Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Bluhm, Elizabeth A. , Abney, Kent D. , Balkey, Simon , Brock, Jason C. , Coriz, Fawn , Dyke, James T. , Garcia, Daniel J. , Griego, Brenda J. , Martinez, Benjie T. , Martinez, David , Martinez, Joe Ray , Martinez, Yvonne A. , Morgan, Lonny , Roybal, Judy D. , Valdez, Jose A. , Ramsey, Kevin B. , Bluhm, Brian K. , Martinez, Carl D. and Valdez, Manuelita M.(2005) 'Plutonium Oxide Polishing for MOX Fuel Production', Separation Science and Technology, 40: 1, 281 — 296

To link to this Article: DOI: 10.1081/SS-200042232

URL: http://dx.doi.org/10.1081/SS-200042232

Separation Science and Technology, 40: 281–296, 2005
ISSN 0149-6395 print/1520-5754 online
DOI: 10.1081/SS-200042232

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Abstract: Los Alamos National Laboratory (LANL) has been tasked to produce 120 kg of plutonium as highly purified PuO_2 for the European Mixed Oxide (MOX) Lead Test Assembly managed by Duke, COGEMA, Stone & Webster (DCS). To meet stringent and challenging technical requirements for PuO_2 production, the LANL aqueous polishing team recently established consistency in generating quality material from weapons-grade PuO_2. Polishing was required to remove undesirable quantities of trace-metal impurities, particularly gallium, and to produce a material with appropriate powder characteristics, such as particle size and surface area. The process flow sheet

Funding was provided by the LANL Fissile Materials Disposition Program (NN60). Los Alamos National Laboratory is operated by the University of California under Contract W-7405-ENG-36. The authors would like to thank Patrick Martinez, Randy Drake, and Debbie Dale of Actinide Analytical Chemistry (C-AAC) for their assistance with trace analytical measurements.

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for PuO_2 polishing was based on aqueous purification and included the unit operations of dissolution, ion exchange, oxalate precipitation, and calcination.

INTRODUCTION

A joint agreement between the United States (U.S.) and Russia requires the disposition of excess plutonium from decommissioned nuclear weapons. The proposed method for disposition is manufacturing mixed oxide (MOX) fuel, a plutonium/depleted uranium oxide blend, for generating electricity in nuclear power plants. Under this program, the Department of Energy's (DOE) Office of Fissile Materials Disposition (NA-26), in conjunction with Duke, COGEMA, and Stone & Webster (DCS), will convert approximately 34 metric tons of U.S. surplus weapons-grade plutonium into fuel for commercial power generation.

In late 2001, DOE authorized Los Alamos National Laboratory (LANL) to begin generating polished plutonium dioxide (PuO_2) for subsequent use in MOX Lead Test Assemblies (LTA) to support fuel qualification and licensing by the Nuclear Regulatory Commission (NRC) (1). Polishing impure PuO_2 reduces unwanted trace impurities, particularly gallium and boron that could affect fuel fabrication processes or affect the performance of the cladding or fuel during reactor operation (2). In addition to impurity levels, the polished PuO_2 must meet minimum physical requirements, as described in the ASTM C757-90 industry standard "Specification for Nuclear-Grade Plutonium Dioxide Powder, Sinterable" (3).

In early 2002, LANL demonstrated the ability to purify Pu originating from the Materials Disposition Programs to meet the impurity levels required by MOX–LTA by processing 5 kg PuO_2 during a first demonstration phase of the project. Although the impurity levels at the end of the first demonstration phase met most of the specification limits, there were several elements whose concentration approached their limit, indicating that these elements could present quality control issues during production phase (4). Several problematic elements included Ga, Al, F, C, B, and Si. The first demonstration phase also indicated that analytical chemistry methods, in particular detection limits for Ga and B, needed to be improved. It is not surprising that these sets of elements had higher levels than other impurities in the PuO_2 product. Gallium is initially high in the Pu feed material, around 3120 $\mu g/g$ (Pu) (see Table 1). Aluminum and F are reagents added during polishing of the PuO_2. Carbon results from incomplete calcination (the final processing step). Boron and Si are potential contaminants that can leach from the glass processing vessels.

