvent systems for regeneration of the spent solution. Primary emphasis was placed on removing the contaminants from solution and regenerating the solvent.

3.0 TEST PROGRAM RESULTS AND DISCUSSION

3.1 Introduction

As previously presented, two chemical cleaning solvents were developed in the decontamination test program for removing radioactive species from metal surfaces. The first solution contained 20 grams per liter ammonium carbonate, pH adjusted to 9.0 with ammonium hydroxide. Ten milliliters of hydrogen peroxide were also added as an oxidant. This solvent proved to be very effective in the dissolution of powdered uranium dioxide. However, the solvent exhibited no corrosive tendencies to carbon steel. Since the radioactive contaminants have penetrated the grain boundaries of the components to be cleaned, it was believed the solvent must have corrosive tendencies to remove several mills of metal surface and expose the radioactive contaminants. The second solvent was identical to the first along with the addition of 50 grams per liter of ethylenediaminetetraacetic acid (EDTA), pH adjusted to 9.0 with ammonium hydroxide. At this pH the EDTA is primarily present as the triammonium salt.

Recycle testing was performed separately on the two decontamination solutions. The results of the testing are presented separately in the following sections. The dissolved uranium content of the test solutions was analyzed before and after treatment. The success criteria established for this testing was a dissolved uranium removal efficiency from the solvent of greater than 99.9%.

3.2 Treatment of Spent Ammonium Carbonate Solvent

The first set of recovery tests was run on the spent solvent containing only the ammonium carbonate. The solvent was treated by means of pH adjustment to effect precipitation and also by ion exchange. Results of testing of both techniques are discussed in this section.

3.2.1 Test Solvent

Spent ammonium carbonate solvent from Task 2 of this program was used for testing under this task. The initial pH of the test solution ranged from 9.0 to 9.1. The dissolved uranium content of this spent solvent was 3778 ppm. In all cases the treated solvent was analyzed for dissolved uranium for comparison to this original dissolved uranium content.

3.2.2 Precipitation Techniques

Table 1 contains a summary of the tests performed using precipitation techniques to treat the spent ammonium carbonate solution. Each of these tests is discussed in detail in the following section.

Test 1

Since ammonium carbonate cannot be easily removed from solution, the first approach for treating the spent solvent was to raise the solution pH and attempt to precipitate heavy metals

and radioactive species as the alkaline hydroxides. A sample of spent solvent was pH adjusted very slowly to 12.5 using pelleted sodium hydroxide. A yellow precipitate formed. After 2 hours the solution was filtered through a 0.45 micron Millipore filter membrane and submitted for uranium analysis. This treated sample contained 1468 ppm. This equated to a removal efficiency of only 61%. These test results indicated that a large portion of the uranium is tightly complexed by the ammonium carbonate.

Tests 2 and 3

Another test was run which first expelled the carbonate from solution followed by treatment with sodium hydroxide. A 100 mL sample was reduced in pH from 9.09 to 1.29 using 1N hydrochloric acid. The pH adjustment required 100 mL of the acid. At a pH of 6.43, carbon dioxide gas bubbles started to form in the spent solvent. After reaching the prescribed pH, the solution was stirred for a period of 1 hour until all signs of carbon dioxide evolution stopped. The solution pH was then elevated back to 9.0 using 97.5 mL of 0.563 N sodium hydroxide. At a solution pH of 9.0 a small amount of white precipitate formed. After standing for 2 hours the solution was filtered and submitted for uranium analysis. The solution was found to contain 31.3 ppm uranium (99.2% removal). This test was repeated (designated Test 3), except the solution pH was increased to 12.5. The filtered sample was found to contain 10.3 ppm of uranium (99.7% removal). These results were encouraging but still below the target of greater than 99.9% removal.

Test 4

A test was run to determine the effects of hydrazine on the precipitation of uranium at the elevated pH. Thirty mL of 30% hydrazine was added to 200 mL of spent solvent. The resultant solution pH was 9.68. The solution pH was increased with sodium hydroxide to a level of 12.5. A precipitate started to form at a solution pH of 9.8. After reaching a pH of 12.5 the solution was allowed to settle for a period of 1 hour then filtered through a 0.45 millipore. Analysis of the filtered sample showed 144 ppm uranium (96.2% removal).

Tests 5 and 6

The effect of hydrazine in the presence of a heavier precipitate was investigated with the use of lime. Thirty mL of hydrazine was added to 200 mL of spent solvent. After thirty minutes, lime (calcium oxide) was added to reach a solution pH of 9.8 followed by the addition of sodium hydroxide to a pH of 12.5. A heavy precipitate formed. After 10 minutes the solution was filtered and submitted for analysis. Uranium remaining in solution was 480 ppm.

