

This article was downloaded by:

On: 25 January 2011

Access details: *Access Details: Free Access*

Publisher *Taylor & Francis*

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Separation Science and Technology

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713708471>

Magnetic Separation as a Plutonium Residue Enrichment Process

L. R. Avens^a; U. F. Gallegos^a; J. T. McFarlan^a

^a Actinide Materials Chemistry Group Los Alamos National Laboratory, Los Alamos, New Mexico

To cite this Article Avens, L. R. , Gallegos, U. F. and McFarlan, J. T.(1990) 'Magnetic Separation as a Plutonium Residue Enrichment Process', *Separation Science and Technology*, 25: 13, 1967 — 1979

To link to this Article: DOI: 10.1080/01496399008050437

URL: <http://dx.doi.org/10.1080/01496399008050437>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

MAGNETIC SEPARATION AS A PLUTONIUM RESIDUE ENRICHMENT PROCESS

L.R. Avens, U.F. Gallegos, and J.T. McFarlan
Actinide Materials Chemistry Group
Los Alamos National Laboratory
P.O. Box 1663, MS E-510
Los Alamos, New Mexico 87545

ABSTRACT

We have subjected several plutonium contaminated residues to Open Gradient Magnetic Separation (OGMS) on an experimental scale. OGMS experiments on graphite and bomb reduction residues resulted in a plutonium rich fraction and a plutonium lean fraction. Values for the bulk quantity rejected to the lean fraction varied between about 20% to 85% of the feed bulk. The plutonium content of the lean fraction can be reduced from about 2% in the feed to the 0.1% to 0.5% range dependent on the portion of the feed rejected to this lean fraction. These values are low enough in plutonium to meet economic discard limits and be considered for direct discard. Magnetic separation of pyrochemical salts gave less favorable results. While a fraction very rich in plutonium could be obtained, the lean fraction plutonium content was too high for direct discard. This may still have chemical processing applications. OGMS experiments at low magnetic field strength on incinerator ash did give two fractions but the plutonium content of each fraction was essentially identical. Thus, no chemical processing advantage was identified for magnetic separation of this residue.

INTRODUCTION

Magnetic separation is a method for the segregation of materials based on differences in magnetic susceptibility. One magnetic separation method called Open Gradient Magnetic Separation (OGMS) can be used on dry powders. When a powder encounters a strong magnetic field, particles with an overall positive susceptibility (paramagnetic) are attracted toward the highest field gradient while those with an overall negative susceptibility (diamagnetic) are repelled from the highest field gradient. This phenomenon serves as a basis for physical separation.

Our interest in the magnetic separation of plutonium residues began when we learned of Oak Ridge National Laboratory magnetic separation work on uranium (1,2). Some success was shown in the

separation of uranium from magnesium fluoride. We choose to examine magnetic separation of plutonium residues since the magnetic susceptibility of plutonium and its compounds is similar to, or in some cases greater than, that of uranium.

Since many of the actinide elements and compounds are paramagnetic we should be able to separate many common impurities from plutonium by OGMS. All compounds with a negative susceptibility should react in the opposite direction from plutonium compounds in a magnetic field. Also, if the residue could be ground into a powder fine enough for all the particles present to be discrete compounds, then a perfect separation would be possible. However this is not practical. As the particles become very small (below about 50 micrometers) random motion in the separation area and other physical forces such as electrostatic attraction become similar in magnitude to the force of gravity or the force created by the magnetic field. These phenomena have been noted by other workers (3,4). This is one reason a complete or perfect separation is difficult to achieve. Since we are forced to work with larger particles, we can assume that many of the large particles consist of mixtures of most of the compounds. The deflection of a particle under the influence of a magnetic field is proportional to the weighted average of the volume magnetic susceptibilities of the particle components.

Given these facts we can see that complete separation should not be a goal of this separation method. A more realistic goal would be to process residues in such a manner as to obtain a plutonium rich fraction and a plutonium lean fraction. The rich fraction would then be processed for plutonium recovery while the lean fraction could be discarded directly if its plutonium content met economic discard limits. If such a separation could be made numerous benefits would be realized, e.g. reduced feed bulk into a dissolver, richer solution from dissolution into ion exchange or solvent extraction, and reduced salt load in subsequent evaporation steps.