In addition to impurity levels, two other processing issues were identified during the first demonstration phase (4). Dissolution efficiencies for the batch HNO_3/HF dissolutions were around 55–65%, requiring several passes to achieve approximately 75–80% total PuO_2 dissolution. Second, the

Table 1. ARIES feed composition for ATL10–36 vs. proposed maximum specification limits

Element	ARIES average composition (μg/g Pu)	Maximum specification limit (6) (μg/g Pu)
Aluminum	77	150
Bismuth	0.8	100
Boron	12	10*
Cadmium	1.5	10*
Calcium	30	500
Carbon	160	500
Chloride	40	250 (F + Cl)
Chromium	43	100
Cobalt	2.9	100
Copper	19	100
Dysprosium	0.4	1
Europium	0.4	1
Fluoride	30	250 (F + Cl)
Gadolinium	0.42	3
Gallium	3120	200
Indium	2.9	20
Iron	188	500
Lead	8.5	200
Lithium	10	100
Magnesium	10	500
Manganese	7.9	100
Molybdenum	5.1	100
Nickel	99	200
Niobium	0.67	100
Nitrogen	40	300
Phosphorus	56	100
Potassium	94	100
Samarium	0.5	2
Silicon	73	200
Silver	0.8	100
Sodium	29	300
Sulfur	30	250
Thorium	1.2	100
Tin	4.4	100
Titanium	7.9	100
Tungsten	11	200
Uranium	527	5,000
Vanadium	6	300
Zinc	9.2	100

*ASTM C757-90 suggests 3 μg/g for B, Cd, and Gd.

calcination process was experiencing a temperature spike during the initial heating step, which resulted in exposing the product material to temperatures in excess of 670°C, where 670°C was the maximum allowable temperature.

Realizing that there was some remaining technical risk, DOE authorized LANL to continue with a second demonstration phase (5 kg) and begin production of an additional 110 kg of PuO_2. The challenge to the aqueous processing team was to identify process improvements based on "best technical judgment" and to implement these improvements without jeopardizing the quality of the PuO_2 product. While it would have been preferable to perform parametric studies of the identified improvements, production timelines did not allow for such detailed studies; therefore, the process improvements had to be evaluated during the performance of production runs. This paper discusses the implementation of these process improvements during the second demonstration (5 kg) and continuing into the first half of the 110 kg production phase. In some cases, there were clear indications that the process change resulted in improvement in a quality parameter measured on the PuO_2 product. In others, a throughput improvement was implemented with no apparent decrease in product quality.

BASELINE PROCESS

The PuO_2 polishing flow sheet was based on aqueous purification in a nitric acid system and included the unit operations of dissolution, ion exchange, oxalate precipitation, and calcination. Figure 1 shows the aqueous polishing

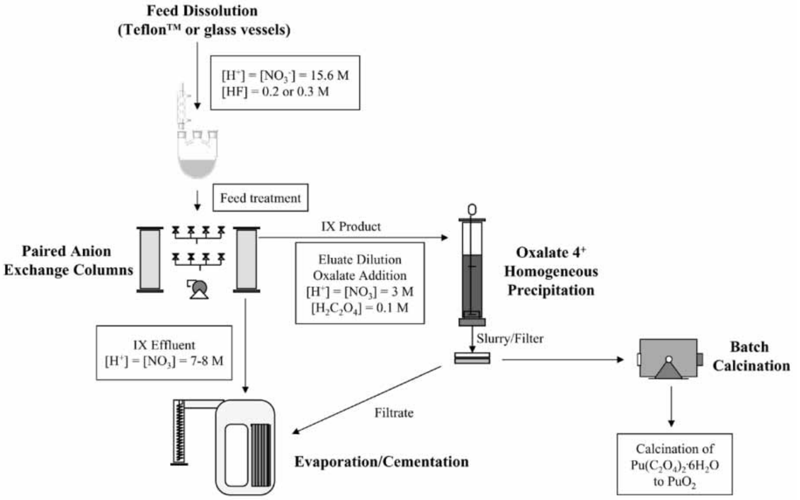

Figure 1.

flow sheet, including the dry operations and waste treatment and disposal. All aqueous processing was performed within the Advanced Testing Line for Actinide Separations (ATLAS) (5). ATLAS (ATL) batch numbers were assigned for consecutive runs. The second demonstration phase consisted of batches ATL10-13, while production runs cover ATL14-36.