This test was repeated without the use of hydrazine. Lime was used to a pH of 9.8 and caustic (NaOH) to a pH of 12.5. After filtration the solution was found to contain 21.8 ppm of uranium. This testing indicated that the presence of a strong reducing agent (hydrazine) hindered the removal efficiency using this precipitation technique. The 99.4% removal efficiency obtained in Test 6 was still below the target of greater than 99.9%.

Table 1
Results - Treatment of Spent Ammonium Carbonate Solutions
By Precipitation Techniques

Starting Solvent: 3778 ppm Dissolved Uranium - pH 9.1

Test #	pH Adjustment	pH Additives	Other Chemical Additives	Dissolved Uranium (ppm)	Removal Efficiency (%)
1	12.5	NaOH	None	1468	61
2	1.29/9.0	HCl/NaOH	None	31.3	99.2
3	1.29/12.5	HCl/NaOH	None	10.3	99.7
4	12.5	NaOH	N ₂ H ₄	144	96.2
5	9.8/12.5	Ca(OH) ₂ /NaOH	N ₂ H ₄	480	87.3
6	9.8/12.5	Ca(OH) ₂ /NaOH	None	21.8	99.4
7	1.0/9.8/ 12.5	HCl/ Ca(OH) ₂ / NaOH	None	<3.0	>99.9
8	1.0/12.5	HCl/NaOH	CaF ₂	34.5	99.1
9A	2.0	HNO ₃	NaCl	433	88.5
9B	2.0	HNO ₃	Na ₃ PO ₄	<3.0	>99.9
9C	2.0	HNO ₃	NaNO ₂	726	80.8
9D	2.0	HNO ₃	NH ₄ HF ₂	268	92.9

Test 7

Another sample was run to determine the effects of lime and caustic on uranium precipitation in the absence of a carbonate residual. A 200 mL sample was treated with 8 mL of concentrated hydrochloric acid. The solution pH was reduced to 1.0. Once the outgassing of the solution stopped, the solution pH was elevated to 9.8 with lime and to 12.5 with sodium hydroxide. After 1 hour the solution was filtered through a Whatman 40 filter paper then through a 0.45 micron Millipore. Uranium concentration of the filtered sample was found to be less than 3.0 ppm (the detection limit of the method employed). This combination of pH adjustments resulted in the target removal efficiency of greater than 99.9%.

Test 8

The use of lime in the precipitation process generally results in an excessive volume of heavy sludge. This was the case in the above test runs using lime. Another test was run to evaluate the effects of calcium on the precipitation of uranium without the addition of lime. Two hundred mL of sample was pH adjusted to 1 with concentrated hydrochloric acid. One gram of calcium fluoride was added and the solution pH readjusted to 12.5 with sodium hydroxide. A yellow orange precipitate formed. The sample was filtered and analyzed for uranium. The solution was found to contain 34.5 ppm uranium. This technique did not meet the objective of greater than 99.9% removal.

Test 9

A final set of tests was run using the spent ammonium carbonate solvent. These tests attempted to precipitate uranium at the lower pH in conjunction with evolution of carbonate. Four 200 mL aliquots were reduced to a pH of 2.0 using 7.0 mL of concentrated nitric acid per aliquot. One reagent was added to each of the aliquots. The reagents consisted of one gram quantities of sodium chloride (Test 9A), trisodium phosphate (Test 9B), sodium nitrite (Test 9C) and ammonium bifluoride (Test 9D). The reagents were added, the solutions mixed thoroughly and allowed to stand for 20 minutes. Each sample was then filtered through a 0.45 micron millipore and analyzed for uranium. Uranium results were 433 ppm, <3.0 ppm, 726 ppm and 268 ppm respectively. Only Test 9B (trisodium phosphate) met the greater than 99.9% removal criteria.

3.2.3 Ion Exchange Techniques

Removal of uranium by ion exchange resin was also evaluated using the spent ammonium carbonate solution. A 1 inch diameter column was loaded with 150 mL of resin. In some instances the spent solvent was pretreated. The same spent solvent as used in the precipitation phase of the program was used during the ion exchange phase (3778 ppm dissolved uranium).

Table 2 contains a summary of the tests performed using ion exchange techniques to treat the spent ammonium carbonate solution. Each of these tests is discussed in detail in the following section.

Test 10

In Test 10, the 1 inch diameter ion exchange column was loaded with standard 8% divinylbenzene gel type I strong acid cation resin. The resin was regenerated with sulfuric acid to a level of 6 pounds per cubic foot. The spent ammonium carbonate solvent was first filtered through a Whatman 40 filter paper to remove suspended matter, then processed through the resin column at the normal solution pH of 9.0. The resin column immediately filled with gas bubbles as the carbonate was converted to carbon dioxide gas. Gas evolution resulted as the hydrogen ions on the resin were exchanged for ammonia in solution. This caused the solution pH to drop and the carbonate was converted to carbon dioxide gas.

