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Fusion Techniques for the Oxidation of Refractory Actinide Oxides

by

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Summary

Small-scale experiments were performed to demonstrate the feasibility of fusing refractory actinide oxides with a series of materials commonly used to decompose minerals, glasses, and other refractories as a pretreatment to dissolution and subsequent recovery operations. In these experiments, 1-2 g of plutonium or neptunium oxide (PuO_2 or NpO_2) were calcined at 900°C, mixed and heated with the fusing reagent(s), and dissolved. For refractory PuO_2, the most effective material tested was a lithium carbonate (Li_2CO_3)/sodium tetraborate ($Na_2B_4O_7$) mixture which aided in the recovery of 90% of the plutonium. The fused product was identified as a lithium plutonate (Li_3PuO_4) by x-ray diffraction.

The use of a $Li_2CO_3/Na_2B_4O_7$ mixture to solubilize high-fired NpO_2 was not as effective as demonstrated for refractory PuO_2. In a small-scale experiment, 25% of the NpO_2 was oxidized to a neptunium (VI) species that dissolved in nitric acid. The remaining neptunium was then easily recovered from the residue by fusing with sodium peroxide (Na_2O_2). Approximately 70% of the neptunium dissolved in water to yield a basic solution of neptunium (VII). The remainder was recovered as a neptunium (VI) solution by dissolving the residue in 8M nitric acid. In subsequent experiments with Na_2O_2, the ratio of neptunium (VII) to (VI) was shown to be a function of the fusion temperature, with higher temperatures (greater than approximately 400°C) favoring the formation of neptunium (VII).

The fusion of an actual plutonium-containing residue with Na_2O_2 and subsequent dissolution was performed to demonstrate the feasibility of a pretreatment process on a larger scale. Sodium peroxide was chosen due to the potential of achieving higher actinide recoveries from refractory materials. In this experiment, nominally 10 g of a graphite-containing residue generated during plutonium casting operations was initially calcined to remove the graphite. Removal of combustible material prior to a large-scale fusion with Na_2O_2 is needed due to the large amount of heat liberated during oxidation. Two successive fusions using the residue from the calcination and the residue generated from the initial dissolution allowed recovery of 98% of the plutonium. The fusion of the residue following the first dissolution was performed at a higher temperature (600°C versus 450°C during the first fusion). The ability to recover most of the remaining plutonium from the residue suggest the oxidation efficiency of the Na_2O_2 fusion improves with higher temperatures similar to results observed with NpO_2 fusion.

Introduction

The generation of refractory materials containing residual amounts of plutonium and other actinides was frequently a byproduct of past nuclear weapons production activities. The continued storage of approximately 20,000 kg of incinerator ash at the Rocky Flats Environmental Technology Site is a good example of this practice. The ash, generated by the combustion of job control waste from plutonium-containing gloveboxes, was originally stored for subsequent recovery of plutonium. Traditionally, a nitric acid/fluoride dissolution flowsheet was used to leach plutonium from the ash. Multiple contact times with fresh dissolver solution were generally required to dissolve a majority of the plutonium. Rather than use the traditional, but inefficient, nitric acid/fluoride dissolution flowsheet, the development of a simple solid state fusion for oxidation of plutonium from the 4+ to the more soluble 6+ valence state would be more effective and economical. Dissolution of the pretreated residue in dilute nitric acid and subsequent plutonium recovery could then be accomplished using existing F-Canyon facilities at the Savannah River Site.

Fusion reactions are routinely used to solubilize minerals prior to dissolution and elemental analysis. The feasibility of this approach was demonstrated for pretreatment of small samples of refractory PuO_2 by fusing with Na_2O_2 and potassium superoxide (KO_2) before subsequent dissolution in 1M nitric acid. Plutonium recoveries approaching 90% were obtained.[1] The prospect of using other reagents commonly used for the decomposition of inorganic materials[2] was investigated using a series of small-scale (1 g of PuO_2) experiments. Additional experiments were conducted to determine the feasibility of fusing refractory NpO_2 as a pretreatment to dissolution with the most promising materials. Based on the potential of achieving higher actinide recoveries from refractory materials, fusion with Na_2O_2 was chosen to demonstrate the feasibility of pretreating and recovering plutonium from a graphite-containing residue.

Small-scale Experiments

A series of small-scale fusion experiments was performed to determine the feasibility of using analytical reagents commonly used for decomposition of inorganic materials as a pretreatment prior to dissolution of refractory PuO_2. The experiments were performed by mixing nominally 1 g of PuO_2 with materials normally used as solubilizing agents for the dissolution of minerals, glasses, and other refractory materials. The PuO_2 used in these experiments was calcined at 900°C for 2 hr to produce a difficult to dissolve material which could consistently be used to evaluate the efficiency of a variety of reagents in aiding the dissolution process. A summary of the materials and material amounts used in each experiment is provided in Table 1.