Chemical (solution) methods of plutonium recovery processing generate large quantities of waste. A dry separation method, however, would not create any "new" waste and would allow direct discard of a portion of the residue. Such a method would certainly be beneficial to the DOE defense complex in light of recent waste minimization guidance. Hence, we have evaluated magnetic separation as a plutonium residue enrichment process for several contaminated residues and report the results here.

EXPERIMENTAL

The equipment used for this magnetic separation work is the Magnetic Barrier Laboratory Separator (Model LB-1) purchased from the S.G. Frantz Company Inc. (5). The separator was installed in a glovebox enclosure and slightly modified by removing all electronic controls and locating them outside the glovebox. Also, an alternative power supply (Model TCR-150-S4-1D-10T) was purchased from Electronic Instruments Inc. to allow operation at 2.0 Amps. At this power level the magnetic field is approximately 20,000 gauss. The LB-1 separator has a magnetic force to field intensity ratio more than three times greater than the Model L-1 separator used in the ORNL work (1).

The LB-1 separates mixtures in three different operating modes; diamagnetic, paramagnetic, or ferromagnetic. The selected mode depends on the magnetic susceptibility of the species one wishes to isolate or concentrate. In general, the sample is fed onto a vibrating tray at a predetermined rate. Flow rate along the tray is determined by a combination of slope (longitudinal angle) and vibration amplitude. The application of a magnetic field effects separation based on the magnetic susceptibility of the particles. The force exerted on the particles is in a direction perpendicular to the particle flow and field direction. Diamagnetic particles experience a force which urges them toward the operator while the converse is true of paramagnetic particles. At the lower end of the tray a divider at the center line separates the particles into two fractions (Figure 1).

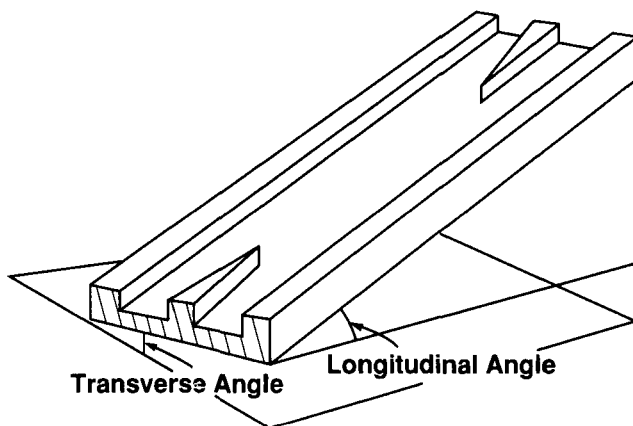


Fig. 1. A Drawing of the LB-1 magnetic separator tray. As shown, the transverse tray angle is negative and the operator is located to the right of the tray.

Diamagnetic separation is effected by additional sloping of the feed tray in a direction perpendicular to the direction of flow and away from the operator (negative transverse angle). The sample is fed into the apparatus at the top of the tray and toward the front (i.e. nearest the operator). In the absence of a magnetic field the particles move away from the operator and to the rear of the chute. When the field is applied, diamagnetic particles are repelled and prevented from crossing the chute center line by the field gradient while paramagnetic particles are drawn across the chute center line by both gravity and the field. In operation, the fraction collected nearest the operator is enriched in the diamagnetic portion of the feed.

Conversely, paramagnetic separation requires the chute be oriented with a positive transverse angle (i.e. toward the operator). The feed material is introduced at the rear of the tray (away from the operator). Now gravity draws the particles down the tray and towards the operator with paramagnetic particles prevented from crossing the tray center line by the magnetic field. We have used the paramagnetic separation mode more than the diamagnetic separation mode in this work because we wished to concentrate the paramagnetic plutonium compounds from the feed matrices which are largely diamagnetic. However, the diamagnetic mode has use if a fraction very low in paramagnetic species is desired.