Table 2
Results - Treatment of Spent Ammonium Carbonate Solutions
By Ion Exchange Techniques

Starting Solvent: 3778 ppm Dissolved Uranium - pH 9.0

Test #	pH Adjustment	Resin Type	Resin Form	Dissolved Uranium (ppm)	Removal Efficiency (%)
10	None	Strong Acid Cation	Hydrogen	NA*	NA*
11	1.0 with H ₂ SO ₄	Strong Acid Cation	Hydrogen	<3.0	>99.9
12	None	Strong Base Anion	Hydroxide	<3.0	>99.9
13	None	Strong Base Anion	Carbonate	<3.0	>99.9

* Not Applicable, Gas Blockage of the Column - No Analysis Performed

Test 11

Test 10 was rerun but the test solution was first reduced to a pH of 1.0 and the carbon dioxide allowed to escape. The resin column was regenerated with sulfuric acid before this test. Spent solvent was passed through the column at a rate of 8 to 15 ml per minute. Samples were collected after 200 and 500 mL of solvent passed through the column. Test results for both samples collected showed less than 3.0 ppm uranium. These results met the removal criteria of greater than 99.9% and indicated good ion exchange capacity.

Test 12

A standard type I strong base anion resin was also evaluated for uranium removal. One hundred and fifty mL of a standard quaternary amine type I resin was loaded into the 1 inch diameter by 12 inch long column. The resin was regenerated at the rate of 4 pounds of caustic per cubic foot of resin. The spent solvent was passed through the column at the existing pH of 9.0. The solution was first filtered to remove suspended matter then passed through the resin column at the rate of 8 to 15 mL per minute. A sample was collected for uranium analysis after 500 mL of solvent had passed through the column. The uranium content was found to be less than 3.0 ppm (greater than 99.9% removal).

Test 13

Another test was run with the resin regenerated in the carbonate form. If successful the exchange material would have a much higher capacity as the resin would not be quickly exhausted by the high carbonate loading. The resin was regenerated with ammonium carbonate to a level of 4 pound per cubic foot as carbonate. The spent solution was filtered and passed through the column at the rate of 8 - 15 mL per minute. A sample of the column effluent was analyzed after passing 500 mL of solvent through the bed. Results showed less than 3.0 ppm uranium in solution, again meeting the removal criteria.

3.2.4 Summary - Solvent Recycle With Ammonium Carbonate Solvent

The ammonium carbonate solvent recycle test program resulted in five successful methods for removing uranium from the spent ammonium carbonate solvent. In each case the uranium content was reduced from 3778 ppm in the spent solvent to less than the detectable limit of 3.0 ppm as determined by ICP (greater than 99.9% removal). The five successful recycle methods were:

1. acidification to pH 1.0 followed by the use of lime to a pH of 9.8 then caustic to a pH of 12.5
2. solvent pH reduction to 1.0 followed by the application of strong acid cation resin regenerated in the hydrogen form
3. application of strong base anion resin regenerated in the carbonate form at the existing solvent pH
4. application of strong base anion resin regenerated in the hydroxide form at the existing solvent pH
5. a precipitation technique involving application of trisodium phosphate after reducing the solvent pH to 2.0.

The treatment process using anion resin in the carbonate form appears to have the best merit for recovery applications because it removes only uranium from solution. In essence this method regenerates the solvent for reuse, assuming there is not a large pickup of heavy metals such as copper and iron. The method of reducing the solvent pH followed by cation exchange complicates the process and removes both the uranium and carbonate solvent from solution. The precipitation techniques also remove both the uranium and carbonate solvent from solution.

3.3 Treatment of EDTA/Ammonium Carbonate Solvent

The first set of recovery tests was run on the spent solvent originally consisting of 50 grams per liter EDTA, 20 g/L ammonium carbonate and the solution pH adjusted to 9.0 with ammonium hydroxide. Ten mL of 50% hydrogen peroxide was added to the test solution at the

beginning of each test run; in some tests, hydrogen peroxide additions were also made during the test. As with the carbonate only solvent, precipitation and ion exchange techniques were both evaluated and will be presented in this section.

3.3.1 Test Solvent

Spent EDTA/carbonate solvent from Task 2 of this program was used for testing under this task. The initial pH of the test solution ranged from 9.0 to 9.1. The dissolved uranium content of this spent solvent was 3704 ppm. In all cases the treated solvent was analyzed for dissolved uranium for comparison to this original dissolved uranium content.