The fusion experiments were performed by mixing the PuO_2 and fusion materials in a small (30 to 100 ml) alumina crucible. The crucibles were placed in a muffle furnace and heated to the desired temperature for 1 to 4 hr. The fused products were initially leached with water to remove soluble salts. The remaining solids were leached with 8M nitric acid for plutonium recovery.

Table 1 Summary of Small-Scale Fusion Experiments

Fusion Reagents	Material Amounts (grams)	Fusion Temperature (°C)	Heating Time (hr)
$Li_2CO_3/Na_2B_4O_7$	2/1	1000	2
$Na_2S_2O_7/K_2S_2O_7$	8/2	325	0.75
$Li_2CO_3/LiBO_2$	1/1	1000	2
$LiOH/NaNO_3/Na_2B_4O_7$	0.68/1/0.28	1000	2
KHF_2	1	200	4
$LiOH/Na_2B_4O_7$	0.6/1	1000	2
$Na_2CO_3/Na_2B_4O_7$	2/1	1000	2
$Na_2CO_3/Na_2B_4O_7$	2/1	1000	2
$LiBO_2$	2	1000	2
$LiF/LiBO_2$ (eutectic mixture)	1.9/6.2	1000	2
$Li_2B_4O_7/NaNO_3$	2/0.5	1000	1
$LiBF_4$	1	1000	2

The dissolutions were performed by placing the crucible on a hot plate/stirrer to provide agitation and heating. Following a 30 to 60 min dissolution period, the crucible contents were filtered and a sample of the filtrate analyzed for plutonium. The plutonium recovery was then determined based on the solution analysis and the amount of PuO_2 used in the experiment. Where appropriate, solid residues were analyzed by x-ray diffraction to identify the fused plutonium species and the spectra of plutonium in nitric acid solution used to identify the plutonium valence.

The most effective material tested for solubilizing the high-fired PuO_2 was the $LiCO_3/Na_2B_4O_7$ mixture which allowed recovery of 90% of the plutonium. The fused plutonium product was identified as a lithium plutonate (Li_3PuO_4) by x-ray diffraction. When the Li_3PuO_4 was dissolved, the plutonium was further oxidized by nitric acid from the 5+ to the 6+ valence state (based on the solution spectra). The use of a 4:1 sodium persulfate ($Na_2S_2O_7$)/potassium persulfate ($K_2S_2O_7$) mixture was somewhat effective in solubilizing the high-fired PuO_2. The experiment was performed by adapting an analytical procedure for high-fired actinide oxides reported by Angeletti and Bartscher.[3] Interestingly, the dissolution of the fused plutonium product was in the 4+ oxidation state. A small sample of the fused product was examined by x-ray diffraction, but no plutonium compounds could be identified. The drawbacks to this procedure include the large 10:1 excess of the persulfate mixture required for the fusion and the concern of introducing large amounts of sulfate into a nitric acid-based plutonium recovery operation which would cause a corrosion problem with the 304L stainless steel equipment.

Surprisingly, the use of other fusion reagents containing both oxidizers and fluxing agents similar to the $Li_2CO_3/Na_2B_4O_7$ mixture were not as effective in solubilizing high-fired PuO_2. In several cases lithium ion was also present in the mixture. Its presence, although the mechanism is not understood, may play a role in successful oxidative fusion. Material combinations similar to the

$Li_2CO_3/Na_2B_4O_7$ mixture which were investigated included: Li_2CO_3/lithium metaborate ($LiBO_2$), lithium hydroxide (LiOH)/sodium nitrate ($NaNO_3$)/$Na_2B_4O_7$, sodium carbonate (Na_2CO_3)/$Na_2B_4O_7$, lithium fluoride (LiF)/$LiBO_2$ (eutectic mixture), and lithium tetraborate ($Li_2B_4O_7$)/$NaNO_3$. The ability of several other material combinations to oxidize refractory PuO_2 were also investigated. The use of a $LiOH/Na_2B_4O_7$ mixture was unsuccessful, presumably due to the lack of an oxidizer. The use of $LiBO_2$ and the $Li_2B_4O_7/NaNO_3$ mixture may have been limited by the relatively high melting point of the starting material(s) which hindered the ability of the reagent(s) from contacting the PuO_2. The $LiBO_2$ fusion also lacked the presence of an oxidizing carbonate or nitrate in the fusion mixture.