Feed Material

Within the Plutonium Facility at LANL, the Advanced Recovery and Integrated Extraction System (ARIES) process line provided the feed material which consisted of impure, weapons-grade PuO_2. ARIES is the prototypic process that will be used in the Pit Disassembly Conversion Facility (PDCF) for providing impure PuO_2 feed to the MOX Fuel Fabrication Facility (MFFF), with both facilities being constructed at the Savannah River Site. ARIES converts plutonium metal to oxide at elevated temperatures resulting in a PuO_2, which is approximately 87% Pu; however, it does not meet the preliminary impurity specifications for the MOX-LTA project and required further polishing. Table 1 provides a listing of the average impurity content for a typical blend of ARIES PuO_2 and the preliminary impurity levels required for MOX fuel (6). These trace impurity levels set a benchmark for LANL aqueous polishing operations and PuO_2 product quality. To err on the conservative side, boron was benchmarked against the ASTM C757-90 standard (3) of 3 μg/g rather than the10 μg/g outlined by Chidester et al. (6).

Dissolution

The ARIES PuO_2 was dissolved in 5.0 L borosilicate glass vessels using heating mantels with digital controllers and thermocouples. Dissolution was performed in 15.6 M reagent grade nitric acid (HNO_3) and 0.2 M hydrofluoric acid (HF), followed by refluxing at a nominal 110°C for several hours. Solutions were filtered, and the remaining heels were redissolved until achieving ~75–80% total dissolution efficiency.

Anion Exchange

The dissolved Pu was separated from impurities using a nitric acid anion exchange process using Reillex-HPQ™ resin (7). The Pu-rich feed solution was adjusted to 7 M HNO_3 using 0.35 M HNO_3 to optimize the sorption of Pu on the resin as $Pu(NO_3)_6^{2-}$. An equimolar amount of aluminum nitrate was added to complex F (used in dissolution). Hydrogen peroxide was also added to ensure complete valence adjustment to the (IV) oxidation state (8).

The resin bed was charged with 7 M HNO_3 after which the treated Pu feed was loaded at the top of the column. Once the feed solution was completely loaded, the resin bed was washed with 7 M HNO_3. For Pu elution, 0.35 M HNO_3 was added to the resin bed and allowed to sit overnight in order to ensure complete desorption of the Pu from the resin. On the following day, elution continued with additional 0.35 M HNO_3 until the Pu was recovered from the resin.

Oxalate Precipitation

Plutonium(IV) oxalate precipitation was performed to further purify and convert the plutonium eluate solution into a solid oxalate cake. The Pu(IV) precipitation has advantages over the Pu(III) oxalate precipitation in that it provides better separation of Pu from U, Fe, and Al and gives optimal particle morphology for the final PuO_2 product (9). The anion exchange eluate was adjusted to 2.5 M HNO_3, gradually warmed to a nominal 65°C temperature while being continuously mixed. Oxalic acid was slowly added to precipitate a brownish-colored Pu(IV) oxalate complex. The oxalate material was allowed to settle, followed by vacuum filtration and washing with dilute oxalic acid in 2.0 M HNO_3. Each batch was air dried under reduced pressure resulting in a damp cake that was transferred to calcination operations.

Calcination

To convert the Pu(IV) oxalate to PuO_2, the oxalate cake was calcined in a fused silica boat placed in a muffle furnace. In general, the furnace controller was programmed for a 150°C/hour ramp rate to 650°C and then maintained at temperature for a minimum of 4 h. Bed temperature was measured by three different thermocouples located within the oxalate cake itself and remained in full contact during the entire calcination process. All calcinations were performed in ambient dry air (~0.032 kPa water) glovebox atmosphere.