Note that two laboratory test procedures were used in Task 2 with the solvent containing EDTA, ammonium carbonate and hydrogen peroxide. One procedure consisted of two test variations at a temperature of 150°F and the other procedure consisted of two test variations at a temperature of 200°F. Each test variation consisted of either initially exposing the solvent to uranium dioxide powder at ambient temperature followed by increasing solution temperature or by using the higher temperature first then going to ambient for a designated period of time. Solvent recovery work used a considerable amount of solvent from both the 150°F and 200°F tests. As will be discussed in this section, recovery tests on the solvents exposed at the lower temperature were not sufficiently successful, whereas a procedure was developed to both remove the EDTA and uranium from solvents applied at the higher temperature. The following section presents the results of the various techniques used in these recovery tests.

3.3.2 Recovery Tests on Solvents Exposed to 150°F

B&W has a considerable amount of experience in the recovery of EDTA from solution by chemical precipitation. It is known that the tetraacid form of EDTA is insoluble at a solution pH of 2.0 or less. Previous B&W recovery tests with solvents containing EDTA complexed with iron showed that at least 95% of free and complexed EDTA can be recovered simply by reducing the solution pH to <2.0.

3.3.2.1 Precipitation Techniques

Table 3 contains a summary of the tests performed using precipitation techniques to treat the spent 150°F EDTA/ammonium carbonate solution. Each of these tests is discussed in detail in the following section.

Tests 14 and 15

The first two recovery tests looked at the effects of reducing the solvent pH with both hydrochloric and nitric acids. In both cases the solution pH was dropped to less than 2.0, but no precipitate formed. In one test the solution pH was readjusted to pH 12.5 to determine if uranium could be precipitated from solution. In this case a precipitate again did not form. The tests were repeated after adding 10 mL of hydrogen peroxide to each of the 200 mL aliquots of the solvent. Again there was no formation of a precipitate at either the low or high pH.

Table 3
Results - Treatment of Spent 150°F EDTA/Ammonium Carbonate Solutions
By Precipitation Techniques

Starting Solvent: 3704 ppm Dissolved Uranium - pH 9

Test #	pH Adjustment	pH Additives	Other Chemical Additives	Dissolved Uranium (ppm)	Removal Efficiency (%)
14	<2.0	HNO ₃	None	NP*	0
15	<2.0/12.5	HCl/NaOH	None	NP	0
16	12.5/<2.0	NaOH/HCl	Air Sparge	NP	0
17	12.5/<2.0	NaOH/HCl	CaF ₂	NP	0
18	<2.0	HCl	NaOCl**	1108	70.1
22	<2.0	HCl	Heat***	162	95.6

* NP = No Precipitate formed, therefore no analysis performed

** Various other coagulants also evaluated, only sodium hypochlorite resulted in a precipitate

*** Solution heated to 200°F and held for 2 hours prior to pH adjustment

Tests 16 and 17

The next group of tests looked at the effects of increasing the solvent pH to 12.5 to precipitate uranium, followed by bubbling air through the solvent to expel the ammonia, then reducing the solvent pH to <2.0 for precipitation of EDTA. The first test in this group consisted of initially raising the solution pH to 12.5 with sodium hydroxide. Again, there was no formation of a precipitate.

For the next test, 1 gram of calcium fluoride was added to the solvent followed by increasing the solvent pH to 12.5. A small amount of white precipitate formed at the elevated pH. The solution was filtered through a 0.45 micron filter. Air was then bubbled through the solution until all noticeable smell of ammonia had dissipated. The solution pH was then reduced to less than 2.0 with hydrochloric acid. There was no precipitation of EDTA. At this point it was evident that neither EDTA nor uranium could be easily precipitated from the spent solvent.

Test 18

A series of tests (all designated in this report as Test 18) were run using a number of chemicals that are frequently used in coagulation processes to generate precipitates and also chemical species that have been used to recover uranium from solution by precipitation. Chemicals such as sodium bisulfite, lanthanum chloride, sodium acetate, chrome in the form of

sodium dichromate and sodium hypochlorite were evaluated. Only the hypochlorite formed a precipitate when added to spent solvent that had been pH adjusted to less than 2.0. After filtering off the EDTA precipitate the filtrate was analyzed for free EDTA. The sample was found to contain 15.7 grams per liter EDTA and 1108 ppm uranium. This test was repeated, but the results were similar.

Test 22

After the Test 18 series, ion exchange was evaluated. The results from the ion exchange testing are detailed in the following section. However, results were not favorable and a final precipitation test was run in this series (designated Test 22). All testing of spent solvents with EDTA to this point were solvents that were heated to 150°F during the dissolution tests. The last test with this type of sample consisted of heating the solvent to 200°F for 2 hours followed by pH reduction to a pH of <2.0. As the solution cooled a precipitate formed. The precipitate was filtered off and the filtrate analyzed for both EDTA and total uranium. Residual EDTA was found to be <1.8 grams per liter (detection limit for the test method) and the uranium was 162 ppm. This was the last test run with solutions that were tested at the lower temperature because it appeared that solvents that were exposed to higher temperatures were more amenable to solvent recovery. In addition, at this time in the program it appeared that the higher solvent temperature would in all probability be the recommended approach.

