LA-UR- 04-7309

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Submitted to: Space Technology and Applications International Forum
(STAIF 2005)
Albuquerque, NM
February 13-17

• Los Alamos
NATIONAL LABORATORY

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Abstract. Pu-238 heat sources are used to fuel radioisotope thermoelectric generators (RTG) used in space missions. The demand for this fuel is increasing, yet there are currently no domestic sources of this material. Much of the fuel is material reprocessed from other sources. One rich source of Pu-238 residual material is that from contaminated combustible materials, such as cheesecloth, ion exchange resins and plastics. From both waste minimization and production efficiency standpoints, the best solution is to recover this material. One way to accomplish separation of the organic component from these residues is a flameless oxidation process using molten salt as the matrix for the breakdown of the organic to carbon dioxide and water. The plutonium is retained in the salt, and can be recovered by dissolution of the carbonate salt in an aqueous solution, leaving the insoluble oxide behind. Further aqueous scrap recovery processing is used to purify the plutonium oxide. Recovery of the plutonium from contaminated combustibles achieves two important goals. First, it increases the inventory of Pu-238 available for heat source fabrication. Second, it is a significant waste minimization process. Because of its thermal activity (0.567 W per gram), combustibles must be packaged for disposition with much lower amounts of Pu-238 per drum than other waste types. Specifically, cheesecloth residues in the form of pyrolyzed ash (for stabilization) are being stored for eventual recovery of the plutonium.

INTRODUCTION

Molten Salt Oxidation (MSO) is a technology that was developed in the 70's by Rockwell for DOE. DOE complex sites (West, 1992) used MSO as a method for treating organic and radioactive wastes (McKenzie et al., 1975); however, the recovery of Pu retained in the salt was not pursued and MSO was temporarily sidelined when incineration became the preferred disposal method. Incineration is no longer a treatment option and MSO has been revisited as a viable disposal technique (Hsu, 2000). Our improved MSO process addresses two new challenges: 1) placement of the process inside a glovebox not specifically designed to accommodate the process, and 2) recovery of the plutonium "contaminant" from the salt.

We developed and tested two MSO systems in a facility outside of the radiation controlled area (RCA). One, a batch mode unit where ash and salt solids are mixed and placed in the crucible before heating the salt; and second, a continuous mode unit that mechanically adds ash via an air stream to already molten salt in the crucible. The batch mode unit is now installed in the glovebox and can process 100 grams of the Pu-containing pyrolyzed ash per day, while the continuous mode unit can process 4-5 g ash/min, up to 1200 g/day during a 4 hr feed-time. The testing of both units in a non-radiological laboratory, as well as the installation and use of the batch unit in the glovebox to recover Pu-238, are discussed in this report.

METHOD

A schematic of a general MSO system is shown in Figure 1. The basic components of an MSO unit are as follows: 1) Feed system, 2) Air supply, 3) Downcomer/Feed tube assembly, 4) Crucible, 5) Secondary containment, 6) Off-gas conditioning and exhaust. A secondary metal vessel sits in an electric furnace and holds the crucible. The crucible contains the molten salt/ash mixture.
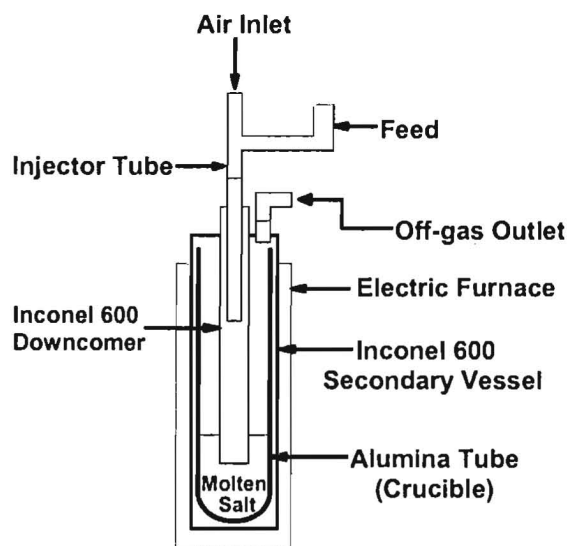

FIGURE 1. Schematic of Molten Salt Oxidation (MSO) Unit.

