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# PURIFICATION OF $^{238}PuO_2$ SCRAP FOR HEAT SOURCE FUEL.

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## ABSTRACT

The Nuclear Materials Technology (NMT) Division of Los Alamos National Laboratory has initiated a development program to recover & purify plutonium-238 oxide from impure feed sources in a glove box environment. A glove box line has been designed and a chemistry flowsheet developed to perform this recovery task at large scale. The initial demonstration effort focused on purification of $^{238}PuO_2$ fuel by $HNO_3$/HF dissolution, followed by plutonium(III) oxalate precipitation and calcination to an oxide. Decontamination factors for most impurities of concern in the fuel were very good, producing $^{238}PuO_2$ fuel significantly better in purity than specified by General Purpose Heat Source (GPHS) fuel powder specifications. A sufficient quantity of purified $^{238}PuO_2$ fuel was recovered from the process to allow fabrication of a GPHS unit for testing. The results are encouraging for recycle of relatively impure plutonium-238 oxide and scrap residue items into fuel for useful applications. The high specific activity of plutonium-238 magnifies the consequences and concerns of radioactive waste generation. This work places an emphasis on development of waste minimization technologies to complement the aqueous processing operation. Results from experiments on neutralized solutions of plutonium-238 resulted in decontamination to about 1 millicurie/L. Combining ultrafiltration treatment with addition of a water-soluble polymer designed to coordinate Pu, allowed solutions to be decontaminated to about 1 microcurie/L. Efforts continue to develop a capability for efficient, safe, cost-effective, and environmentally acceptable methods to recover and purify $^{238}PuO_2$ fuel.

## INTRODUCTION

Plutonium-238 is an excellent radioisotope for heat source applications in space because of availability, power density, useful lifetime, minimal shielding requirements, and oxide stability. General Purpose Heat Source (GPHS) units containing ~150 g $^{238}PuO_2$ are used in Radioisotope Thermoelectric Generator (RTG) units such as those used to provide electrical power for the Cassini mission to Saturn. Smaller amounts of $^{238}PuO_2$ fuel, typically ~2.7 g, are also used in Radioisotopic Heater Units (RHU) employed recently in Cassini and on the Mars Pathfinder Rover to keep critical components warm in space.

The United States is not presently producing new quantities of plutonium-238 isotope in reactors. One possible domestic feedstock of this nuclear fuel for heat source needs in the immediate future is the existing quantity of relatively impure plutonium-238 isotope residing in scrap and residues. Chemical separations resulting in recovery of high-purity plutonium-238 oxides from this feedsource could be used to produce a significant quantity of GPHS and RTG units. The PF-4 glove box facility operated by Nuclear Materials Technology (NMT) Division has been utilized for high-purity recovery of the weapons isotope, plutonium-239, from a wide variety of scrap materials. This report describes a portion of our efforts to demonstrate technologies for the efficient, safe, cost-effective, and environmentally acceptable recovery and purification of $^{238}PuO_2$ fuel.

## EXPERIMENTAL

The plutonium-238 oxide fuel used as feed for this study was fired at temperatures up to 1600 °C. Dissolution of plutonium oxide fired to this temperatures is slow and often incomplete in refluxing nitric acid (Cleveland 1979). Addition of a small amount of hydrofluoric acid increases the rate of dissolution, but this rate was too slow for our