3.3.2.2 Ion Exchange Techniques

Table 4 contains a summary of the tests performed using ion exchange techniques to treat the spent 150°F EDTA/ammonium carbonate solution. Each of these tests is discussed in detail in the following.

Test 19

Uranium removal from spent solvent was evaluated using a standard type I gel type strong base anion resin regenerated in the hydroxide form. The laboratory standard 1 inch diameter by 12 inch long column was loaded with 150 mL of ion exchange material. After the column was regenerated with caustic and thoroughly rinsed down, 200 mL of the solvent was passed through the column at the rate of 8 - 15 mL per minute. An effluent sample was then collected and analyzed for uranium content. The uranium concentration of the spent solvent was reduced from 3704 ppm to 29.2 ppm after ion exchange. Twenty nine ppm at the column effluent after a 200 mL throughput is a rather high leakage rate from ion exchange resin. From this data, it appeared that the resin column had become exhausted due to the high concentration of carbonate in solution.

Test 20

Test 19 was repeated with the resin column regenerated in the carbonate form with ammonium carbonate. After passing 200 mL of solvent through the column a sample was collected for analysis. This sample was found to contain 585 ppm. For this particular type of ion exchange resin it appeared that the resin has a higher affinity for carbonate than for uranium.

Table 4
Results - Treatment of Spent 150°F EDTA/Ammonium Carbonate Solutions
By Ion Exchange Techniques

Starting Solvent: 3704 ppm Dissolved Uranium - pH 9.0

Test #	pH Adjustment	Resin Type	Resin Form	Dissolved Uranium (ppm)	Removal Efficiency (%)
19	None	Strong Base Anion	Hydroxide	29.2	99.2
20	None	Strong Base Anion	Carbonate	585	84.2
21	<2.0 with H ₂ SO ₄	Strong Acid Cation	Hydrogen	1010	72.7

Test 21

Test 20 was repeated using a standard type I strong acid cation resin regenerated in the hydrogen form with sulfuric acid. The spent solvent was pH adjusted to 2.0 with sulfuric acid prior to treatment by ion exchange. An effluent sample was collected after passing 200 mL of solvent through the resin bed. Uranium content was found to be 1010 ppm.

3.3.3 Recovery Tests on Solvents Exposed to 200°F

3.3.3.1 Precipitation Techniques

A neutralization curve was established for the spent solvent containing 50 g/L EDTA, 20 g/L ammonium carbonate and 10 mL of 50% hydrogen peroxide. A 100 mL sample was titrated using concentrated sulfuric acid. The original sample pH was 9.3 and it required 3.4 mL of concentrated sulfuric to reduce the solution pH to 1.42. The titration curve is shown as Figure 1. This information was required to provide an estimate of chemical usage for field recycle efforts.

Just two trials were performed using precipitation-only techniques with the spent 200°F EDTA - carbonate solution. Both trials resulted in precipitation, but were below the target removal efficiency of greater than 99.9%. Details of these two trials follows.

Test 22

Precipitation of both EDTA and uranium was attempted by adjusting the solution pH to 1.0 with sulfuric acid. A white precipitate formed which was filtered after allowing sufficient time for all precipitation to take place. The solution was then pH adjusted to 12.5 with sodium hydroxide. A brown precipitate formed. This solvent was originally tested in the presence of carbon steel corrosion test coupons, consequently the brown precipitate which formed was presumably iron hydroxide. After filtering off the precipitate the solution was analyzed and found to contain 1048 ppm uranium (a removal efficiency of only 72%).

Test 23

Test 22 was repeated with the addition of 1 gram trisodium phosphate after reaching the final pH of 12.5. A similar technique was successful with the carbonate only solvent. The solution was filtered and the filtrate analyzed for uranium. Test results showed 230 ppm uranium. This was a marked improvement (94% removal efficiency), but still far from the target of greater than 99.9% removal efficiency. At this time, a decision was made to concentrate efforts on ion exchange techniques.

Figure 1
Neutralization Curve for 100 Milliliters
Spent EDTA/Carbonate Solution

3.3.3.2 Conventional Ion Exchange Techniques

Conventional ion exchange resins were initially evaluated with the solvents exposed to higher temperature. The spent EDTA/carbonate solvent used in this evaluation contained 3455 ppm dissolved uranium.

Table 5 contains a summary of the tests performed using conventional ion exchange techniques to treat the spent 200°F EDTA - ammonium carbonate solution. Each of these tests is discussed in detail in the following.