In the continuous feed unit, the organic feed is introduced by accelerated air flow through the injector tube into the molten salt. The feed tube is situated above the molten salt to reduce corrosion and clogging at the feed tube end. It is sheathed by the Inconel downcomer, which is inserted into the molten-salt bed an inch from the bottom of the crucible to maximize residence time in the salt. The downcomer creates a cooling blanket of air around the feed tube and protects it from corrosion.

During continuous processing, both air and feed are forced through the downcomer into the crucible where the molten sodium carbonate salt provides a hot matrix for the reaction. The oxidation reaction is most efficient at high temperatures (> 850°C) and up to 10 wt% sodium sulfate can be added to the salt to achieve a higher reaction rate (up to 10x) (Stelman, et al., 1976) and increased reaction efficiency.

Our particular needs required us to devise an MSO unit that would have a small footprint and fit into a space within a glovebox enclosure 1.1 m high that is occupied by a 33 cm high by 15.2 cm inner diameter tube furnace situated in a well. We must also work within the "safety envelope" of allowed actions in the glovebox. One limiting rule is that no manual activities can be done while any part of the unit is > 100°C.

Our goal is to recover plutonium oxide from the salt after the oxidation. The salt however retains several metal oxides generated by the corrosion of the metal pieces of the MSO unit but can contain up to 20 wt% of "contaminants" before it must be replaced. Our goal is to maximize the plutonium oxide concentration in the salt by minimizing the formation of less desirable other oxides.

One difficulty with the dissolution step of the recovery process is the volume of liquid generated from dissolving a kilogram or more of the metal oxide-salt cake. For example, the solubility of pure sodium carbonate is 29 g/100 ml and the volume required to dissolve 1 kg (~3.4 L for pure Na_2CO_3) of the less soluble salt-cake product presents significant storage and waste generation issues. The separation challenge is preceded by the need to remove the salt-cake from the crucible. Salt virtually is "cemented" to metal during the cooling process. Other workers removed salt via a vacuum transfer which "instantly" froze the salt, and did not stick to the transfer container. Vacuum transfer of molten salt is not within our safety envelope of activities in the glovebox.

In short, the technical challenges unique to recovering plutonium from our MSO process which we need to overcome are: 1) formation of insoluble metal oxide contaminants during the oxidation process, 2) generation of hazardous solution waste, and 3) mechanical removal of the salt-cake from the crucible to retrieve the plutonium.

PROJECT OVERVIEW

Our feed source is pyrolysis ash, which is generated when plutonium contaminated cheesecloth used for glovebox cleanup is heated to remove its hydrocarbon content. Pyrolysis ash is a light (64 kgs/m^3), flocculent material that retains some fibrous structure after ashing. These properties of the ash present problems during feeding and the small fibers bridge as the ash is moved by mechanical means.

Corrosion is one of the most challenging problems of MSO processing. The harsh alkaline, oxidizing, and high temperature environment in the salt is highly corrosive to metals and many ceramics. Corrosion products like iron, nickel and chromium oxides from "corrosion resistant" metal alloys contaminate the salt containing the desired plutonium oxide. Since the corrosion products shorten the lifetime of the salt and create difficulties in plutonium purification, selection of materials for the downcomer and crucible, which are in direct contact with the molten salt, is a subject of continuing research and testing.

Most MSO users employ larger units, which prevent them from using ceramic materials with more promising anti-corrosion characteristics. The choice of ceramics requires consideration of the thermal shock stability of the material as well as the corrosion resistance. For example, alumina has been shown to be very corrosion resistant to molten salt, but like many ceramics, it can break unpredictably when subjected to thermal gradients or thermal cycling. Therefore, we have experimented extensively with ceramics to find one suitable to our needs.