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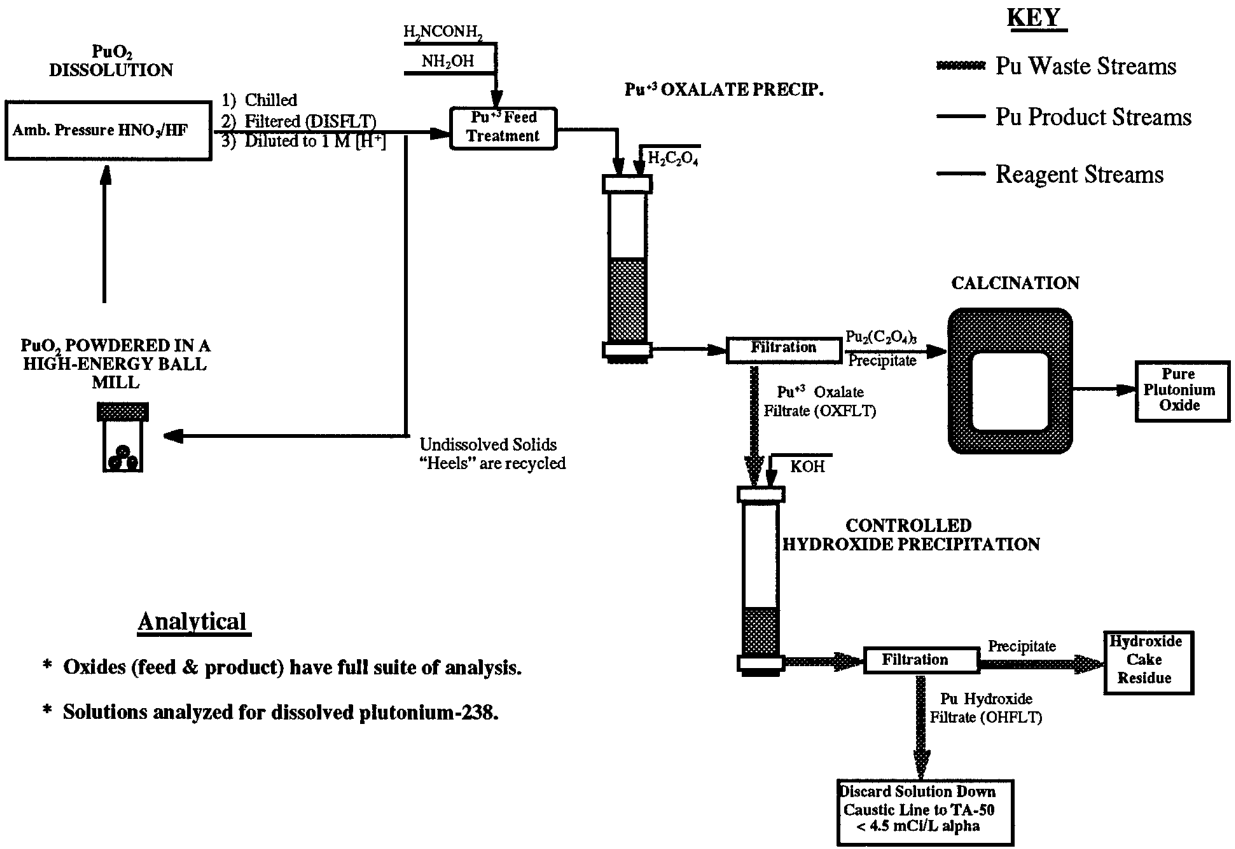
purposes. Better dissolution results were obtained for $^{238}PuO_2$ feed treated in a high-energy ball mill. Particle size analysis of the milled $^{238}PuO_2$ feed indicated that an average particle size of less than 1 micron could be obtained. The milled oxide was stirred in a mixture of nitric and hydrofluoric acids refluxing at ambient pressure. This provided good dissolution efficiency after 7-8 hours at reflux. Several dissolutions at 65 g scale were accomplished. Reagent grade chemicals and distilled or deionized water were used to minimize introduction of impurities. Teflon or polypropylene apparatus was used for handling $^{238}Pu$ solutions to avoid leaching silicates from glassware. A flowsheet illustrating the process is shown in Figure 1.



**Analytical**

- **Oxides (feed & product) have full suite of analysis.**
- **Solutions analyzed for dissolved plutonium-238.**

FIGURE 1. Current Plutonium-238 Aqueous Process Flow Diagram.

Hydroxylamine nitrate was used as the primary reducing agent, and with the exception of one run, either sulfamic acid or urea as a holding agent. All steps were performed as rapidly as possible to minimize the effects of radiolysis. For this study, recovered plutonium(III) oxalate was air-dried and then heated to dull red heat in a stainless steel, slip-top can on a hot plate for about 60 minutes to convert the plutonium to an oxide. Heating for longer times produced no significant weight loss.