Table 5
Results - Treatment of Spent 200°F EDTA/Ammonium Carbonate Solutions
By Conventional Ion Exchange Techniques

Starting Solvent: 3455 ppm Dissolved Uranium - pH 9.0

Test #	pH Adjustment	Resin Type	Resin Form	Dissolved Uranium (ppm)	Removal Efficiency (%)
24	None	Strong Base Anion	Hydroxide	2000	42.1
25	<2.0 with H ₂ SO ₄	Strong Base Anion	Hydroxide	<3.0	>99.9%*
26	<2.0 with H ₂ SO ₄	Strong Base Anion	Carbonate	19.8	99.4
27	None	Strong Base Anion	Hydroxide	<3.0	>99.9%*
28	None	Strong Base Anion	Carbonate	60.2	98.3
29	<2.0 with H ₂ SO ₄ - 12.5 with NaOH	Strong Base Anion	Hydroxide	133	96.2

* Although >99.9% removal efficiency was achieved, the resin capacity was limited (see detailed discussion)

Test 24

A conventional strong base type I anion exchange resin regenerated in the hydroxide form (with caustic) was used to process the solvent at the application pH (9.0). The solvent was filtered, then passed through the laboratory standard 1-inch diameter by 12 inch deep column. Solution flow rate was 8 - 15 mL per minute.

A sample of the influent solvent was taken along with a column effluent after processing 200 mL of solvent. Both samples were analyzed for uranium. As previously reported, the original solvent contained 3455 ppm U. The effluent sample contained 2000 ppm.

Tests 25 and 26

Test 24 was repeated using an anion resin both in the hydroxide (Test 25) and carbonate (Test 26) forms. The test solution was first pH adjusted to <2.0 with sulfuric acid to reduce the carbonate loading on the resin. Effluent samples were collected after passing 200 mL of solvent through each column. Test results showed 19.8 ppm uranium in the effluent of the carbonate regenerated resin (Test 26) and 2.4 ppm with the resin in the hydroxide form (Test 25). After 200 mL of solvent, the Test 25 results met the acceptance criteria of greater than 99.9% removal. The influent sample was also analyzed for EDTA after the solution was pH adjusted and filtered. Results showed 4.3 grams per liter of EDTA remaining in solution.

Flow was continued through both columns and additional samples collected after processing 300 mL of solvent. Test results showed 205 ppm uranium in the column regenerated with sodium hydroxide and 45.5 ppm in the carbonate column. These results indicated that a conventional strong base type I anion exchange resin has a limited capacity for uranium, apparently due to the solvent's high anion content.

Test 27

Another test run was made with the anion exchange column to check capacity. The ion exchange column was regenerated in the hydroxide form using sodium hydroxide. A filtered sample of spent solvent that was tested at 200°F was passed through the column at the original solvent pH of 9.0 - 9.3. Samples were collected after passing 150, 300 and 450 mL of solvent through the column. The samples were found to contain <3.0 ppm, 35.8 ppm and 51.4 ppm uranium respectively. These tests show that uranium can be effectively removed by ion exchange. However, the resin has a very low capacity for uranium.

Test 28

To complete the data package for the anion exchange resin, the column test was repeated with the anion exchange resin regenerated in the carbonate form (with ammonium carbonate). Effluent samples were collected after passing 150, 300 and 450 mL of spent solvent through the column. Test results showed the samples to contain 60.2, 76.5 and 108 ppm respectively. This test was repeated, except that the solution pH was dropped to <2.0 with sulfuric acid. The sample was filtered to remove the precipitate prior ion exchange. A sample of spent solvent was

collected after passing 300 mL of solution through the column. Samples of the original solvent, solvent after filtration and solvent after ion exchange were submitted for uranium analysis. Test results showed the samples to contain 3390 ppm, 1041 ppm and 133 ppm uranium respectively.

Test 29

The next test was run to determine the uranium removal efficiency of anion exchange, in the hydroxide form, after pretreating the solvent to remove EDTA at the low pH followed by high pH adjustment to remove iron and some of the uranium. The spent solvent was first pH adjusted to <2.0 with sulfuric acid. After all the carbonate was expelled and all precipitation of EDTA had ceased, the solution was filtered, then pH adjusted to 12.5 with sodium hydroxide. The brown precipitate that formed was filtered off and the solution was passed through the anion exchange column regenerated in the hydroxide form. A sample of column effluent was taken after processing 200 mL of solvent. A sample of filtered spent solvent was also taken for uranium and iron analysis. The filtered spent solvent was found to contain 648 ppm uranium and 148 ppm iron as Fe. The column effluent sample contained 133 ppm uranium. Although the carbonate was removed from the spent solvent the anion content was apparently still excessive due to the sulfate contribution from the addition of sulfuric acid.