Analytical Characterization

All trace analytical analyses were performed by the Chemistry-Actinide Analytical Chemistry (C-AAC) group at LANL. Oxide samples for analysis are prepared by dissolving in either a HNO_3/HF mixture or *aqua regia.* Aluminum and Si content were determined by Inductively Coupled Plasma—Atomic Emission Spectrometry (ICP-AES), B and Ga by Inductively Coupled Plasma—Mass Spectrometry (ICP-MS), and C by oxidizing the carbon to CO_2 at 1200°C in a furnace followed by infrared analysis of the off-gas to quantitate CO_2 generation. Chlorine and F were extracted from the PuO_2 by pyrohydrolysis and elemental concentration was determined by using ion chromatography.

Particle size, specific surface area, and moisture content of product PuO_2 for batches ATL10-36 were determined in order to qualify process modifications to the calcination process. Particle size was determined by using a Coulter Counter (Multisizer3, Beckman Coulter, Miami, FL) and surface area with a 6-point BET (Brunauer, Emmett, Teller) method on a NOVA 3000 (Quantachrome, Boynton Beach, Florida) surface area analyzer. Moisture content was determined by the Loss-on-Ignition method. The sample was initially weighed, then placed into a furnace and slowly heated to 1000°C. After heating, the sample was cooled and reweighed to determine if weight loss occurred. For pure oxides, a weight loss indicates water removal; however, if salts or other volatile compounds are present, the weight loss could also be due to these constituents.

PROCESS IMPROVEMENTS

Following the first demonstration phase and during the processing of both the second demonstration phase and the production runs, several process changes were implemented. There were two driving factors in designing a process change: (a) reduction of product impurity levels and (b) improved throughput in order to meet production schedules.

Washing Procedures—Anion Exchange

It was found during the first demonstration phase that to meet Ga removal specifications, a significantly large wash volume was required during the anion exchange process. We are currently using a minimum of 500 L of 7 M HNO_3 to remove impurities from the Pu loaded column. Five independent tests (ATL5, 7–9, 17) were conducted to analyze Ga content during typical ion exchange runs to ensure complete removal from the resin bed during the load and wash process. All effluent profiles display similar behavior. Figure 2 illustrates a typical example of Ga concentration ($\mu g/g$ Pu) in the wash effluent for production run ATL17. Results of analyzing effluent volumes at 25 L intervals for Ga concentration generated the profile shown in Fig. 2. The first Ga elution peak rises to nearly 250 ppm as the feed solution displaces from the column, then decreases to approximately 6 ppm by 300 L and to 0.4 ppm by 400 L. After 600 L, a rise in the Ga concentration was observed from presumably the release of entrapped Ga from the resin bed due to the loading of the 0.35 M HNO_3 strip solution. This second elution peak occurs closely to the Pu elution band and could potentially cause

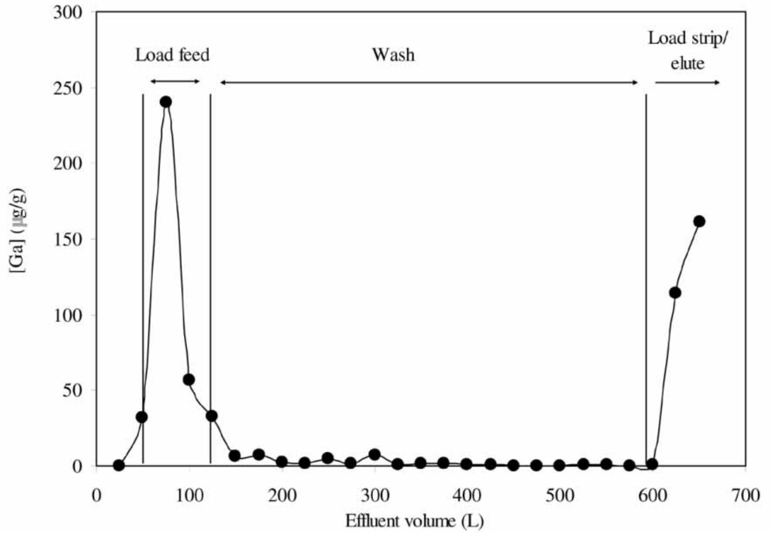

Figure 2.

a cross-contamination problem with the clean eluate. However, the oxalate precipitation process should remove any Ga that bleeds into the clean Pu fraction.