## RESULTS AND DISCUSSION

Plutonium (III) oxalate precipitation was used in this demonstration because of it's simplicity, speed and adequacy of purification. In particular, plutonium (III) oxalate precipitation requires no temperature control, shows little detrimental effect from excess oxalic acid, and has a rapid reaction, precipitation, and filtration time. The large amount of radiolysis associated with plutonium-238 complicates control of the Pu oxidation state in nitric acid. In general, the radiolysis of concentrated nitric acid produces an oxidizing environment, and a larger proportion of Pu(VI) (Katz 1986). The presence of radiation-generated oxidizing agents in nitric acid increases the quantity of reducing agents required to effect complete reduction to Pu(III). Others have observed that $^{238}Pu$ is best reduced to Pu(III) by combinations of "holding reagents" and "reducing agents" (Greinetz 1980 and Burney 1984). Holding agents react with nitrite and other oxidizing species, slowing the rate of Pu(III) oxidation, while the reducing agent rapidly reduces Pu(VI) and Pu(IV) to Pu(III).

Results were obtained on plutonium-238 oxide feed and purified products described in this study. These included measurement of actinide and non-actinide impurities in plutonium-238 oxide. Other results were obtained for the various process solutions based on radiochemical measurements of the plutonium-238 content.

Non-Actinide Element Decontamination:

The first row in Table 1 lists the element limits in µg/g that are specified in the GPHS standard (Los Alamos 1996). Three different sources of plutonium-238 oxide fuel, as designated by Feed #1, Feed #2, and Feed #3, were used in the experiments. Table 1 shows analytical results for element impurities (µg/g) in the feed materials and plutonium oxide products produced by oxalate precipitation. The numbers in boldface type in Table 1 represent a value from an average of duplicate analysis for the impurities. Italicized numbers indicate analytical results that are at or below the indicated detection limits.

Good decontamination was achieved for silicon, chromium, iron and nickel, with mixed results for aluminum. The column entitled Sum non-Actinides is the summation of all elemental impurities listed above the detection limit. Entries marked "*" in the Sum non-Actinides column indicate an indeterminate value, typically because one element exceeded the range of the measurement method. Product # 1A, for example, was an experiment in which hydroxylamine nitrate was added as a reducing agent, but no holding reagent was included. A poor precipitation occurred leaving a highly colored solution. NaOH was added to force more plutonium to precipitate, resulting in a high Na content in Product # 1A in this preliminary experiment.

TABLE 1. Non-Actinide Impurities (µg/g) in Plutonium-238 Oxide Fuel.