Ferromagnetic separations require the use of the "Low Field Control" unit. This option allows operation of the separator at low magnetic fields, thereby allowing separation of materials containing ferromagnetic particles.

Very sensitive separations can be made through: 1) adjustment of longitudinal and transverse tray angles, 2) control of the magnetic field strength, and 3) multiple sample passes under various conditions.

Sand, slag, and crucible (SS&C) residue, bomb reduction sand, contaminated graphite, electrorefining (ER) salt, direct oxide reduction (DOR) salt and incinerator ash were the test matrices evaluated by OGMS. The origin, chemical recovery process technology, and economic discard limits for these plutonium contaminated residues are detailed in reference six. In general the residues were crushed and sized through standard sieves to remove fine particles. Particular information concerning particle sizes is included in the discussion of each residue studied. Experimental runs used 30 to 200 grams of bulk residue. The longitudinal tray angle was held at 20 degrees throughout our experimental work.

Plutonium values were obtained by two methods; nondestructive neutron counting and certified destructive radiochemical analysis by the analytical chemistry group (CLS-1) at Los Alamos National Laboratory. For some residues, high levels of americium or light elements (fluoride, magnesium, carbon, etc.) introduced considerable uncertainty in the neutron count results. However, since the neutron results were used as a comparison between rich and lean fractions, rather than absolute plutonium values, these uncertainties do not affect our conclusions.

Usually, a plutonium contaminated residue was run through the separator under a variety of operating conditions with neutron counting used to give an indication of separation performance. When a particular separation protocol was determined (based on the neutron counting) the samples were remixed and the separation repeated using the determined separation parameters. Samples were then collected for certified analysis. When the quantity of plutonium contaminated residue permitted, three samples were subjected to magnetic separation to increase statistical confidence in the result.

RESULTS AND DISCUSSION

Sand, Slag, and Crucible

Sand, Slag and Crucible (SS&C) residue results from the chemical reduction of plutonium tetrafluoride to plutonium metal using calcium metal as the reductant (6). The byproduct is a calcium fluoride slag. The reduction occurs in a magnesium oxide crucible that cannot be reused. The crucible is insulated from the pressure vessel with magnesium oxide sand.

SS&C is a lean residue, generally containing only 1-5% plutonium. The plutonium recovery method at Los Alamos involves grinding and pulverization of the residue prior to nitric acid dissolution (6). The plutonium is recovered and purified by ion exchange from the nitric acid solution.

Economic discard levels vary from site to site and year to year and are highly dependent on the particular residue (6). In general, recent discard levels for SS&C have been between about 5 and 10 grams of plutonium per kilogram of bulk material. We hoped to achieve separation such that an appreciable quantity of the feed material would meet these economic discard limits.

For our magnetic separation experiments we intercepted a batch of SS&C residue after crushing and pulverization. We first wished to investigate the effect of particle size on separation. Thus, a portion of the SS&C was sieved through two screens with openings of 250 and 90 micrometers. This gave three fractions with: 1) particle sizes larger than 250 micrometer, 2) smaller than 250 but larger than 90 micrometer, and 3) smaller than 90 micrometer. X-Ray diffraction showed that the fraction less than 90 micrometer in size was mostly calcium fluoride. Also, Scanning Electron Microscopy, Electron Backscatter, and X-ray Emission Spectroscopy of various samples of SS&C indicated that the most likely chemical form of the plutonium was exclusively plutonium oxide. The plutonium present in the residue appeared as plutonium rich inclusions in magnesium oxide, calcium oxide or fluoride rather than discrete particles of plutonium oxide.

Paramagnetic separation test results and operating parameters for these samples are shown in Table 1. The parameters used for these experiments were arrived at through separate experiments where neutron counting was used as the diagnostic. In the initial experiments a small portion (about 0.5%) of ferromagnetic material was invariably found adhered to the magnet pole pieces. In any large scale operation removal of the ferromagnetic material prior to separation at high field would be required.