3.3.3.3 Uranium Selective Resins

All testing with the spent EDTA solvent had shown that precipitation was not effective in reducing uranium content to less than 3.0 ppm and that conventional ion exchange resins are not economically feasible due to their low exchange capacity for uranium. Ion exchange was shown to be effective in removing uranium but the high dissolved ion content of the solvent rapidly depletes the resin capacity. At this point a number of resin vendors were contacted to determine the availability of resins which have a high selectivity for uranium. A number of uranium selective resin samples were received and evaluated in this test program.

Table 6 contains a summary of the tests performed using uranium selective ion exchange resins to treat the spent 200°F EDTA/ammonium carbonate solution. Each of these tests is discussed in detail in the following.

Test 30

A sample of Dowex 21K anion resin was evaluated in this program. The resin was received in the chloride form; however, per the manufacturer's recommendation, it was double regenerated with sodium hydroxide at the level of 4 pounds sodium hydroxide per cubic foot per regeneration before testing. The resin was regenerated in the standard 1 inch diameter by 12 inch long column. The column was thoroughly rinsed with demineralized water prior to testing. Flow rate was again set between 8 and 15 mL per minute. Per the vendor recommendation, the solvent pH was reduced to <2.0 with sulfuric acid prior to flow through the column. Samples were collected from the column effluent after passing 200 and 500 mL of solvent through the column. Test results showed 336 ppm and 1815 ppm uranium in the effluent samples respectively. This was well below the target removal level.

Table 6
Results - Treatment of Spent 200°F EDTA/Ammonium Carbonate Solutions
By Uranium Selective Ion Exchange Resins

Starting Solvent: 3455 ppm Dissolved Uranium - pH 9.0

Test #	pH Adjustment	Resin Type	Resin Form	Dissolved Uranium (ppm)	Removal Efficiency (%)
30	<2.0 with H ₂ SO ₄	Dowex 21K Anion	Hydroxide	336	90.3
31	None	Dowex M41 Anion	Hydroxide	1437	58.4
32	<2.0 with H ₂ SO ₄	Amberlite IRC 718 Cation	Hydrogen	21	99.4
33	None	Amberlite A-27 Anion	Hydroxide	123	96.4
34	None	Bayer TP-207 Cation	Hydrogen	3.9	99.9
35	<2.0 with H ₂ SO ₄	Purolite S-940 Cation	Hydrogen	<3.0	>99.9
36	<2.0 with H ₂ SO ₄	Purolite S-940 Cation	Hydrogen	<3.0	>99.9
37	<2.0 with H ₂ SO ₄	Purolite S-940 Cation	Hydrogen	<3.0	>99.9

Test 31

A sample of Dowex M 41 anion resin was also tested in the same manner as Test 30. This resin was also received in the chloride form and was double regenerated using sodium hydroxide at the level of 4 pounds per cubic foot (per the manufacturer's instructions). The solvent was passed through the ion exchange column at the original pH (9.0 - 9.4). Samples of column effluent were collected after processing 200 and 500 mL of solvent. Uranium analysis showed the samples to contain 1437 ppm and 3809 ppm uranium respectively.

Test 32

Rohm and Haas Amberlite IRC 718 cation resin was evaluated in this test program. The resin sample was received in the sodium form but was double regenerated in the acid form using sulfuric acid at a level of 6 pounds acid per cubic foot of resin (per the manufacturer's instructions). The acid was thoroughly rinsed from the column to a neutral end point as determined by pH paper. The spent solvent to be treated was first reduced to a pH of <2.0 with sulfuric acid and the precipitate filtered off. Solvent flow rate through the column was set at 8 - 15 mL per minute. Effluent samples were collected after processing 200 mL and 500 mL of solvent. Uranium analysis showed 21 ppm and 293 ppm uranium respectively. This was an improvement but still well below the target removal efficiency.

Test 33

A Rohm and Haas anion resin designated as Amberlite A-27 was evaluated in this test program. This resin was received in the chloride form. Per the manufacturer's recommendation the resin was first regenerated into the carbonate form and then regenerated into the hydroxide form. After thoroughly rinsing the resin, the spent solvent was passed through the column at the original pH of about 9.0. Column effluent samples were collected after passing 200 mL and 500 mL of solvent through the column. Test results showed 123 ppm and 108 ppm uranium in the samples respectively.

Test 34

A cation resin sample from Bayer Lewatit designated as TP 207 was tested in this program. This resin was received in the sodium form. It was converted to the hydrogen form, per the manufacturer's instructions, using sulfuric acid at the rate of 6 pounds acid per cubic foot of resin. The spent EDTA solvent was pH adjusted to <2.0 to prevent outgas formation in the resin column. The effluent stream was sampled after passing 200, 500, 750 and 1,000 mL of solvent through the column. Uranium test results showed the samples to contain 3.9, 43.5, 96.1 and 31.1 ppm uranium respectively. Test results for the 200 mL sample were approximately equivalent to the results of the conventional type I resin previously tested and were not acceptable.