Two experiments were performed using potassium hydrogen fluoride (KHF_2) and lithium tetrafluoroborate ($LiBF_4$) as fusion reagents. These materials should produce a more soluble plutonium compound by fluorination with the expected product being plutonium tetrafluoride (PuF_4). In anticipation of producing PuF_4, the fused product was leached with a 2M nitric acid/0.5M aluminum nitrate ($Al(NO_3)_3$) solution. Dissolution of the fused KHF_2/PuO_2 mixture resulted in the recovery of 16% of the plutonium. An optimization of the processing conditions would likely result in an increased recovery. Only a trace of plutonium was detected in the filtrate following dissolution of the product from the $LiBF_4$ fusion.` The failure of this fusion was attributed to the high furnace temperature (1000°C) which likely decomposed the $LiBF_4$ before it had a chance to react with the PuO_2. The plutonium recoveries from each of the small-scale experiment are summarized in Table 2.

Table 2 Plutonium Recovery Following Small-Scale Fusions

Fusion Reagents	Plutonium Recovery (%)
$Li_2CO_3/Na_2B_4O_7$	90
$Na_2S_2O_7/K_2S_2O_7$	56
$Li_2CO_3/LiBO_2$	34
$LiOH/Na_2B_4O_7/NaNO_3$	25
KHF_2	16
$LiOH/Na_2B_4O_7$	12
$Na_2CO_3/Na_2B_4O_7$	10
$Na_2CO_3/Na_2B_4O_7$	10
$LiBO_2$	3
$LiF/LiBO_2$ (eutectic mixture)	2
$Li_2B_4O_7/NaNO_3$	<1
$LiBF_4$	<1

Neptunium Oxide Fusion

A small-scale experiment was performed with high-fired neptunium oxide (NpO_2) to measure the effectiveness of fusion with a $Li_2CO_3/Na_2B_4O_7$ mixture as a pretreatment prior to dissolution. High-fired NpO_2 is quite refractory and can be as difficult to dissolve as high-fired PuO_2. The NpO_2 used in this experiment was heated to 900°C prior to fusion. Nominally 2 g of NpO_2 were mixed with 4 g Li_2CO_3 and 2 g $Na_2B_4O_7$ and fused at 1000°C for 1 hr. The $Li_2CO_3/Na_2B_4O_7$ fusion oxidized 25% of the NpO_2 to a neptunium (VI) species that was easily dissolved in nitric acid. The valence of the neptunium was determined by the solution spectrum. The insoluble residue from this experiment was dried and fused with Na_2O_2 (2g Na_2O_2/g NpO_2) at 380°C for 4 hr. Following the fusion, approximately 70% of the NpO_2 residue dissolved in water to yield a basic solution of neptunium (VII), identified by the neptunium (VII) adsorption spectrum. The remainder of the neptunium was recovered as a neptunium (VI) solution by dissolving in 8M nitric acid.

To understand the role of temperature in the oxidation of NpO_2 to higher valence states, a series of Na_2O_2 fusions was performed with step-wise increases in temperature. High-fired (900°C) NpO_2 was combined with Na_2O_2 equal to 2.8 times its mass. The fusion mixture was heated for 2-3 hr at 300-500°C in 50°C steps, and sampled after heating at each temperatue. The samples were washed with 2M NaOH to dissolve neptunium (VII); the base-insoluble fraction was then dissolved in 8M nitric acid. The ratio of neptunium (VII) to neptunium (VI) was determined by assay of the solutions. Essentially all of the NpO_2 was converted to soluble higher valences. The results from each fusion are shown in Table 3.

Table 3 Fusion of NpO_2 with Increasing Temperature

Temperature (°C)	Np (VII) (mg)	Np (VI) (mg)	$\frac{\text{Np (VII)}}{\text{Total Np}}$ (%)
300	NR	NR	-
350	29.6	76.7	28
400	50.9	24	68
450	13.6	7	66
500	417	24	95

NR – No Reaction

From data in Table 3, one can see that the ratio of neptunium (VII) to neptunium (VI) increased with temperature up to 500°C. To extend the data to higher temperatures, additional experiments were performed from 550 to 650°C. In these experiments, the neptunium oxidations were performed by mixing 6.5 mole Na_2O_2/mole NpO_2 and fusing for 2-3 hr. Dissolution of the fused products in 2M NaOH demonstrated that 99% of the NpO_2 was converted to a green ternary neptunium (VII) oxide powder, presumably by equation (1).

$$NpO_2 + 3/2Na_2O_2 \rightarrow Na_3NpO_5 \tag{1}$$

Oxidation at 620°C with a 1.4 mole Na_2O_2 to 1 mole NpO_2 ratio yielded a brown powder, which was probably the ternary neptunium (VI) oxide formed by equation (2).

$$NpO_2 + Na_2O_2 \rightarrow Na_2NpO_4 \tag{2}$$

Another 1.4 mole of Na_2O_2 was added, and the mixture heated another 2 hr. The product was a green solid that analyzed 95% neptunium (VII). The minimum amount of Na_2O_2 to achieve complete conversion of NpO_2 to neptunium (VII) was judged to be approximately 300% of the stoichiometric requirement. The temperature limit of the fusion is probably the melting point of Na_2O_2, 675°C,[4] where molten Na_2O_2 could attack the fusion vessel.