While there are a variety of ways to report the results, we choose to give the bulk split and the plutonium split. The bulk split is simply the percentage of the feed that reports to the diamagnetic and paramagnetic fractions. The plutonium split is the plutonium content of either the diamagnetic or

Table 1: Magnetic separation results on sized sand slag and crucible material.

Sample	Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split	
		Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic (g Pu/kg bulk)	Plutonium Rich Paramagnetic (g Pu/kg bulk)
>250μM	15	67.3	32.7	7.6	32.1
<250μM > 90μM	5	31.6	68.4	6.0	21.9
< 90μM	30	7.7	92.3	10.5	18.7

paramagnetic fraction in grams of plutonium per kilogram of bulk in that fraction. For example, in Table 1 the sample with particles larger than 250 micrometer splits into a diamagnetic fraction that consist of 67.3% of the feed for the experiment. This fraction has a plutonium content of about 7.6 g Pu/kg of bulk.

Separation was most effective on the largest particle size fraction with over 67% of the material being rejected as diamagnetic in one pass with a plutonium content of 7.64 g Pu/kg of bulk. Separation was less effective on the intermediate sized fraction. Here, about 32% of the feed was rejected with a plutonium content of 6.00 g Pu/kg of bulk. Even at the relatively high transverse angle of 30 degrees, only 7.7% of the bulk of the fine fraction was rejected. Also, the plutonium content of this fraction was too high to be considered for direct disposal. Repeated pass experiments through the magnetic field were not performed on these samples.

The success with the sieved material led us to examine the paramagnetic separation of SS&C as received. We quickly found that no set parameters would reliably give a lean fraction low enough in plutonium to be considered discardable.

Another group of experiments was conducted using repeated passes through the field in the paramagnetic mode. Here, three feed samples were passed through the magnetic field at a relatively high transverse tray angle, then only the lean fraction was recycled through the field at successively reduced transverse tray angles until the lean fractions had a plutonium content below 10 g Pu/kg of bulk.

The results from these experiments are shown in Table 2. To achieve a discardable component, four passes through the field were required. Also, the discardable fraction consisted of only about 33% of the original feed.

Table 3 shows the results from a similar experiment where particles smaller than 45 micrometer were removed from the SS&C before magnetic separation. Here only three passes were required to achieve the discard level on all three samples. Also note that two of the three samples reached the discard level after only two passes. Another important point is that the discard fraction comprises about 66% of the feed bulk. This lean fraction contains only about 25% of the plutonium in the feed batch.

The question arises that if separation at 15 degrees transverse tray angle gives a discardable lean fraction, why not hold this parameter constant and attempt to achieve a one pass separation? Experiments to demonstrate this were unsuccessful. It appears that if one attempts to separate too large a fraction in one pass, the separation efficiency suffers. Modest feed rates (about 10 g bulk/min), separating modest

Table 2: Magnetic separation results on sand slag and crucible residue as received.

Sample	Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split ^(a)	
		Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic Fraction (g Pu/kg bulk)	Plutonium Rich Paramagnetic Fraction (g Pu/kg bulk)
13	40	80.0	20.0	17.6	32.9
13A		60.3	39.7	13.2	32.5
13B		68.2	31.8	15.0	29.9
13	30	77.2	22.8	17.4	49.9
13A		59.0	41.0	15.0	26.0
13B		64.5	35.5	11.3	30.2
13	20	45.8	54.2	8.9	30.9
13A		50.7	49.3	13.7	30.0
13B		58.0	42.0	12.6	28.4
13	15	32.9	67.1	9.3	34.0
13A		30.6	69.4	3.8	27.3
13B		35.1	64.9	7.2	26.7

^(a) All plutonium values were obtained by neutron counting. While some scatter is observed in the values, we have observed that neutron counting of this residue is generally accurate $\approx 25\%$ of the chemical analysis value.

fractions of material appear to give "complete" separation under that set of conditions. For example, other experiments showed that if SS&C was run at 35 degrees transverse angle and the plutonium rich paramagnetic fraction was run again under the same set of conditions, no separation occurred on the second pass.