| | Al | B | Be | Ca | Cd | Cr | Cu | Fe | Mg | Mn | Mo | Na | Ni | Pb | P | Si | Sn | Zn | Sum non-Actinides |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| GPHS Rev A | 500 | 5 | 5 | 300 | 50 | 500 | 200 | 800 | 100 | 50 | 250 | 250 | 500 | 100 | 25 | 750 | 50 | 50 | 2550 |
| Feed # 1 | **145** | *5* | *1* | **9** | *10* | **390** | *1* | **830** | **18** | **13** | *20* | *50* | **120** | **20** | **710** | **560** | *5* | *5* | **2203** |
| Pdt. # 1A | **200** | *5* | *1* | **3.5** | *10* | **285** | *1* | **335** | *10* | **9** | *20* | * | **5** | *10* | *20* | *5* | *5* | *5* | * |
| Pdt. # 1B | **34** | *5* | *1* | **65** | *10* | **19** | *1* | **67** | *10* | **2** | *20* | *50* | **9** | *10* | *20* | **53** | *5* | *5* | **377** |
| Feed # 2 | **155** | *5* | *1* | **95** | *10* | **820** | **13** | **1500** | *10* | **18** | *20* | *50* | **455** | **20** | *20* | **850** | *5* | **23** | **4051** |
| Pdt. # 2A | **185** | *5* | *3* | **40** | *10* | **15** | **20** | **39** | *10* | **1.5** | *20* | *50* | **8** | *10* | *20* | **7** | *5* | *5* | **436** |
| Pdt. # 2B | **155** | *5* | *3* | **40** | *10* | **19** | **18** | **78** | *10* | **3** | *20* | *50* | **14** | *10* | *20* | *5* | *5* | *5* | **453** |
| Pdt. # 2C | **55** | *5* | 2 | **450** | *10* | **23** | **100** | **62** | *10* | **6** | *20* | *50* | **18** | *10* | *20* | **23** | *5* | *5* | **656** |
| Pdt. # 2D | **62** | *5* | 2 | **>750** | *10* | **30** | **3** | **67** | *10* | **1** | *20* | *50* | **10** | *10* | *20* | **11** | *5* | *5* | * |
| Pdt. # 2E | **225** | *5* | 2 | **>750** | *10* | **56** | **3** | **145** | *10* | **1** | *20* | *50* | **25** | *10* | *20* | **91** | *5* | *5* | * |
| Feed #3 | **155** | *5* | *1* | **7** | *10* | **540** | **100** | **555** | *10* | **10** | *20* | *63* | **99** | *10* | | **515** | *5* | *15* | **2121** |
| Pdt. # 3A | **176** | *5* | *1* | **350** | *10* | **194** | **75** | **52** | *10* | **10** | *20* | *50* | **17** | *10* | | **32** | *5* | *5* | **1024** |
| Pdt. # 3B | **110** | *5* | *1* | **500** | *10* | **25** | **50** | **54** | *10* | **8** | *20* | *50* | **19** | **13** | | **23** | *5* | *15* | **919** |
| Pdt. # 3C | **5** | *5* | *1* | **300** | *10* | **17** | **50** | **32** | *10* | *1* | *20* | *50* | **14** | **15** | | **12** | *5* | *5* | **553** |
| Pdt. # 3D | **144** | *5* | *1* | **300** | *10* | **220** | **50** | **255** | *10* | **6** | *20* | *50* | **97** | **25** | | **13** | *5* | *5* | **1217** |

Only in the case of calcium has there been noted an apparent increase in the amount of a measured impurity in the $PuO_2$ fuel. It is anticipated that calcium would coprecipitate with plutonium under the conditions of the plutonium(III) oxalate precipitation process (Cleveland 1979). High-purity water, acids and other reagents were used in this demonstration specifically to avoid the possibility of introducing contaminants such as Ca. In spite of these precautions, an increase in Ca was observed in some runs while not in others. Separate analysis of reagents used was instituted in a search for a possible source of calcium contamination, with results to date for all reagents and water below detection limits. Pu can act as an interferent with Ca measurements in the DC arc analysis technique used on the $PuO_2$ samples. Work is continuing to determine if the calcium data are significant or a measurement artifact.

## Actinide Element Decontamination:

The first row in Table 2 lists the actinide impurities in µg/g as specified in the GPHS standard. The numbers in boldface type represent values (µg/g) observed for the actinide impurities. Those numbers presented in italics indicate results that are at or below the indicated detection limits. $^{234}$U was by far the largest actinide impurity observed in the feed material because it is the daughter product of $^{238}$Pu by alpha decay. Good decontamination was achieved for U. Np, Am and Th showed little or no decontamination from Pu under the conditions of plutonium (III) oxalate precipitation. As these actinides are well below the GPHS standard in this feed, they were not a major concern. Analysis for actinide impurities of several more $^{238}PuO_2$ product samples, including those originating from Feed # 3, is pending.