Our space considerations have dictated some modifications from the "classic" MSO unit design. The typical height of an MSO unit is based on experiments done by Rockwell, which studied the salt splash level as a function of air speed at the salt surface. This results in a unit ideally having a 6:1 ratio of height:diameter (Grantham et al, 1975) to minimize salt entrainment and clogging of the exhaust system. Our glovebox height is the limiting factor in the unit design, having a height of 1.1 m. The furnace sits in a well, which requires consideration of how to remove the unit from the furnace. It is also only 38.1 cm tall, which will expose part of the unit outside the furnace, another departure from previous MSO design.

HISTORY

Initially, development of the continuous feed unit was our entire focus. We built a prototype unit to establish the working parameters of the system. A bank of analytical units was integrated into the exhaust system to observe CO_2, CO, O_2 and total hydrocarbon. A larger unit was designed based on those studies (Figure 2), but developing a suitable mechanical feed system for delivering the ash remained an obstacle. While pursuing feed system development for the continuous unit, a batch mode unit was developed in order to determine if the process could be performed in a glove box and how the process would be affected by the thermal, oxidative, and radioactive properties of plutonium-238 oxide.

The main difference between the batch and continuous mode systems is how the feed material is introduced. The crucible of the batch mode unit is loaded with feed mixed with salt prior to heating the unit, while the continuous mode unit supplies a steady stream of feed via an accelerated air stream into the crucible after the salt is molten. The continuous feed is more complex to operate, but much more efficient and the continuous mode system can be operated without removing the salt until it has reached its contaminant limit of 20 wt% insoluble oxides. See Table 1 for a list of other differences. Our batch mode MSO unit began operation on July 1, 2003. To date (10/04), only 3 proof of principle runs have been performed using Pu-238 oxide contaminated ash.

Further modifications to the unit shown in Figure 2 have significantly shortened the overall height of the unit and fittings. The 2.5 cm OD braided steel feed tube was replaced with 1.9 cm OD tubing. The piping above the unit is

now significantly shortened. About 25 cm was removed from the height of the unit from that design change. Another 10 cm height was shortened by cutting down the height of the unit itself. The small screwfeeder needed to be replaced with a larger one having an agitator in the hopper to break up bridging.
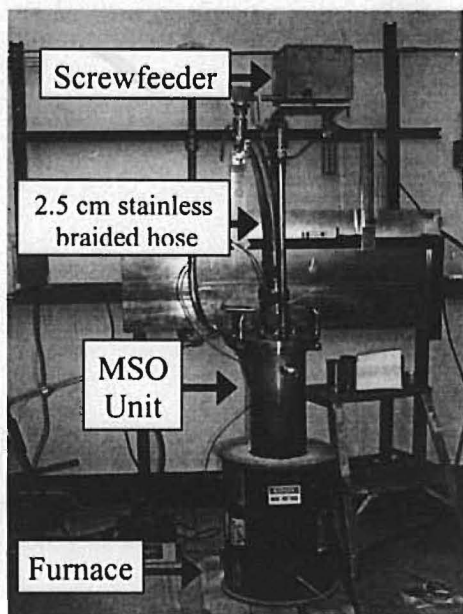

FIGURE 2. Continuous Feed MSO Unit. See Text above for further Design Changes from this to Present Unit.

RESULTS

Our goal is to develop an automated continuous feed unit that maximizes the rate of feed introduction into the molten salt and requires as a minimal amount of hands-on operator time. The feed system and containment materials

TABLE 1. Differences between Batch and Continuous Mode MSO Processing.

Characteristic	Continuous Mode Unit	Batch Mode Unit
Size	14 cm OD x 50.8 cm tall	14 cm OD x 43.2 cm tall
Feed mode	Mechanically metered by screwfeeder. Delivery into the melt by accelerated air stream created by the eductor.	Mix with salt before heating
Air supply mode	Compressed air accelerated by eductor into unit. Various pressures not exceeding 40 psi.	Compressed air at 20 psi controlled by flow meter.
Downcomer/Feed Assembly	3.8 cm OD outer tube (downcomer) almost to bottom. 1.9 cm feed air tube inside downcomer and 20.3 cm from bottom of the crucible	1.5 cm Inconel 600 sparge tube almost to bottom of crucible (no downcomer)
Air volume	1 -3 scfm	4-6 scfh
Throughput	950 g feed / run (4 hr). Reaction rate determined feed rate of 4 g/min (average).	50-90 g feed / run. Volume limited.

need to be optimized but the continuous feed unit is operable. Throughput, efficiency, and feed system studies have been performed. Space utilization of the continuous unit in the glovebox is being performed using a plywood mock up of the host glovebox space. The corrosion rates of many candidate ceramic materials combining thermal shock resistance and minimal corrosion have been determined.