The same samples were subjected to magnetic separation in the diamagnetic mode. The results in Table 4 indicate that 20 to 30% of the feed can be rejected with the very low plutonium content (about 1 to 2 g Pu/kg bulk). This is quite impressive since these results were obtained in only one pass. The scatter in the results is likely due to incomplete sieving of the sample with the high plutonium value. Similar results were observed in the paramagnetic separation results (Table 3).

One would expect that the rich fraction from the diamagnetic separation experiments could be reprocessed in the paramagnetic mode to achieve higher overall separation performance (more feed rejected with a low plutonium content). However, attempts to demonstrate this separation protocol were unsuccessful.

Magnetic separation of SS&C as received in the diamagnetic separation mode gave a maximum of 20% of the bulk rejected into the lean fraction. However, the plutonium content of this fraction was above 20 g Pu/kg of bulk. This again demonstrates the detrimental effects of fine particles.

Table 3: Magnetic separation results on sand slag and crucible with particles smaller than 45 micrometer removed.

Sample	Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split ^(a)	
		Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic Fraction (g Pu/kg bulk)	Plutonium Rich Paramagnetic Fraction (g Pu/kg bulk)
15	35	87.9	12.1	17.2	48.3
16		85.0	15.0	13.6	67.3
17		83.3	16.7	15.3	79.5
15	25	73.7	26.3	5.7	31.6
16		76.3	23.7	6.0	76.1
17		77.1	22.9	17.9	67.5
15	15	63.6	36.4	5.2 (6.3)	53.3 (45.8)
16		66.1	33.9	5.4 (7.1)	54.5 (30.5)
17		70.1	29.9	10.4 (8.0)	60.5 (46.7)

^(a)Numbers in parenthesis indicate chemical analysis values. Other plutonium values are neutron count results.

Table 4: Magnetic separation results on sized SS&C in the diamagnetic mode.

Sample	Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split ^(a)	
		Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic Fraction (g Pu/kg bulk)	Plutonium Rich Paramagnetic Fraction (g Pu/kg bulk)
15	-2	18.9	81.1	0.9	30.0
16		24.4	75.6	6.5	26.5
17		19.3	80.7	0.9	33.1
15	0	28.3	71.7	1.8	38.2
16		29.3	70.7	5.1	31.6
17		23.0	77.0	1.5	27.4

^(a)Plutonium values were obtained by neutron counting.

A few experiments were conducted to examine the effect of feed rate on separation performance. We found that separation suffers at rates above about 10 g/min. Similar results were obtained for most of the residues examined in this study.

Bomb Reduction Sand

A recent change at Los Alamos is the segregation of the bomb reduction sand from the slag and crucible to meet fine particle limits for waste destined for the Waste Isolation Pilot Plant. The bomb reduction sand is fused electrical grade magnesium oxide and is used to insulate the reduction crucible from its outer pressure containment vessel in the bomb reduction process. Several experiments were conducted to achieve magnetic separation of a plutonium rich fraction from this material. The LB-1 was operated exclusively in the paramagnetic mode for these separations.

Table 5 shows the results from two samples of sand. Again a trace of ferromagnetic material was observed. The sized sand was sieved before it was introduced into the glovebox line for use in the bomb reduction process. (The sand used was held on a screen with 90 micrometer openings.) After the sand was used in the normal mode of operation in the bomb reduction process, it was passed through the magnetic field only once at a transverse tray angle of 15 degrees. As the results show, most of the plutonium was concentrated in only 15 percent of the bulk while the lean fraction is well into the discard range. The separation performance here is quite impressive. It has been pointed out that the lean fraction could be recycled to the bomb reduction process. Magnetic separation with recycle of the lean fraction to bomb reduction would lead to a substantial reduction in the sand requiring chemical recovery. This reduction in bulk to dissolution is significant as it has been estimated that over 1000 l of solution waste is generated in the chemical recovery of 1 kilogram of plutonium from this lean residue.

Table 5: Magnetic separation results for bomb reduction sand.