TABLE 2. Actinide Impurities (µg/g) in Plutonium-238 Fuel.

| Sample | Am-241 | Np-237 | Pu-236 | U-234[a] | Th | Sum of Actinides |
|---|---|---|---|---|---|---|
| GPHS Rev A | 5000 | 5000 | 2 | 5000 | 5000 | 10,000 |
| Feed # 1 | **350** | **978** | *1* | **14000** | **1600** | 16929 |
| Pdt. # 1A | **331** | **936** | *1* | **1900** | **1600** | 4768 |
| Pdt. # 1B | **340** | **237** | *1* | **60** | **1700** | 2338 |
| Feed # 2 | **135** | **637** | *1* | **7000** | **530** | 8303 |
| Pdt. # 2A | **57** | **891** | *1* | **60** | **560** | 1569 |
| Pdt. # 2B | **91** | **603** | *1* | **70** | **530** | 1295 |
| Pdt. # 2C | **132** | **517** | *1* | **130** | **350** | 1130 |
| Pdt. # 2D | **136** | **577** | *1* | **280** | **410** | 1404 |
| Pdt. # 2E | **108** | **315** | *1* | **0** | **290** | 714 |

a. Values listed in the table for U-234 and Am-241 are decay corrected for the time interval between precipitation and analysis. The rapid ingrowth of U-234 adds to the uncertainty in these values.

## Process Effluents Solution Data:

TABLE 3. Data for Plutonium-238 Oxalate Filtrate Solutions.

| Sample | Volume (L) | [H+] (M) | $^{238}$Pu (g/L) | alpha (mCi/L) | Total Pu (g/L) | $^{238}PuO_2$ Recovered (g) | Soln. Pu % Recovered |
|---|---|---|---|---|---|---|---|
| OXFLT-1A | 1.22 | 0.57 | 3.26E-02 | 558.01 | 4.09E-02 | 9.03 | 99.38 |
| OXFLT-1B | 1.05 | 1.04 | 2.87E-01 | 4912.58 | 3.60E-01 | 7.79 | 94.78 |
| OXFLT-2A | 1.90 | 1.33 | 2.51E+00 | 42963.67 | 3.05E+00 | 16.99 | 72.08 |
| OXFLT-2B | 1.74 | 1.39 | 6.28E-01 | 10749.48 | 7.64E-01 | 9.40 | 86.18 |
| OXFLT-2C | 1.90 | 1.14 | 1.72E-01 | 2944.12 | 2.09E-01 | 28.87 | 98.46 |
| OXFLT-2D | 1.90 | 1.14 | 1.73E-01 | 2961.24 | 2.10E-01 | 27.68 | 98.39 |
| OXFLT-2E | 0.86 | 1.19 | 2.25E-01 | 3851.33 | 2.74E-01 | 4.97 | 94.90 |
| OXFLT-3A | 2.80 | 0.98 | 1.64E-01 | 2810.00 | 2.00E-01 | 14.35 | 95.76 |
| OXFLT-3B | 2.35 | 1.10 | 5.74E-02 | 982.00 | 7.00E-02 | 13.36 | 98.62 |
| OXFLT-3C | 3.24 | 0.92 | 4.30E-02 | 736.00 | 5.24E-02 | 27.63 | 99.31 |
| OXFLT-3D | 3.00 | 1.05 | 4.76E-02 | 814.00 | 5.80E-02 | 29.04 | 99.33 |
| OXFLT-3E | 2.90 | 0.88 | 1.07E-01 | 1840.00 | 1.31E-01 | 28.78 | 98.52 |
| OXFLT-3F | 2.90 | 1.02 | 9.06E-02 | 1550.00 | 1.10E-01 | 31.81 | 98.87 |

Table 3 lists data for plutonium-238 oxalate filtrate solutions (OXFLT) in this work. An acid-base titration was performed on feed solutions prior to oxalate precipitation to measure and allow adjustment of free acidity in the solution to approximately 1 Molar. Dissolved $^{238}$Pu was adjusted to the trivalent state and precipitated with oxalic acid, followed by a filtration step for recovery. Plutonium-238 in the filtrate was measured radiochemically by gas proportional alpha counting, and converted to a total plutonium value, based on measured plutonium isotopic values for the feed material. The "Soln. Pu % Recovered" data is based on the amount of Pu recovered as oxide in the process and the amount of Pu remaining in the oxalate filtrate solution following filtration. Recovery percentages were generally 95% or greater for all runs with the exceptions of products OXFLT-2A and OXFLT-2B that were recovered from higher molarity acid.