The batch mode unit yielded a technique to remove the salt-plug from the crucible within the confines of a glovebox. We still plan to use the glovebox unit to develop a method to recover Pu-238 oxide from the salt and further study the effects of the presence of plutonium on the materials and reaction.

TABLE 2. Corrosion Rates in $g/(mm^2 \cdot day)$ for some Ceramic Materials.

Material	**1st week	2nd week	3rd week	4th week	5th week	Avg.
AlN	1.425E-05	9.570E-06	2.530E-05	2.482E-05	3.166E-05	2.112E-05
$Mg-ZrO_2$ sq	-5.592E-06	-2.400E-06	7.059E-07	3.986E-06	8.521E-06	1.044E-06
Al_2O_3 rod	***	***	***	1.456E-05	1.330E-05	1.393E-05
Al_2O_3 sq	***	***	***	1.128E-05	7.351E-06	9.317E-06

**Materials were kept at 950°C for a total of five weeks, with weights taken at the end of each week. Alumina coupons begun at Week 4.

Downcomer and Crucible Materials

A significant technical challenge for any MSO operation stems from the extreme corrosive capacity of the alkaline salt in an oxidizing environment at high temperatures. Extensive studies by Rockwell in the '70s showed a nickel metal alloy, Inconel 600, to have the best corrosion resistance for an easily obtainable commercial material (Newcomb, 1995). It has been used as a crucible as well as outer vessel on other units.

Three problems exist which preclude us from using an Inconel 600 crucible in our process: 1) The vacuum transfer step of the recovery process typically used in other MSO operations is not an option because safety concerns. 2) If slowly cooled, salt sticks like cement to a metal crucible. 3) Although Inconel 600 is corrosion resistant, it releases metal oxides into the salt, thereby decreasing the lifetime of the salt and increasing the need to remove more contaminants during plutonium oxide purification.

One solution is to develop a crucible from ceramic which can be more corrosion resistant than metals. In general, salt adheres less to the surface of ceramic materials and allows removal of the salt plug from a ceramic crucible (Figure 3). Also, the build up of corrosion products (e.g. alumina, zirconia, nickel oxide) in the salt is negligible. However, the fragility of these materials is a disadvantage and many ceramic materials are fail during thermal cycling. The thermal sensitivity of ceramics can decrease run time since they require slower temperature ramps.

Many ceramics were tested for corrosion resistance including the Inconel 600 for comparison (Newcomb, 1995). Some of our results are shown in Table 2. We chose to make aluminum nitride crucibles to test in our recovery process because aluminum nitride showed excellent corrosion resistance and ability to withstand thermal cycling. Casting the aluminum nitride into a suitably sized crucible was difficult because the material had to be cast in three pieces (2 tubes and a flat bottom, all three pieces 1.3 cm thick) which were cemented together. We have not yet tested the aluminum-nitride crucibles.

We originally used ceramic materials in both the crucible and downcomer/sparge tube, but because of its sensitivity to thermal cycling, the ceramic downcomer/sparge tube needed to be removed from the molten salt during the process. This operation violates our safety envelope for doing operations on hot objects.
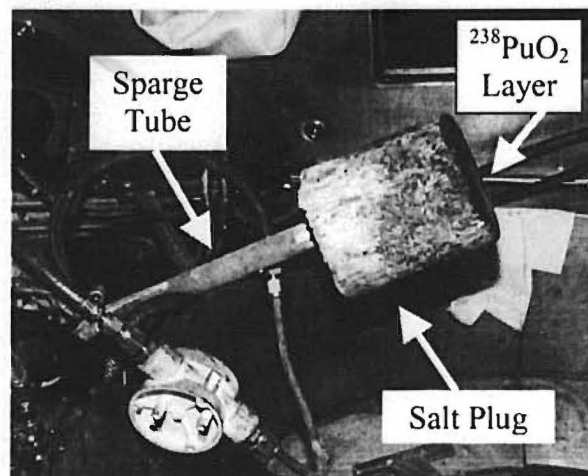
FIGURE 3. Sparge Tube is Used as a Handle to Remove the Salt from the Ceramic in a Single Plug. First Result of Batch Mode Run in a Glovebox using Pu-238 Contaminated Ash. Darkening on the Bottom is Layer of $^{238}PuO_2$