Sample	Bulk Feed Split		Plutonium Split	
	Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic Fraction (g Pu/kg bulk)	Plutonium Rich Paramagnetic Fraction (g Pu/kg bulk)
SIZED SAND	84.7	15.3	5.1	53.6 ^(a)
SAND AS RECEIVED	68.7	31.3	8.6	31.3 ^(b)

^(a) Plutonium values were obtained by chemical analysis.

^(b) Plutonium values were obtained by neutron counting.

Magnetic separation of the sand as it was received from the bomb reduction process was not as favorable. The result reported is from the 15 degree magnetic separation pass. However, two passes at a transverse tray angle of 30 and 20 degrees were performed on this sample prior to the 15 degree run. After the 30 and 20 degree runs the lean fraction still held 14 g Pu/Kg of bulk. Also, it should be noted that the rich fraction is twice as large here as in the sized sand result. Since we did not attempt a one pass 15 degree run on this sand sample we cannot say that a one pass separation is not possible. However, we feel that the advantages of sizing the sand prior to use in the bomb reduction process are readily apparent.

Graphite

Graphite is widely used as casting molds for actinides (6). Recovery methods generally center around either crushing the molds or grinding away the surface where the plutonium contamination resides. The resulting graphite powders are then leached in nitric acid to selectively dissolve the actinides. Economic discard levels for this residue are generally in the range of 5 g Pu/kg of bulk.

For our magnetic separation experiments a portion of a graphite mold was crushed in a mortar and sieved through 60 mesh (250 micrometer opening) and 170 mesh (90 micrometer opening) screens. The material not passing the 170 mesh screen was subjected to magnetic separation in the paramagnetic mode. The results in Table 6 show that over 40% of the graphite can be rejected with a plutonium content of about six grams of plutonium per kilogram of graphite. Doubtless a fraction more lean in plutonium could have been obtained with further passes at lower transverse tray angle. Experiments were discontinued due to the relatively high radiation field from the americium content of the sample.

Table 6: Magnetic separation results for plutonium contaminated graphite powder.

Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split ^(a)	
	Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic Fraction (g Pu/kg bulk)	Plutonium Rich Paramagnetic Fraction (g Pu/kg bulk)
35	59.5	40.5	18.0	38.9
20	46.0	54.0	13.7 (6.1)	34.0 (26.6)

^(a)Plutonium values in parenthesis are chemical analysis values. Other plutonium values are from neutron counting. The neutron counts in this case were inflated by the presence of relatively large quantities of americium.

Incinerator Ash

Incinerator ash results from the burning of plutonium contaminated materials. The particle size of the ash was quite small and no sizing of this residue was performed. The residue is relatively rich in plutonium (up to 30%) but is extremely difficult to chemically process for plutonium recovery. The processing difficulties arise from the variability of ash composition and recalcitrant nature of the plutonium oxide in the residue.

Essentially all DOE sites have used incineration as a volume reduction method. Due to the difficulty of processing the economic discard limits for this residue are relatively high at about 25 g Pu/kg of bulk. Current process technology involves a nitric acid leach with plutonium recovery from the acid solution.

Table 7 shows our magnetic separation results. Since the ash contains a large portion of ferromagnetic material, our separation experiments were conducted at low magnetic field strength and the results are reported for the more magnetic and less magnetic fractions. For calcined ash we found that parameters could be adjusted such that a separation could be achieved. However, both the more magnetic fraction and the less magnetic fraction contained similar quantities of plutonium.

Table 7: Magnetic separation results for several incinerator ash samples.

Sample	Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split ^(a)	
		Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic (g Pu/kg bulk)	Plutonium Rich Paramagnetic (g Pu/kg bulk)
Calcined Ash	25	87.8	12.2	184	162
Calcined Ash	30	14.7	85.3	217	200
Fluorinated Ash	30	97.8	2.2	263	118
Leached Ash Heel	30	86.0	14.0	49	28

^(a) All Plutonium values are derived from neutron counting.