Solutions from the oxalate filtration process were neutralized with KOH, allowed to stand at least overnight and then filtered through Whatman 42 paper. Table 4 lists data for plutonium-238 hydroxide filtrate solutions (OHFLT) in runs where a sample aliquot was immediately removed and diluted in 0.1 M $HNO_3$. The alpha activity observed in hydroxide filtrate solutions after a single filtration ranged from approximately 3–170 millicurie/L. Solutions below the alpha activity limit (4.5 millicurie/L) of the PF-4 caustic drain line are allowed to be transferred to a centralized collection point in the wastewater treatment facility for further decontamination.

The "Soln. Pu % Recovered" data is based on the amount of Pu initially present in the oxalate filtrate and the amount of Pu remaining in the filtrate following the hydroxide precipitation process. The "Overall Soln. Pu % Loss" data shows the total loss of $^{238}$Pu in process effluent streams from the point of dissolution through the sequential precipitation and filtration steps.

TABLE 4. Data for Plutonium-238 Hydroxide Filtrate Solutions Sampled Immediately After Filtration.

| Sample | Volume (L) | Pu-238 (g/L) | alpha (mCi/L) | Total Pu (g/L) | Soln. Pu % Recovered | Overall Soln. Pu % Loss |
|---|---|---|---|---|---|---|
| OHFLT-1A | 1.59 | 3.13E-04 | 5.36 | 3.93E-04 | 98.75 | 0.0078 |
| OHFLT-1B | 1.44 | 4.32E-04 | 7.39 | 5.42E-04 | 99.79 | 0.0108 |
| OHFLT-2A | 2.30 | 1.62E-04 | 2.77 | 1.97E-04 | 99.99 | 0.0022 |
| OHFLT-2B | 2.10 | 8.85E-04 | 15.15 | 1.08E-03 | 99.83 | 0.0235 |
| OHFLT-2C | 2.20 | 8.68E-04 | 14.86 | 1.06E-03 | 99.42 | 0.0090 |
| OHFLT-2D | 2.20 | 4.53E-04 | 7.75 | 5.51E-04 | 99.70 | 0.0049 |
| OHFLT-2E | 1.00 | 7.30E-04 | 12.50 | 8.88E-04 | 99.62 | 0.0192 |
| OHFLT-3A | 4.00 | 1.02E-02 | 173.80 | 1.24E-02 | 91.16 | 0.3749 |
| OHFLT-3B | 4.00 | 1.83E-03 | 31.35 | 2.23E-03 | 94.57 | 0.0748 |
| OHFLT-3C | 4.00 | 7.00E-03 | 119.90 | 8.54E-03 | 79.89 | 0.1393 |
| OHFLT-3D | 4.00 | 6.09E-03 | 104.28 | 7.43E-03 | 82.92 | 0.1153 |

Several of the hydroxide filtrate solutions were allowed to stand for a few weeks and then supernatant solution sampled. Data for these solutions, designated with a W at the end of the Sample name, are presented in Table 5. Alpha activity for solutions sampled in this manner were considerably decreased than the same solutions (OHFLT-3A through OHFLT-3D in Table 4) sampled immediately after filtration. A longer period of time appears to allow additional plutonium to precipitate or settle from the solution.

TABLE 5. Data for Plutonium-238 Hydroxide Filtrate Solution Supernatant Sampled Weeks Later.

| Sample | Volume (L) | Pu-238 (g/L) | alpha (mCi/L) | Actual Pu (g/L) | Soln. Pu % Recovered |
|---|---|---|---|---|---|
| OHFLT-3AW | 4.00 | 5.67E-05 | 0.97 | 6.92E-05 | 99.95 |
| OHFLT-3BW | 4.00 | 5.50E-05 | 0.94 | 6.70E-05 | 99.84 |
| OHFLT-3CW | 4.00 | 6.19E-06 | 0.11 | 7.55E-06 | 99.98 |
| OHFLT-3DW | 4.00 | 1.32E-04 | 2.26 | 1.61E-04 | 99.63 |
| OHFLT-3EW | 4.00 | 8.35E-04 | 14.30 | 1.02E-03 | 98.93 |
| OHFLT-3FW | 4.00 | 4.06E-05 | 0.70 | 4.95E-05 | 99.94 |

The bulk of the effluent solutions generated were combined, treated with additional caustic and iron sulfate, again filtered, and transferred to the wastewater treatment facility when decontamination below the PF-4 caustic drain line alpha limit (4.5 millicurie/L) was achieved.