We switched to Inconel 600 sparge tube on the batch MSO instrument and discovered that the ash reaction went to completion faster and more completely than with a ceramic sparge. This rate increase may result from the inability of ash that froths to the top of the molten salt to adhere to the Inconel sparge tube sitting in and above the molten salt.

If the sparge tube was allowed to sit in the molten salt mixture as it was allowed to cool, we discovered that the metal sparge tube stuck to the salt plug like a popsicle stick and provided a handle to assist in pulling the salt plug from the crucible (Figure 3). The salt can be "wiggled" out of the ceramic crucible by using a little water to loosen the salt plug from the ceramic. The continuous feed unit also yields "popsicles".

Feed System

Two approaches were taken to solve the resistance of our main feedstock to mechanical metering. One was to mix an additive with the ash that would make it more amenable to mechanical feeding. For example, it is much easier to pump liquid into the system, so ash in a liquid matrix would make a better feedstock. Two organic liquids, ethylene glycol and mineral oil, were mixed with (non-rad) pyrolysis ash and tested. Another additive tested was a polystyrene polymer (PiccoTex) that was heated to 110°C to form a liquid in which the ash was suspended and the mixture solidified when cooled to room temperature. The resulting ash/plastic solid could be crushed into 0.3 cm pieces and fed easily by screwfeeder. Each additive tested allowed only a maximum of 10% by weight ash in the organic matrices. This dilution of the ash resulted in an unacceptable decrease of ash throughput in the continuous MSO unit and has the added disadvantage of increasing a worker's (radiation) exposure in the additional preparation time required.

The other option was to find a mechanical feeder that could feed ash. Screwfeeders abound, but tend to be too large to put into a glovebox. A small screwfeeder (AccuRate. See Figure 2.) was tested with several modifications to help the ash to fall in the path of the helix. Further searching identified stainless steel screwfeeders (K-Tron and Brabender) with features that effectively feed the pure ash. They both feature a stainless steel hopper having a rotating agitator, like that of a flour sifter, which breaks up the bridging and pushes the material into the screw

mechanism. A double screw more effectively "grabs" the ash and pushes it uniformly along the length of the helices. Vendor tests proved that the ash could be effectively moved at a minimum rate of 1 g/min (or faster) with sufficient accuracy for our purpose. We have not yet chosen one. Modifications may be made to customize them to our needs.

The remaining difficulties lie at either end of the feeder. First, the hoppers are too small to supply enough feed for an entire run. The material is light enough that the feed rate is affected by the volume of material in the hopper. Work presently is concentrated in these areas. Second, a Venturi nozzle is used to accelerate the material through the feed tube and into the salt. The device has a narrowing point that is a bottleneck where ash clogs. The effect manifests as small puffs of blowback. Further pressurization of the system would make it more difficult to work with in the safety envelope of the facility, but other modifications are being pursued.

Salt Composition

Commercial MSO operations use sodium carbonate because it is inexpensive. Sodium carbonate melts at 850°C. The disadvantage of using that salt is that the typical (compromise) reaction temperature range of around 900°C is not much higher than the melting point. During processing, temperature fluctuations occur due to opposing changes in air flow rate (cooling) and the exothermic oxidation reaction. In the past, these changes have been great enough to cause partial solidification of sodium carbonate salt during processing, which halts the run.

We employ a 40 mol% K_2CO_3 - 60 mol% Na_2CO_3 eutectic that melts at 706°C (Janz & Lorenz, 1961). In addition to having the lower melting point, the mixture has a higher solubility due to the contribution of the potassium carbonate, which is 3 times more soluble than the sodium salt (29 g/100 ml). We determined the solubility of the potassium-sodium mixture to be about 50 g/100 ml.