Since the paramagnetic susceptibility varies between various compounds of an element we attempted magnetic separation of fluorinated ash. Here a lean fraction depleted in plutonium was obtained but the plutonium level was so high and the size of the fraction so small that little processing advantage would result from such a separation.

We also demonstrated magnetic separation of leached incinerator ash heel. Here a lean fraction consisting of 14 percent of the bulk was obtained. The plutonium content of this fraction was near the discard level but it is questionable that separation of such a small fraction would be useful.

It is possible that some incinerator ash matrices exist that would give more favorable results with OGMS. This was not pursued.

Pyrochemical Salts

Electrorefining (ER) is an electro-chemical purification process for plutonium metal (6). The salt used in this process is usually a sodium-potassium chloride mixture. Direct Oxide Reduction (DOR) is a pyrometallurgical process where plutonium oxide is reduced to plutonium metal in molten calcium chloride. The limited development of chemical recovery process technologies and the problems of handling corrosive chloride solutions in stainless steel glovebox enclosures have led to economic discard levels which vary between about 5 to 25 grams of plutonium per kilogram of bulk salt for these residues.

Our magnetic separation results for these residues are reported in Tables 8 and 9. The ER salt sample was received in the crushed and pulverized condition and was split into three smaller samples for the separation experiments. The DOR salt was a massive piece of calcium chloride/calcium oxide. The lower portion of the salt, which contained most of the plutonium in the salt cake, was cleaved from the bulk, crushed, and sieved. This is an important departure from experiments on other residues studied in this report. By isolating only the rich fraction of the salt, we enriched the feed material prior to the magnetic separation experiment. The fraction larger than 170 mesh but smaller than 60 mesh was used for magnetic

Table 8: Magnetic separation results for electrorefining salt.

Sample	Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split ^(a)	
		Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic (g Pu/kg bulk)	Plutonium Rich Paramagnetic (g Pu/kg bulk)
A	15	98.0	2.0	63.2	341
B	0	45.8	54.2	31.5	101
B	-2	50.0	50.0	72.5	148
C	-2	88.1	11.9	39.9	263

^(a)Plutonium results for samples A and C are from neutron counting. Results for Sample B are from chemical analysis.

Table 9: Magnetic separation results for direct oxide reduction salt.

Transverse Tray Angle (Degrees)	Bulk Feed Split		Plutonium Split ^(a)	
	Plutonium Lean Diamagnetic Fraction (%)	Plutonium Rich Paramagnetic Fraction (%)	Plutonium Lean Diamagnetic Fraction (g Pu/kg bulk)	Plutonium Rich Paramagnetic Fraction (g Pu/kg bulk)
35	99.4	0.6	15.8	≈1000
15	99.1	0.9	15.8	≈ 792
5	82.7	17.3	(11.0)	(55.6)

^(a)Results in parenthesis are from chemical analysis; others are from neutron counting.

ER salt was subjected to separation in the paramagnetic mode. A small fraction (2%) very rich in plutonium was obtained but the lean fraction was much too high in plutonium to be considered discardable. Further test in the paramagnetic separation mode showed that if the transverse angle was reduced to the point that more material was transported into the rich fraction, selectivity suffered (i.e. the plutonium content of the separated fractions was very near their initial value). The remainder of the experiments were conducted in the diamagnetic separation mode.

Similar to the paramagnetic separation results, diamagnetic separation results from sample B show that a fraction rich in plutonium can be obtained. However, the lean fraction is still far above any reasonable discard limit.

Sample C was sieved to remove particles smaller than 90 micrometer. While this appears to improve separation, the lean fraction is still high in plutonium.

One reason as to why magnetic separation of ER residue fails to give a very lean fraction may be due to the chemical form of the plutonium in the salt. If a portion of the plutonium in the ER salt exists as a plutonium halide, then the plutonium is actually dissolved in the salt matrix rather than a mixture of heterogeneous phases. Attempts to verify this through surface analysis methods failed as difficulty was encountered preparing the samples.

