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ACTINIDE RECOVERY TECHNIQUES UTILIZING ELECTROMECHANICAL PROCESSES

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Abstract

Under certain conditions, the separation of actinides using electromechanical techniques may be an effective means of residue processing. The separation of granular mixtures of actinides and other materials is based on appreciable differences in the magnetic and electrical properties of the actinide elements. In addition, the high density of actinides, particularly uranium and plutonium, may render a simultaneous separation based on mutually complementary parameters. Both high intensity magnetic separation and electrostatic separation have been investigated for the concentration of an actinide waste stream. Waste stream constituents include an actinide metal alloy and broken quartz shards. The investigation of these techniques is in support of the Integral Fast Reactor (IFR) concept currently being developed at Argonne National Laboratory under the auspices of the Department of Energy.

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Introduction

The incorporation of two key features has made the Integral Fast Reactor (IFR) project the focus of DOE's reactor research and development program [1]. Advanced reactor technology coupled with an integrated fuel cycle have led to a single, coherent nuclear power plant concept, Figure 1. Its development and demonstration are being performed at the Argonne National Laboratory in Idaho.

Operation of the Experimental Breeder Reactor-II (EBR-II), a sodium cooled, pool-type, metal fueled reactor, is the first principal component to the concept. Historically, the reactor has been a test bed for nuclear fuels and materials, although recently, the demonstration of its inherent safety characteristics has been its most significant feat. The second component, spent fuel reprocessing, closes the loop for complete on-site power generation. Modifications to an existing facility, the Fuel Cycle Facility (FCF), are currently being finalized in preparation for the pyroprocessing of spent fuel.

Three integral operations will transform the spent fuel into new fuel. The first, electrorefining, separates actinide species from fission products using electric current to drive the chemical reactions. Some fission products, specifically the long-lived, are carried along with the actinides for recycle back to the reactor while the majority of fission products remain in the electrorefiner. These fission products are then removed from the electrorefiner and incorporated into metal waste forms. In terms of waste disposal, the resultant stream is much less radioactive with decay times on the order of hundreds instead of thousands of years. A cathode processor will then consolidate the actinide ingot by high temperature distillation followed by the return of the distillate to the electrorefiner for continued use. Finally, the injection casting of new fuel alloyed with zirconium completes the reprocessing cycle.