Second, a modeling study predicts that the increased neutron production associated with (α, n) reactions, as a result of the highly alpha-emitting Pu-238, is higher for sodium interactions and a factor of three lower for the 40 mol% potassium-60 mol% sodium mixture that we use (Shores, 2002). The predictions from these calculations have not been tested experimentally.

Sodium sulfate acts to increase the oxidation reaction rate by up to a factor of 10 when used in 1-10 wt% amounts in the primary salts (Stelman et al., 1976). We have shown that the reaction goes to completion more quickly and that CO_2 production is increased (compared to CO) with the addition of as little as 1% sulfate. The amount of sulfate is important in the case of the batch mode operation. The addition of sulfate guarantees that the reaction goes to completion within our run time. Without sulfate, the reaction rate "tails off" leaving a small layer of unreacted ash on top of the salt. This makes it impossible to remove the salt plug via the "popsicle stick" method.

Past work has shown that the sulfate has little effect on the reaction rate of easily oxidized material like LDPE (low density polyethylene). But ash is the least reactive material that we have tested. Experimentally, the addition of sodium sulfate during a continuous mode run increased efficiency (ratio of CO_2:CO in the off-gas increased from about 1:10 to about 1:30), but did not significantly increase throughput.

Size, Throughput, and Efficiency

MSO reactors can be any size. The available space in the glovebox is dictating the size of our unit. The height to diameter of crucible ratio for a unit is typically 6:1 (minimally 4:1) (Grantham, 1975). This parameter is based on the volume and velocity of air being bubbled through the salt and the amount of splashing and salt entrainment resulting from this activity. The oxidation reaction occurs in the frothing of the molten salt mixed with air, so factors affecting that are important for the success of the reaction.

Our batch unit uses a crucible that has a 10.2 cm OD and is 30.5 cm high to match the size of the furnace. The ash volume is limited since ash and salt must fit into the crucible before heating. The ratio of salt to ash is important to the success of the batch mode reaction. The ash/salt ratio affects the viscosity of the melt, which affects reaction efficiency. We found the salt:ash mix have an acceptable ratio of 15-25 g salt per gram ash (ave. 60 g) coupled with

an air flow rate of 4-6 scfh is sufficient to oxidize ash in the batch mode unit in 3 hrs while not inducing salt entrainment.

Those ash loading constraints are not an issue in the continuous mode unit. The amount of air vs. the height of splash then becomes important. Our unit has a ht:diameter ratio of 4:1. Salt entrainment has been decreased by the use of a triple-layer baffle followed by an expansion trap in the exhaust line. A row of small holes has been drilled around the bottom of the downcomer. The effect is to produce smaller bubbles, which create greater reaction surface area and decrease splash height. Also, smaller bubbles subject the crucible to less mechanical force. These observations were made using of a zinc chloride solution (at room temperature) made to the density of molten salt. The liquid was placed clear plastic crucible-sized container and the feed/air part of the unit used to deliver air.

The other factor affecting splash height is the effect of exposure of half of the unit outside of the furnace. It greatly increases heat loss. This requires stricter balance of the reaction (feed) and air flow rates to stabilize temperature. The exposed part of the unit has the effect of reducing the kinetic energy of the salt sufficiently that it deposits near the top of the crucible, reducing salt escape into the line, but increasing the risk of clogging at the baffle and exhaust. We are experimenting with the effects of added insulation or heating around the exposed area to adjust these effects to our advantage.

Our goal is maximum throughput. Therefore, reaction efficiency (maximum CO_2, minimum HC and CO) can be sacrificed, within the envelope of our air quality emission standard. The greatest amount of carbon monoxide off-gas produced was in the range of 1%. Over a 4-8 hour processing period per day, this is an acceptable level. Our levels of hydrocarbon produced by ash processing in the continuous mode are at 200 ppm or lower. In the batch mode, hydrocarbons come off the ash (93% C, 4% H, 3%O) in a concentrated manner during the heating of the salt/ash mixture under argon. With the addition of a simple 4 M NaOH scrubber, the hydrocarbon content of the final exhaust is reduced to by a factor of 4 after scrubbing.