The DOR salt sample was passed through the field first at 35 degrees with only the lean fraction being recycled at 15 then 5 degrees transverse tray angles. The first and second passes through the field gave a very small rich fraction that appeared as a dark colored powder. This was probably plutonium metal that had not coalesced into the product plutonium metal button in the DOR process. On the third pass the transverse tray angle was reduced to the point where depletion of the lean fraction was more successful. This still left the lean fraction of the salt with a plutonium content of 11 g Pu/kg of bulk. However, the bulk fraction of the salt not processed (the parent block) had a plutonium content considerably lower than this value (about 3 g Pu/kg).

An economic assessment of these results would show if separation of a very small plutonium rich fraction from this residue would be useful.

CONCLUSIONS

We have shown that OGMS can be used to enrich certain residues in plutonium. Hence, potential chemical processing applications exist in that some of the residues studied allowed the separation of a portion of the feed into a lean fraction low enough in plutonium content to be considered for direct discard. This is true for SS&C residue, bomb reduction sand, and plutonium contaminated graphite. The benefit that results from this type of operation is the concentration of the plutonium in feed materials prior to plutonium recovery operations. Highgrading lean feeds: increases the capacity of the dissolution equipment since reduced bulk is processed, increases the plutonium concentration in subsequent process solutions allowing more efficient ion exchange or solvent extraction operations, and decreases solution volume to evaporators or other contaminated solution handling processes. The quantity of reagents used for the chemical recovery operation is also reduced.

For a few residues such as DOR salt and ER Salt, OGMS can be used to produce a rich fraction and a lean fraction where the latter might not meet economic discard levels. Potential processing applications still exist, however, where processing facilities have a "fast" processing portion and a "slow" processing portion. The advantage here is that most of the plutonium could be processed more quickly and efficiently in the fast portion of the plant.

The incinerator ash samples we studied could not be processed by OGMS to give a plutonium rich and plutonium lean fraction. An improved OGMS separator or selected ash pretreatment methods might improve the separation of this residue. Also, since the incinerator ash consist of rather small particles, High Gradient Magnetic Separation might be successfully applied.

Residues that were separated successfully by OGMS consist of heterogeneous particles. In the residues of this type (SS&C, sand, and graphite) there exist a broad spectrum of plutonium content in the particles. The particles high in plutonium content are, of course, more highly paramagnetic and thus, are deflected more strongly in the magnetic field. We suggest that in ER salt, at least some of the plutonium is dissolved in the salt phase making physical separation methods impractical. For incinerator ash it appeared that the plutonium is equally distributed between the more magnetic and less magnetic fractions.

Higher field or modified design of OGMS equipment might give more sensitive or more complete separations.

ACKNOWLEDGMENT

The authors wish to thank Mary E. Salazar for her help in the preparation of this report and Thomas D. Wellington of the S.G. Frantz Co. Inc. for the many helpful discussions. We also wish to thank William B. Hutchinson and others in the Analytical Chemistry Group at Los Alamos National Laboratory for their help with the surface and chemical analysis.

REFERENCES

1. Hoegler, J.M., and W.M. Bradshaw, U.S. DOE Report ORNL TM-11117, Oak Ridge National Laboratory, Oak Ridge, TN, 1989.
2. Wichner, R.P., A.A. Khan, and J.M. Hoegler, U.S. DOE Report ORNL TM-11141, Oak Ridge National Laboratory, Oak Ridge, TN, 1989.
3. Holman, A.S., E.C. Hise, and J.E. Jones, U.S. DOE Report ORNL-5764, Oak Ridge National Laboratory, Oak Ridge, TN, 1982.
4. Hise, E.C., I. Wechsler, and J.M. Doulin, U.S. DOE Report ORNL-5763, Oak Ridge National Laboratory, Oak Ridge, TN 1981.
5. Sun, J.J., U.S. Patent 4,235,710, S.G. Frantz Co. Inc., 1980.
6. Christensen, D.C., D.F. Bowersox, B.J. McKerley, and R.L. Nance, U.S. DOE Report LA-11069-MS, Los Alamos National Laboratory, Los Alamos, NM, 1988.