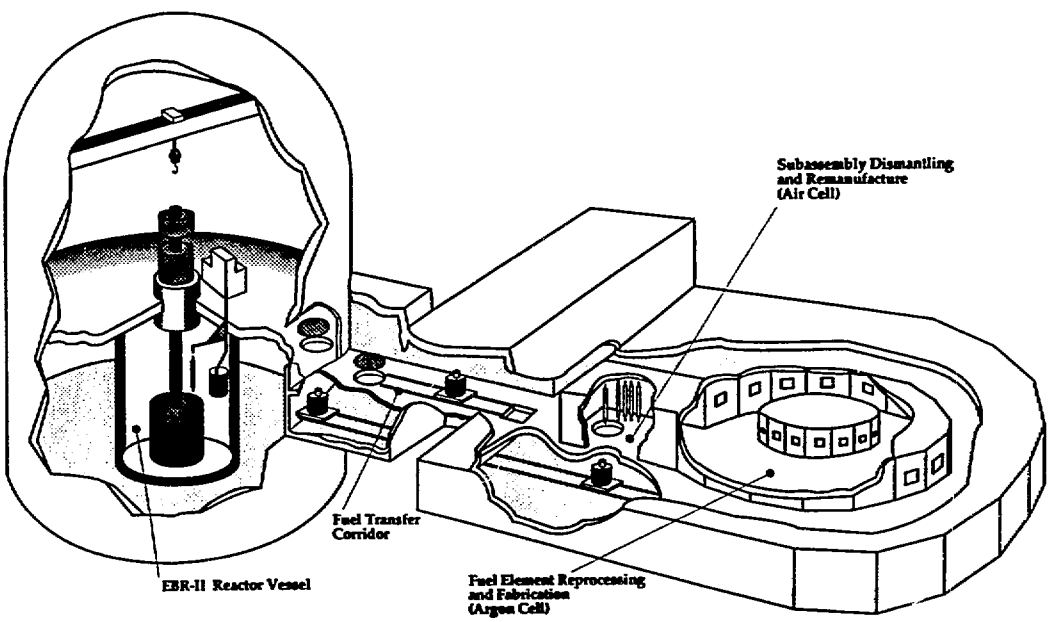

Figure 1. EBR-II Reactor and Fuel Cycle Facility

As a basis for commercialization of the IFR concept, all material losses, particularly actinides, from the fuel cycle must be minimized. Currently, the principal actinide losses will occur following the injection casting of metallic fuel. During demolding of the fuel from their quartz molds, a minor amount of actinide metal alloy remains adhered to the molds despite the use of a zirconia mold wash. Actinide losses from this demolding operation account for approximately 6% of the finished product or about 12 kg a year assuming normal production rates for EBR-II. These values are based on the Fuel Manufacturing Facility's (FMF) injection casting operations for the fabrication of fresh fuel to EBR-II. The FCF casting and demolding operations are modeled after those at the FMF. The alloy currently being cast and investigated for actinide recovery schemes at the FMF, is ninety percent uranium and ten percent zirconium (U-10Zr) by weight. Even though a different alloy (U-20Pu-10Zr) will be utilized for the IFR demonstration program, no appreciable differences in separation performance are expected for the two similar alloys.

The demonstration of fuel reprocessing, and associated tasks, will be performed in an atmosphere controlled, shielded hot cell. Oxygen and moisture contents are maintained at less than 100 ppm in the argon main cell. Hence, any potential process for the separation of actinides from quartz must consider the constraints and limitations for handling radioactive materials. It is for this reason that chemical (wet) separation methods have been excluded from consideration in favor of mechanical (dry) techniques.

Many approaches for the separation of uranium or plutonium from various materials have been documented in the literature [2, 3, 4, 5]. In particular, the research into both wet and dry methods of magnetic separation has shown the most prominence with value recoveries on the order of at least 90%. Although mentioned briefly, electrostatic separation of an actinide residue has been tested and does show promise for classified size fractions [3]. Preliminary studies at Argonne indicate that both comminution/sieving and electrostatic separation have been propitious as a means of recovery for uranium from waste materials [6].

Chemical analyses of the mold scraps, the actinide/quartz waste stream resulting from the demolding process, have been performed extensively to produce the following averaged material breakdown: 21% U, 4% Zr, and 75% quartz by weight. As noted before, these values are based on FMF operations and may vary slightly due to deviations in the casting sequence. A plot of two typical particle size distributions for the mold scraps is given in Figure 2. For both tests, more than 80% of the material is larger than 1mm and in one test up to 91% of the material sieved is larger than 1mm. The variance in the two tests is characteristic of the demolding process.

Although dependent on particle density and geometry, efficient separations are generally achieved with a coarser size fraction (6mm to 0.1mm) for both dry magnetic and electrostatic separations [7]. With finer particles, surface forces tend to predominate, thus causing insufficient recoveries. Hence, the mold scrap material would seem to be an excellent candidate for both types of separation. Also, the high density of actinides might effect a separation, specifically when the centrifugal and gravitational components pertaining to particle mechanisms are considered. Table I gives the electromagnetic properties of selected substances at room temperature. Notice that the actinide species are paramagnetic, positive values, as well as conductive while quartz is neither conductive, nor magnetic.