The continuous mode unit reaches peak efficiency at the lowest feed rates, about 1 g/min, as measured by the total hydrocarbon present in the off-gas (< 50 ppm) relative to CO_2 production. Since hydrocarbon production is not a significant output, the feed rates are dictated primarily by air flow rate. Results of 4 test runs showed: 1) Addition of ash at > 6 g/min resulted in higher rates of clogging at the eductor. 2) Air flow rates of > 2.1 scfm resulted in excessive splashing and clogging at the exit. (Total air throughput was about 3 scfm from suction created by Venturi). An average of 4-6 g/min ash at 2.1 scfm was sustainable for 2 hrs. Production of CO2 and consumption of oxygen were about 10%, CO production was 10% or less of the CO_2. Under those conditions, it was possible to maintain a temperature balance (850-880 °C) between the reaction heat and heat loss from the exposed portions of the unit, although that is still under testing with more insulation of the exposed part.

Plutonium Recovery and Waste Disposal

Several experiments were performed to develop the method for $^{238}PuO_2$ reclamation from salt. First, we have shown from the batch process (Figure 3) and from the results of other workers (Bird, 1994) that the $^{238}PuO_2$ layers on the bottom of the crucible, therefore, the entire salt plug does not have to be dissolved to reclaim the $^{238}PuO_2$. Second, the solubility of the Na_2CO_3/K_2CO_3 eutectic mixture is greater 40% higher than Na_2CO_3 alone, which reduces the amount of liquid waste generated by this process. Finally, using a ceria surrogate mixed with spent salt, the ceria and almost all of the other contaminant metal oxides proved to be completely insoluble and easily filtered from the salt solution. This impure plutonium oxide is the base material for further purification.

This is a waste minimization process, so attention to production of further waste has been scrutinized. The sources of waste are as follows: 1) broken crucibles, 2) spent salt, 3) salt solutions. We are addressing each of these streams with the view of minimizing their impact. We continue seeking a ceramic material that will survive multiple thermal cycles, not only to reduce waste but save time and increase worker safety. We have chosen to use a metal downcomer/sparge tube which will contribute to the metal oxide content of the salt. This will shorten the lifetime of the salt, but enhances worker safety. It is the most corrosion resistant choice and is estimated to last for a year or more. The salt must be replaced when the contaminant content reaches 20 wt%. Since the plutonium is the major contributor and is removed as it builds up from the bottom of the plug, the salt life will be extended. Further, if required, the salt can be recrystallized to further extend its life. The salt solutions may contain chromium and

aluminum. Residue processing (precipitation) to remove contaminants like these is already routinely performed on all solutions before being sent to our liquid waste treatment facility.

CONCLUSION

A batch mode MSO unit is operational inside a Pu-238 glovebox. The operating parameters have been optimized for maximum throughput and minimum waste. The unit can process up to 100 g of Pu-238 contaminated pyrolysis ash per run.

Work continues on the development of a continuous feed unit. We have a mechanical feed system and have run continuously for up to two hours at a feed rate of 4-6 g/min. A conservative estimate for a single run is four hours, yielding approximately 1200 g of ash processed per run. Other advantages are: 1) There is no need to remove the salt each day, resulting in much lower operator exposure. 2) The furnace does not need to be completely cooled between runs, giving more run time per day.

There are still issues to resolve. The feed system is still not very robust. The eductor can clog if the feed rate is uneven and feed rate is not well controlled. Second, we are still testing materials for a robust containment of salt. We have procured two aluminum nitride crucibles that still must be tested. The crucibles were not easily manufactured, so their performance in this configuration is important. New materials are still being corrosion tested.

The continuous feed unit requires more specialized equipment and more extensive testing to assure the safety of the operation in a glovebox. Continuous testing of the operation is needed to learn the long-term effects of the salt on the containment and downcomer, especially in the case where the unit is not completely cooled between runs. This savings in cooling in reheating would significantly increase processing time.

ACKNOWLEDGMENTS

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