An interesting sidelight of the fusion of NpO_2 with Na_2O_2 is the separation of plutonium-238 from neptunium-237 during neptunium oxidation to neptunium (VII). Neptunium that has been used to produce plutonium-238 retains enough of the plutonium-238 that alpha-pulse height analysis shows only 10-20% neptunium in the alpha spectrum. After oxidation to neptunium (VII), the spectrum of the recovered neptunium in the base-soluble fraction averaged 91% neptunium-237 alpha.

Fusion of Graphite Fines Residue

Fusion of refractory PuO_2 with Na_2O_2 and a $Li_2CO_3/Na_2B_4O_7$ mixture prior to dissolution clearly achieved the highest recoveries during recent and past[1] small-scale experiments. Based on this work and the fusion experiments performed with high-fired NpO_2, it appears the use of Na_2O_2 has the higher potential for the development of a plutonium recovery process. To test the ability of Na_2O_2 to solubilize PuO_2 on a multi-gram scale, an experiment was designed to recover plutonium from a graphite-containing residue generated during the cleaning of casting molds at the Los Alamos National Laboratory (LANL). The nominal composition of the "graphite fines" residue is given in Table 4.

Table 4 Composition of Graphite Fines Residue

Component	Composition (wt%)
Graphite (Carbon)	75
Calcium Fluoride	9
Magnesium Oxide	7
Plutonium Oxide	7
Minor Oxides	2

Before attempting the fusion with the actual graphite fines residue, a surrogate material was prepared to demonstrate the pretreatment process. A 4 g mixture containing 73 wt% graphite, 15 wt% calcium fluoride (CaF_2), and 12 wt% cerium oxide (CeO_2), the surrogate for PuO_2, was prepared. The surrogate was mixed with a 30% excess in the amount of Na_2O_2 (44.3 g) required to fully oxidize the graphite (see equation 3).

$$C + 2\ Na_2O_2 \rightarrow CO_2 + 2\ Na_2O \qquad (3)$$

The mixture was placed in a 250 ml alumina crucible and fused at 450°C for 2 hr. During the heating period, the crucible shattered due to thermal shock from the heat liberated during the reaction of the graphite with Na_2O_2. The estimated heat of reaction (for equation 3) at 450°C is 48 kcal/mole.[4] Due to the large amount of heat liberated from the oxidation of the graphite by the Na_2O_2 and the large amount of reagent required to perform the fusion, calcination of the graphite fines (and any other residue containing combustible materials) prior to fusion should be performed. A subsequent fusion of the simulated graphite fines following calcination at 1000°C was performed without difficulty.

To demonstrate the pretreatment of actual graphite fines residue with Na_2O_2 prior to dissolution and recovery of plutonium, nominally 10 g of material were placed in a 100 ml alumina crucible and calcined at 900°C until a constant mass was achieved. Nine hours at temperature were required and approximately 75% of the residue mass was lost during the calcination. The calcined residue was then mixed with 0.924 g of Na_2O_2. The mass of Na_2O_2 was based on the uncalcined residue containing nominally 7 wt% plutonium, the stoichiometry of equation (4), and 100% excess reagent.

$$PuO_2 + 2Na_2O_2 \rightarrow Na_4PuO_5 + 1/2O_2 \qquad (4)$$

The calcined residue and Na_2O_2 were heated at 450°C for 2 hr. The fused product was transferred to a 250 ml beaker and dissolved with 8M nitric acid. The beaker contents were stirred and heated at 80-90°C for 1 hr. A watch glass containing water was placed on the beaker to minimize evaporation during the heating period. After cooling, residual solids were filtered using 0.45 μm paper. The filtrate volume was measured and the solution analyzed for plutonium and americium by liquid scintillation counting and a gamma scan.