Washing Procedures—Precipitation

Following precipitation, the oxalate cake was washed, under vacuum, with dilute oxalic acid in 2.0 M HNO_3. Beginning with the production runs, a new washing procedure was implemented because trace analysis continued to show scattered and inconsistent (0.33–0.70 ppm) Ga values (Table 2). It was also determined earlier that the anion exchange and oxalate precipitation steps were efficiently removing most Ga. Therefore, the wash procedure was then modified such that for each addition of wash solution, vacuum was disconnected to allow intimate mixing between the oxalate cake and wash while continuously stirring with a spatula. The total wash was a least three times the recovered cake volume. As a result, Ga values declined dramatically from approximately 0.34 ppm Ga to 0.02 ppm and stayed relatively stable all the way through ATL36 (Table 2). Figure 3 shows a plot of Ga concentration (µg/g Pu) vs. ATL batch number to demonstrate process consistency in washing. Note that the Si values also decreased following the implementation of the new wash procedure from approximately 180 ppm to 60 ppm.

Table 2. Problematic impurity concentrations (μg/g Pu) for the second 5 kg demonstration (ATL10–13) and production runs (ATL14–36) [less than signs (<) denote detection limits]

Sample ID	Al	B	C	Ga	Si	F + Cl	Dissolution vessel
ATL10	<16	6.1	170	0.70	110	<280	Glass
ATL11	100	<2	490	0.59	250	<260	Glass
ATL12	60	7.9	430	0.33	185	<320	Glass
ATL13	75	<2	470	0.33	183	<180	Glass
ATL14	64	3	430	0.057	52	100	Glass
ATL15	40	<2	350	0.051	16	90	Glass
ATL16	52	<2	370	0.051	39	<110	Glass
ATL17	53	2.6	350	0.091	60	120	Glass
ATL18	60	<1	330	0.059	93	80	Glass
ATL19	8	<1	340	0.028	43	60	Glass
ATL20	32	<1	230	0.042	150	<70	Glass
ATL21	<16	<1	390	0.028	90	<90	Glass
ATL22	<16	<1	400	0.025	90	100	Glass
ATL23	47	14	390	0.06	25	<80	Glass
ATL24	20	<2	510	0.054	100	<60	Teflon^{TM}
ATL25	<15	<1	390	0.033	61	<40	Teflon^{TM}
ATL26	25	2.7	220	0.018	98	<100	Glass
ATL27	17	<2	320	0.061	<20	<60	Teflon^{TM}
ATL28	26	3.1	370	0.085	96	<40	Teflon^{TM}
ATL29	24	7.4	240	0.051	50	<70	Teflon^{TM}
ATL30	47	4.8	420	0.059	60	<80	Teflon^{TM}
ATL31	44	3.1	750	0.045	40	90	Teflon^{TM}
ATL32	16	2.7	300	0.02	<20	180	Teflon^{TM}
ATL33	31	2.7	390	0.028	<20	<80	Teflon^{TM}
ATL34	32	7.2	340	0.024	<20	<90	Teflon^{TM}
ATL35	19	<2	310	<0.011	<20	<60	Teflon^{TM}
ATL35R	20	<2	330	<0.011	118	180	Teflon^{TM}
ATL36	33	<2	440	0.014	100	<60	Teflon^{TM}

Dissolution—Teflon^{TM} Vessel

Due to poor dissolution efficiency and PuO$_2$ product impurity levels, Teflon^{TM} vessels were eventually implemented into the aqueous flow sheet. Several batches of ARIES PuO$_2$ proved difficult to dissolve in glass pots, resulting in low dissolution efficiencies, around 75–80%, even with multiple passes. There was also concern that a more aggressive dissolution would attack the glass dissolution vessel leading to increases in Si and B in the final product. Therefore, the process change to 6.0 L Teflon^{TM} pots was implemented starting with ATL24,25 and then ATL27–36. During production, studies