### Ultrafiltration Solution Data:

Passing small samples of each of the aged OHFLT supernatent solutions through an 0.2 micron filter resulted in no additional decontamination. Samples of supernatent OHFLT-3BW adjusted to different pH values were also passed through an ultrafilter with a 10,000 molecular weight cut-off (~0.008 microns). This also resulted in no significant decontamination. Additional samples of this supernatent solution were treated using Polymer Filtration. This technique employs a water-soluble chelating polymer to bind selected metal ions and then uses an ultrafiltration step to separate the polymer-metal ion complex from the bulk of the solution (Jarvinen 1997). A concentrated solution of a water-soluble polymer containing phosphonic acid chelating groups was added to the supernatent solution to give a 0.2 to 0.4 weight % solution of the polymer. The pH of the solution was adjusted between 4 and 13 and the solution filtered through a 10,000 MW cut-off filter. As shown in Table 6, up to 98% of the remaining alpha activity could be removed at pH 6.5. These preliminary results indicate that two stages of polymer filtration could allow the solutions to be decontaminated to less than 0.5 microcurie/L which is the upper limit for discharging aqueous solutions to the industrial waste line at Los Alamos. The concentrated Pu-238 bound to the polymer could be recovered by acidifying the solution to release the metal and adding a relatively small volume of wash solution to remove the metal by allowing it to pass through the ultrafilter. The polymer solution could then be reused. Another option involves sending the loaded polymer solution directly to a disposal process such as evaporation followed by cementation.

TABLE 6. Removal of Alpha Activity from Hydroxide Filtrate Supernatent OHFLT-3BW by Polymer Filtration

| Sample | wt % polymer | pH | Final alpha activity (mCi/L) | % alpha removal |
|---|---|---|---|---|
| 3BW1 | 0.20 | 4.0 | 1.33E-02 | 44 |
| 3BW2 | 0.40 | 4.0 | 9.50E-03 | 59 |
| 3BW3 | 0.20 | 6.5 | 6.87E-04 | 97 |
| 3BW4 | 0.40 | 6.5 | 5.55E-04 | 98 |
| 3BW5 | 0.20 | 8.5 | 1.06E-03 | 95 |
| 3BW6 | 0.40 | 8.5 | 8.09E-04 | 96 |
| 3BW& | 0.20 | 13.0 | 4.88E-03 | 79 |
| 3BW8 | 0.40 | 13.0 | 2.17E-03 | 91 |

## CONCLUSIONS

NMT Division has the expertise and facilities to make large scale recovery and purification of plutonium-238 for oxide fuel a practical option. Additional supplies of plutonium-238 oxide fuel from many existing sources, including scrap and materials that fail the GPHS standard, can be recovered in a glove box environment. For much of the scrap $^{238}PuO_2$ available as feed, including that with significant $^{234}U$ ingrowth, the plutonium (III) oxalate precipitation procedure is adequate to meet the GPHS fuel standard. Good dissolution efficiencies were observed for high-fired $^{238}PuO_2$ material that was first treated in a high energy ball mill. Overall losses of the $^{238}Pu$ isotope to aqueous effluent streams were small. More that enough purified $^{238}PuO_2$ fuel has been recovered to begin the process for fabrication of a 150g GPHS. The flowsheet shown in Figure 1 will be adapted to allow anion exchange recovery of $^{238}Pu$ from leaner feed residues, andto incorporate additional waste minimization technologies for treatment of process effluent.

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