The filter paper and solids were transferred into a 100 ml alumina crucible and heated at 800°C for 2 hours to destroy the filter paper. A second fusion was then performed using the residue by adding 0.524 g of Na_2O_2, mixing, and heating at 600°C for 2 hr. The fused product was dissolved in nominally 60 ml of 8M nitric acid. The acid was added directly to the crucible since the material was sticky and could not be removed. The sticky consistency of the fused product was attributed to the absorption of moisture while the crucible was stored in the glovebox for approximately 8 days. A stirring bar was added to stir the crucible contents while heating at 80-90°C for approximately 1 hr. A watch glass containing water was placed on the crucible to minimize evaporation during the heating period. Once cool, the crucible contents were filtered through a piece of 0.45µm filter paper. The filtrate volume was measured and the solution analyzed for plutonium and americium by liquid scintillation counting and a gamma scan.

The small amount of remaining solids and filter paper were placed into a 100 ml alumina crucible and heated at 600°C for 2 hr. The residue and approximately 75 ml of a 15.7M nitric acid/0.1M potassium fluoride (KF) solution were transferred to a 250 ml beaker which was covered with a watch glass containing water, stirred, and heated under reflux conditions for 2 hr. The solution was cooled and filtered through 0.45 µm filter paper demonstrating that all of the remaining solids dissolved. The filtrate volume was measured and the solution analyzed for plutonium and americium by liquid scintillation counting and a gamma scan.

The amounts of plutonium recovered from the graphite fines residue following each of the dissolutions are given in Table 5.

Table 5 Plutonium Recovery from Graphite Fines Residue

Dissolution	Recovered Plutonium (g)	Fraction Recovered (%)
Following 1st Fusion	0.25	54
Following 2nd Fusion	0.20	44
Nitric Acid/KF	0.01	2
Total	0.46	100

The recovery of essentially all the remaining plutonium following the second fusion at 600°C suggest the oxidation `efficiency of the Na_2O_2 fusion improves with higher temperatures, similar to the fusion of NpO_2, at least up to the melting point at 675°C.

Conclusions

Small-scale experiments have demonstrated the feasibility of fusing refractory PuO_2 with a series of materials commonly used to decompose minerals as a pretreatment to dissolution and subsequent recovery operations. The most effective material tested for solubilizing high-fired PuO_2 was a $LiCO_3/Na_2B_4O_7$ mixture which facilitated recovery of 90% of the plutonium. The fused product was identified as a lithium plutonate (Li_3PuO_4) by x-ray diffraction. Several other materials were somewhat effective in solubilizing the refractory PuO_2. Fusion with a $Na_2S_2O_7/K_2S_2O_7$ mixture resulted in a 56% plutonium recovery following dissolution in 8M nitric acid; however, a large excess of the persulfate mixture is required and the introduction of large amounts of sulfate into nitric acid-based recovery operations would cause corrosion problems with 304L stainless steel equipment. Other materials showing some promise included $Li_2CO_3/LiBO_2$ and $LiOH/Na_2B_4O_7/NaNO_3$ mixtures which resulted in the recovery of 34 and 25% of the plutonium following dissolution of the fused product in 8M nitric acid.

The use of a $Li_2CO_3/Na_2B_4O_7$ mixture in solubilizing high-fired NpO_2 was not as effective as demonstrated for refractory PuO_2. In a small-scale experiment, 25% of the NpO_2 was oxidized to a neptunium (VI) species that dissolved in nitric acid. The remaining neptunium was then easily recovered from the residue by fusing with Na_2O_2; approximately 70% of the neptunium dissolved in water to yield a basic solution of neptunium (VII) (identified by the adsorption spectrum). The remainder of the neptunium was recovered as a neptunium (VI) solution by dissolving in 8M nitric acid.

A series of experiments with progressively higher reaction temperatures demonstrated that Na_2O_2 fusion of high-fired NpO_2 favored the formation of neptunium (VII) compounds as the fusion temperature increased. At 550°C, 99% of the neptunium was oxidized to a neptunium (VII) ternary oxide. Oxidation of the neptunium to the 7+ valance and dissolution in dilute NaOH can be used to separate neptunium from the residual plutonium-238 alpha activity, if the neptunium has been used for past plutonium-238 production.

The effectiveness of a Na_2O_2 fusion to produce an acid soluble actinide product was judged superior to other materials tested during these studies. This conclusion was based on previous work and the small-scale PuO_2 and NpO_2 fusions. In a larger scale demonstration of the Na_2O_2 fusion using a graphite fines residue from LANL, the necessity of calcining any combustible material prior to the fusion was demonstrated. Excessive heat and gases generated by the oxidation of combustible material by the Na_2O_2 would likely present problems. The large-scale experiment also indicated that the plutonium oxidation efficiency of Na_2O_2 will likely improve with increasing temperatures, similar to the reaction with refractory NpO_2.

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