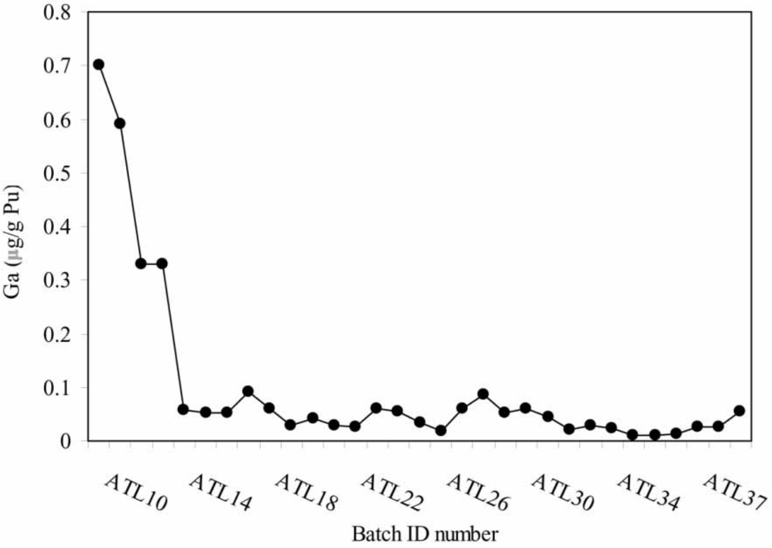

Figure 3.

were conducted (see Tables 3 and 4 for batch I.D. numbers) in the Teflon^{TM} to improve dissolution efficiency by adding more concentrated HF to the initial oxide and generated heels (10). Two methods of HF addition and concentration were examined for their relative efficiency of PuO_2 dissolution.

Tables 3 and 4 list ARIES PuO_2 dissolution results in Teflon^{TM} pots using 15.6 M HNO_3/0.2 M HF and 15.6 M NO_3/0.3 M HF, respectively, for the initial pass and combined residues. A 2 kg batch of ARIES PuO_2 was split into approximately four equal samples in order to run duplicate dissolution studies using each method. After each initial pass, the two remaining residues were recombined and dissolved using the same method as the initial dissolutions. Two different methods were used for HF addition. Method 1 involved adding the HNO_3 first, followed by half of the total HF volume, then the remaining HF volume after 2 h reflux. This standard dissolution method was originally used for the glass dissolvers, ATL10–23. For Method 2, HNO_3 and HF were added to a separatory funnel, and the mixture was allowed to slowly drip into the reaction vessel. Historical experience at LANL has shown that slowly dripping small amounts of HF into the reaction vessel can increase dissolution efficiencies by minimizing insoluble Pu–F complexes.

Based on the data in Tables 3 and 4, Method 1 was preferred for acid addition, but HF concentration made negligible difference on oxide dissolution efficiency for the *initial* pass. Dissolution efficiencies were then improved on the combined residues using Method 1 and increasing the HF

Table 3. Dissolution efficiencies for ARIES PuO_2 using methods 1 and 2 with 15.6 M HNO_3 and 0.2 M HF. An ATL batch ending with "R" denotes residues

Sample I.D.	Initial Pu (g)	Dissolved Pu (g)	Dissolution efficiency (%)
		Method 1	
ATL24A	500	334	66.8
ATL24B	500	404	80.8
ATL25A	500	415	83.0
ATL25B	500	408	81.6
ATL28B	500	430	86.0
ATL28D	500	446	89.2
Average			81.2
ATL25R	547	285	52.1
ATL28R	329	114	34.7
Average			43.4
		Method 2	
ATL24C	500	234	46.8
ATL24D	500	285	57.0
ATL25C	658	404	61.4
ATL25D	500	384	76.8
ATL28A	500	392	78.4
ATL28C	500	403	80.6
Average			66.8
ATL24R	743	384	51.7

Table 4. Dissolution efficiencies for ARIES PuO_2 using 15.6 M HNO_3 and 0.3 M HF for method 1. ATL batches ending with an "R" denote residues