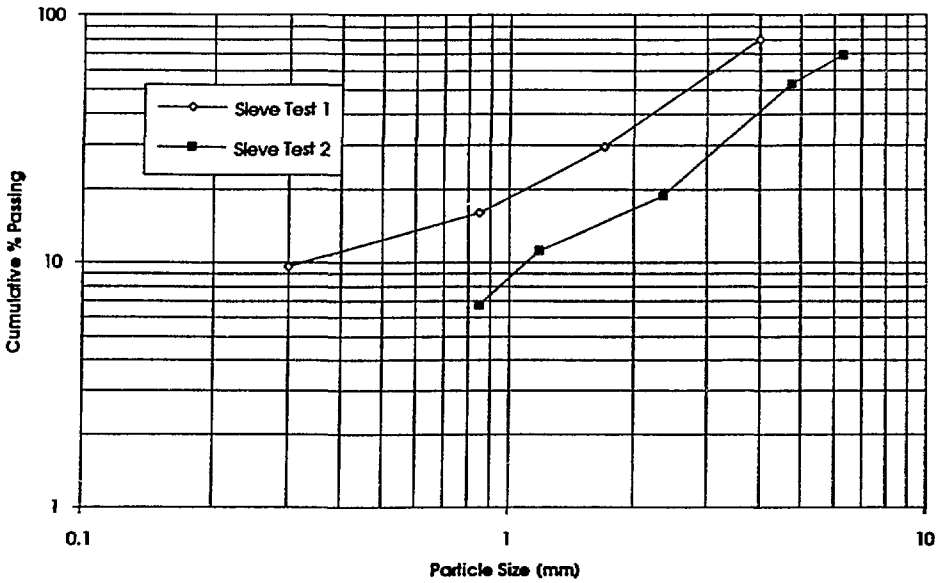

Figure 2. Size Distribution of Mold Scraps

Table I. Electromagnetic Characteristics of Selected Substances

Substance	Specific Magnetic Susceptibility (10^{-6}*cc/gm)	Electrical Conductivity (10^{6}*1/ohm·m)	Electrical Conductivity/Density Ratio (m^2/ohm·kg)
Fe	Ferromagnetic	10.30	1310
Fe_2O_3	+22.45	-----	-----
Sm	+12.37	1.14	151
UO_2	+8.74	-----	-----
PuO_2	+2.66	-----	-----
Pu	+2.52	0.71	36
U-20Pu-10Zr	+1.84	2.72	173
U	+1.72	3.33	175
U-10Zr	+1.68	3.25	203
Zr	+1.34	2.50	385
Al	+0.61	37.67	13,951
Mg	+0.54	22.47	12,915
Cu	-0.08	59.77	6671
Pb	-0.11	4.84	425
Au	-0.14	42.55	2205
Ag	-0.18	62.89	5990
SiO_2	-0.47	10^{-4}-10^{-8}	0
Graphite	-0.50	0.07	32

A separation technique has been developed utilizing both the conductive and magnetic properties of particles. The induction of eddy currents in conducting particles while being exposed to a changing magnetic field has shown excellent results [8, 9]. Determination of eddy current separation potential is a function of the conductivity to density ratio. As can be seen in Table I, aluminum and magnesium exhibit the most conducive ratios for eddy current separation while the actinides are probably not practical.

Experimental

To date, two electromechanical methods have been conducted on the FMF's mold scraps for the recovery of actinides from residue material. Both magnetic and electrostatic separations are discussed and evaluated for their significance.

Magnetic Separation

Scoping studies for the magnetic separation of actinide materials were performed at Hazen Research, Inc. in Golden, CO on a depleted uranium mold scrap sample. The separation characteristics of depleted uranium, enriched uranium, and plutonium should be analogous since the electromagnetic properties are very similar. Two different magnetic methods were investigated at Hazen, both exploiting high intensity magnets (> 2 Tesla) under dry conditions.

Prior to testing, the depleted uranium sample was wet-screened into five size fractions: +5 mesh, -5 /+10 mesh, -10 /+20 mesh, -20 /+48 mesh, and -48 mesh. Wet-screening the sample to remove fines should reduce any possible contamination of equipment by radioactive dust. Subsequent to screening, the +48 mesh material was washed with acetone to promote drying.

As a beginning, an induced roll separator was tested on the -20 /+48 mesh size fraction. An observable lack of segregation of magnetics from non-magnetics led to the blending of these two streams for testing on another separator.

A rare earth permanent magnet separator was then applied to the same material. Because of the obvious improvements in separation on the -20 /+48 mesh fraction, the other three size fractions were also separated using the rare earth belt separator. For all four size fractions, the initial pass of mold scraps through the separator was not completely effective. Consequently, all non-magnetic streams were recycled over the roll magnet until a sufficient separation was achieved, which in no case exceeded two passes.