Test 35

The final set of tests run in this program were performed using a Purolite cation resin designated as S-940. This resin is classified as a chelating type with aminophosphonic acid functional groups. The resin was shipped in the sodium form. Per the manufacturer's instructions for this use, it was converted to the hydrogen form prior to testing. The resin regeneration level was 6 pounds of sulfuric acid per cubic foot. The spent solvent was first pH adjusted to <2.0 with sulfuric acid and the precipitate filtered. Solvent flow through the ion exchange column was established at 8 - 15 ml per minute. Samples of the resin column effluent were collected after passing 500 and 750 mL of solvent through the column. Uranium analysis showed the samples to contain 1.55 ppm and 205 ppm respectively. The 500 mL sample

indicated that this resin had the capability of greater than 99.9% uranium removal efficiency with an associated reasonable capacity.

Tests 36 and 37

Test 35 was repeated two more times. During Test 36 samples were again collected after 500 mL and 750 mL of solvent throughput. Uranium test results were 0.8 ppm and 313 ppm respectively. Test 35 verified the results from Test 34.

For the last test run (Test 36) samples were collected after processing 200 mL and 500 mL of solvent. Uranium test results were <0.3 ppm and 1.08 ppm respectively. Results of this set of tests basically show that uranium is just beginning to break through the resin column at a volume of 500 mL. At this volume of solvent throughput, and assuming a uranium valence of 3, the exchange capacity of the resin for uranium would be 11.5 kilograins per cubic foot. This capacity for uranium is considered excellent. This is based on the fact that the average strong acid cation exchange resin regenerated at a level of 6 pounds acid per cubic foot has an exchange capacity of 14 to 17 kilograins per cubic foot, depending on the calcium to magnesium ratio of the solution.

3.3.4 Summary - Solvent Recycle with EDTA - Carbonate Solvent

Recycle of the spent EDTA - ammonium carbonate solution proved much more difficult than the recycle of the carbonate only solvent. Precipitation techniques were not successful in achieving the desired removal efficiency so that the solvent could be effectively reused. Conventional ion exchange techniques were successful in achieving greater than 99.9% removal efficiency; however, resin usage was considered excessive.

Purolite S-940 cation resin in the hydrogen form was found to achieve the desired removal efficiency of greater than 99.9%. This uranium selective resin also provided an adequate exchange capacity. Since this is a cation exchange resin, it is necessary to first drop the solvent pH to expel the carbonate prior to passing the solvent through the ion exchange column. As discussed in Section 3.2.3, gas blockage of the column occurs if the solvent is passed at its as-received pH of about 9.0.

When using an EDTA/carbonate solvent that has experienced a high temperature exposure (minimum of two hours at 200°F) reducing the pH to less than 2.0 results in precipitation of EDTA. This operation, in conjunction with ion exchange using the Purolite S-940 resin, allows for recycle of the spent solvent.

4.0 CONCLUSIONS

The ammonium carbonate solvent recycle test program resulted in five successful methods for removing uranium from the spent ammonium carbonate solvent with an efficiency of greater than 99.9%. The five successful recycle methods were:

1. acidification to pH 1.0 followed by the use of lime to a pH of 9.8 then caustic to a pH of 12.5
2. solvent pH reduction to 1.0 followed by the application of strong acid cation resin regenerated in the hydrogen form
3. application of strong base anion resin regenerated in the carbonate form at the existing solvent pH
4. application of strong base anion resin regenerated in the hydroxide form at the existing solvent pH
5. a precipitation technique involving application of trisodium phosphate after reducing the solvent pH to 2.0.

The treatment process using anion resin in the carbonate form appeared to have the best merit for recovery applications because it removes only uranium from solution. All other processes were acceptable and considered equal.

Only one method was found that could be used with the EDTA/ammonium carbonate decontamination solvent. This method consisted of reducing the solvent pH to less than 2.0 followed by the application of a uranium selective cation resin (Purolite S-940) in the hydrogen form. When this technique was applied to the solvent that had been heated to 200°F during the decontamination process, greater than 99.9% dissolved uranium removal was achieved along with the separation of the EDTA.

5.0 RECOMMENDATIONS

If a carbonate only solution is used for decontamination, it is recommended to recycle the solvent with a strong base anion resin regenerated in the carbonate form. If an EDTA/carbonate solution is used for decontamination it is recommended to recycle the solvent by first adjusting pH to less than 2.0 and subsequently treating the solution with the Purolite S-940 cation resin regenerated in the hydrogen form.

If a combination of the two solvents is used, it is recommended to recycle the solvents by first adjusting pH to less than 2.0 and subsequently treating the solution with the Purolite S-940 resin. Based on the data generated in this program, this technique will be successful in obtaining greater than 99.9% uranium removal with either solvent system.

6.0 REFERENCES

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