Sample I.D.	Initial Pu (g)	Dissolved Pu (g)	Dissolution efficiency (%)
29A	500	433	86.6
29B	500	391	78.2
29C	500	464	92.8
29D	500	344	68.8
30A	500	430	86.0
30B	500	406	81.2
30C	500	448	89.6
30D	499	394	79.0
Average			82.8
ATL29R	368	248	67.4
ATL30R	321	249	77.6
Average			72.5

concentration to 0.3 M. Dissolution efficiencies for Method 2 and 0.3 M HF were not studied. By implementing the preferred methods listed above, we have been able to increase dissolution efficiency up to 95% with only two passes as compared to 75–80% efficiency in multiple passes with the glass reaction vessels. However, it is still not clear why the higher HF concentration only improved dissolution for the heels and not the initial oxide. Perhaps surface area increased after the first dissolution pass, which contributed to a greater amount of uncomplexed F^- in solution and an increased rate of dissolution.

Dissolution of impure PuO_2 in Teflon^{TM} vessels was expected to reduce B and Si content in the final PuO_2. According to data listed in Table 2, average B and Si content for Teflon^{TM} and glass are not statistically different, even at the 65% confidence interval. Therefore, switching to Teflon^{TM} had no marked effect on lowering these impurity levels in the final oxide product. Further, the averages are also an indicator of the more overall product quality.

Calcination

A new muffle furnace and digital furnace controller were installed and fully implemented within the glovebox line for the second demonstration run. The digital controller regulated time and temperature during calcination and allowed the operator to archive profiles as electronic and hard copy files. For batches ATL10–19 and ATL21, two calcination boats were used due to the size of oxalate cake. The furnace controller was programmed at a 150°C/hour ramp rate to 650°C and then maintained at temperature for a minimum of 4 h (ATL10–14). Starting with ATL15, calcination profiles were modified to include a 6 h presoak at 200°C to drive off excess water in the oxalate cake. Batches ATL20 and ATL22–36 were calcined in larger volume, single boats to facilitate increased production throughput (six ion exchange runs/month for 10 kg total oxide), initiated in April 2003. Run profiles for the single boat continued to use the 200°C presoak followed by a ramp to 650°C. During the implementation of these two calcination process changes, there was slight evidence of effects on product impurity levels. With the exception of ATL24 and ATL32, carbon levels, which come from incomplete calcination, continue to lie just below the 500 ppm requirement (Table 2). These results indicate a possible improvement on the calcination process.

Because calcination conditions ultimately determine PuO_2 physical characteristics, such as surface area, particle size, bulk/tap density, and moisture content, it was imperative to analyze representative samples from ATL10–36 to ensure product quality. Based on the results to date, ATL10–36 met all physical characteristics according to specifications outlined by ASTM C757-90 (3). Tables 6–8 present the average, standard deviation, and relative standard deviation for the determination of moisture content, specific surface area, and particle size. Data indicate that all ATL batches met the ASTM C757-90 specification limit for particle size (95% $<44\,\mu m$ and 100% $<100\,\mu m$) and surface area ($2\,m^2/g < x < 20\,m^2/g$) (3). ASTM C-757 says that moisture should be determined but does not give a limit or an analysis method. LANL is using the DOE 3013 Standard for moisture content, <0.5 wt %. As indicated in Table 8, the average moisture content is 0.22 wt %, which is much below the 3013 Standard of 0.5 wt %.

Impurity Removal Efficiencies

Removal efficiencies for problematic impurity elements are listed in Table 5. Feed and product averages were calculated by averaging all trace analytical sets for ATL10–36. Equation (1) was used to calculate % removal efficiency:

$$100 - \left[\left(\frac{Polished\_Avg}{Feed\_Avg}\right) \times 100\right] \qquad (1)$$

Removal efficiencies presented in Table 5 are comparable to those reported for the first demonstration process. To date, the overall impurity removal efficiency for ATL10–36 was ~86% with an average of 100.00% for Ga removal.