Electrostatic Separation

The testing of an electrostatic separator for the recovery of actinide material is a continuation of earlier investigations [6]. Previously, the amount of quartz material in the mold scraps was controlled for accountability considerations and feasibility testing. For current testing, the entire mold scrap stream was utilized to simulate the FCF process. Two sets of experiments were performed with the electrostatic device available at the FMF on enriched uranium mold scraps.

Based on prior experience with the electrostatic separator, the mold scrap material +4 mesh was reduced in size by a manual crusher to minimize the amount of quartz carry over. Large quartz shards tend to be carried by momentum into the conductive stream rather than adhering to the ground rotor and, thus, reporting to the non-conductive fraction. This modified feed was then passed through the separator until the conducting material was noticeably devoid of quartz. Two passes of the conductive stream were required to clean the sample.

For the second test, the same procedure was followed for the reduction of material +4 mesh. Though, for this test, material -4 mesh was set aside in hopes of decreasing the throughput of mold scraps while minimizing uranium losses. Both the conductive and non-conductive fractions of the +4 mesh material were recycled until observable results were obtained. The non-conducting fraction required three passes whereas the conducting stream required two passes.

Results

Presentation of the results for the separation of actinides from quartz molds includes three essential elements: uranium recovery, grade of recovered stream, and bulk volume reduction. Of course, uranium recovery is self-explanatory in terms of minimizing actinide losses from the fuel reprocessing circuit. Stream grade or quality is significant in that recovered material may be recycled back to the injection casting furnace. Too much quartz recovery could adversely affect subsequent castings. Grade is calculated as the amount of alloy in the recovered stream versus the amount of alloy and waste in the same stream. Lastly, the ability to directly discard a portion of the mold scraps will alleviate hot cell storage requirements.

As shown in Table II, similar efficiencies were found for all four size fractions separated with the permanent magnet apparatus. Note that any uranium alloy passing the 48 mesh screen was considered lost since no recovery was attempted. The highlighted values indicate results for the entire test. Approximately 80% of the mold scrap waste stream can be disposed of while losing only 3.5% of the uranium. Also, more quartz carry over to the recovered stream occurs as particle size is decreased. This is evident by the decreasing grade values.

Table II. Magnetic Separation Results

Fraction (mesh)	Uranium Recovery (%)	Cumulative U Recovery (%)	Volume Reduction (%)	Cumulative Volume Reduction (%)	Grade (%)	Cumulative Grade (%)
+5	99.95	99.95	60.47	60.47	97.76	97.76
-5/+10	100.00	99.98	82.54	76.22	92.74	95.13
-10/+20	99.92	99.97	80.93	76.98	86.00	93.91
-20/+48	99.81	99.96	73.95	76.76	65.61	91.68
-48	-----	96.59	-----	79.00	-----	-----

Table III gives the results for both sets of experiments using the electrostatic separator on enriched uranium mold scraps. A slight improvement was realized for the second test by the exclusion of the -4 mesh material. However, the recovery rates still do not approach those of the magnetic separator. Volume reduction and grade values are comparable to those from magnetic separation.

Table III. Electrostatic Separation Results

Test	Cumulative Uranium Recovery (%)	Cumulative Volume Reduction (%)	Cumulative Grade (%)
1	78.37	83.27	92.67
2	81.96	78.15	96.28

Conclusions

- By exploiting the electromagnetic characteristics of actinide elements, both magnetic and electrostatic separation have been demonstrated as a viable option for the recovery of actinides from scrap materials.

- Although both methods result in similar volume reductions and grades, magnetic separation is preferable in terms of uranium recovery capability.

- Using the magnetic separation recovery rate of 96%, the expected uranium loss via the mold scraps is decreased from 6% to less than 0.5% of the finished product. This translates to less than 0.5 kg per year, a substantial improvement over 12 kg a year.

- Magnetic separation appears to be a feasible solution for the reprocessing of waste materials to complete the fuel cycle. Recycle of the recovered actinides to the injection casting furnace looks promising considering the final stream grade.

- For electrostatic separation, enhanced recovery of actinides may be possible by classifying the feed material prior to separation. Besides the exclusion of material -4 mesh for the second set of tests, no attempts were made to separate specific size fractions.

Acknowledgments

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