SUMMARY AND CONCLUSIONS

To date, LANL has demonstrated the ability to polish approximately 100 kg of plutonium in the form of PuO_2 for use in fabrication of MOX-LTAs. Results for the Second Demonstration and Production phases clearly indicate that LANL has the ability and consistency in unit operations to produce nuclear fuel quality PuO_2. The unit operations of dissolution, anion exchange, and oxalate precipitation located within the ATLAS

Table 5. Removal efficiencies (%) for the seven problematic elements (feed and product averages were taken for ATL10–36)

	Al	B	C	Ga	Si	F + Cl
Feed average (μg/g Pu)	77	12	160	3,120	73	70
Product average (μg/g Pu)	36	3	370	0.106	79	112
Removal (%)	53	73	−130	100.00	−8.1	−60

Table 6. Particle size distribution for ATL10–36

	Mean (μm)	Median (μm)	Avg. % <44 μm	Avg. % <100 μm
Average	14.80	15.00	99.88	100.00
Std. Dev.	2.15	2.48	0.17	0.00
RSD (%)	15	17	0.17	0

ASTM specification is 95% $<$ 44 μm and 100% $<$ 100 μm.

process line were used to purify the impure PuO_2 from the ARIES demonstration line. A batch calcination process was used to convert the plutonium oxalate to PuO_2 product.

Chemical (problematic trace impurity) content, surface area, particle-size distribution, and moisture content were determined for each ATL batch of PuO_2 product. Elemental analysis for ATL10–36 met the specification limits for problematic impurity content outlined in Table 1, with the exception of B, Si, F + Cl on the Second Demonstration run (ATL10–14). High process efficiencies continued throughout the second 5 kg demonstration and production phases. Overall impurity removal efficiency for ATL10–36 was around 86%, with an average of 100.00% for Ga removal. Gallium concerns associated with the First Demonstration run were resolved by changing the washing procedure after oxalate precipitation. In addition, operator training and routine production schedules helped with achieving consistency in PuO_2 product.

One major process improvement was incorporating two 6 L Teflon™ dissolution vessels into the ATLAS line to increase dissolution efficiencies by using a higher concentration of HF on the PuO_2 residues. Two different methods were evaluated for HF addition. Method 1 was preferred for acid addition, but an increase in HF concentration to 0.3 M made negligible difference on oxide dissolution efficiency for the initial pass. However, utilization of Method 1 coupled with an increase

Table 7. Specific surface area (m^2/g) for batches ATL10–36

	Specific surface area (m^2/g)
Average	9.77
Std. Dev.	1.79
RSD (%)	18

ASTM specifications is $2\,m^2/g < x < 20\,m^2/g$.

Table 8. Average moisture content (wt %) for ATL10–12 and 16–36

	Moisture content (wt %)
Average*	0.22
Std. Dev.	0.08
RSD (%)	36

*Moisture data was not obtained for ATL13–15.

in HF concentration to 0.3 M improved the residue dissolution efficiency. Implementation of these process improvements for PuO_2 dissolution has increased batch dissolution efficiency up to 95% with only two passes as compared to 75–80% efficiency in multiple passes with glass reaction vessels.

Dissolution of impure PuO_2 in Teflon^{TM} vessels was expected to reduce the level of boron and silicon in product PuO_2. However, a comparison of silicon and boron values for ATL14–36 as listed in Appendix A of the LANL report "Qualification Report for Plutonium Oxide Production: Product Characterization" (2) indicates the implementation of Teflon^{TM} dissolution equipment did not lower the value of B and Si levels in product PuO_2.

Temperature control and data recording was problematic for calcinations during the first demonstration, ATL5–9, but was resolved before the second demonstration phase by implementing a new furnace and computer-driven temperature controller that provides electronic, archivable files of the temperature profiles. Larger capacity, single boats were implemented to accommodate a sprint processing operation that required a minimum of six ion exchange runs per month. The calcination run profile for single boat and double boat firings was slightly modified to include a 6 h presoak at 200°C. Moisture content, surface area, and particle size distribution of all batches met specification limits and were not degraded by the new calcination profile and larger capacity boats.

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