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# PYROCHEMICAL RECOVERY OF URANIUM FROM MONOLITHIC U-10MO FUEL SCRAP

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#### ABSTRACT

The NNSA Global Threat Reduction--Conversion program is currently engaged in the development of a nuclear fuel that would enable conversion of US high-performance research reactors from high-enriched uranium to low-enriched uranium alloy fuel, monolithic uranium-10wt% molybdenum (U-10Mo) bonded to aluminum cladding. Approximately 2400 kg of the initial U-10Mo fuel meat used to create fuel foils for the five U.S. high-performance research reactors will be recycled annually. A design concept has been developed for a pyrochemical processing system to recover low enriched uranium from the U-10Mo fuel scrap. A description of the system and a design concept for the proposed scrap recovery line is presented. Results from successful electrorefining experiments with U-10Mo alloy and U-10Mo with zirconium bond are discussed.

#### 1. Introduction

The NNSA Global Threat Reduction--Conversion program is currently engaged in the development of a nuclear fuel that would enable conversion of US high-performance research reactors from high-enriched uranium to low-enriched uranium alloy fuel, monolithic uranium-10wt% molybdenum (U-10Mo) bonded to aluminum cladding. Approximately 2400 kg of the initial U-10Mo fuel meat used to create fuel foils for the five U.S. high-performance research reactors will be recycled annually.[1] The U-10Mo recycle stream is a mixture of materials including casting scrap from the uranium casting process, shearing losses from foil sizing, rejected fuel elements, and material from holdup in crucibles and other contaminated equipment. A design concept has been developed for a pyrochemical processing system to recover low enriched uranium from the U-10Mo fuel scrap. This study consists of two major activities, flowsheet development and demonstration of process feasibility. A description of the process chemistry including a mass balance flowsheet that identifies and quantifies product streams and waste streams, processing equipment, and the approximate footprint of the proposed scrap recovery line is presented. Electrorefining experiments performed at the laboratory-scale with U-10Mo alloy and U-10Mo with zirconium bond are also discussed.

#### 2. Process Flowsheet Development

The flowsheet for recovering uranium from U-10Mo fuel scrap consists of head-end processes to prepare the scrap for subsequent treatment by pyrochemical processes that recover and prepare the uranium for recycle to fuel fabrication. There are two head-end processes; clad material is treated by aqueous methods to remove the aluminum cladding from the foils and all foils are chopped or shredded to expose the fuel meat and size it to fit in the electrorefiner anode baskets. After head-end treatment, the fuel scrap is treated by electrorefining to recover the uranium. Distillation removes the residual salt from the uranium after it is removed from the electrorefiner and prior to it being consolidated into ingots. Distillation is also used to recover the residual salt from the Mo and Zr scrap before they are prepared for disposal.

## Mass Balance

A simplified schematic of the conversion process and mass balance is shown in Figure 1. The mass balance flowsheet, designed to treat 2000 kg of LEU, has a theoretical uranium product recovery efficiency from the U-10Mo scrap of >99 wt%. The waste generated from the process is composed of aqueous waste, Mo/Zr metal scrap, and damaged fuel baskets and crucibles that cannot be recycled. The LiCl-KCl molten salt is continually recycled from the uranium consolidation and salt distillation operations to the electrorefiner. Impurity build-up in the LiCl-KCl salt would be slow so that salt purification would not be required until after several years of normal operation. A detailed criticality safety analysis was not part of this study. All uranium-bearing waste streams could be accumulated with the down blending and casting hold-up scrap that could not be successfully removed from the crucibles and potentially be recycled by other separation technologies. The assumptions used in developing the mass balance flowsheet are given below.

- General Assumptions
  - Facility designed for 2000 kg LEU
  - o Facility operated 200 days/yr
  - Fuel baskets and crucibles require annual replacement
- Feed Streams
  - o 20 wt% of down blending and casting scrap recovered from crucibles
  - $\circ$  Thickness of Zr layer estimated to be 25  $\mu m$  (0.001 inches)
- Head-End Process
  - o No U-10Mo losses with hardware waste stream
  - o Complete removal of aluminum cladding by aqueous dissolution
  - o Bulk packing density of chopped fuel in baskets is estimated at 2.5 kg per liter
- Pyrochemical Operations
  - o Electrorefiner
    - Feed alloy contains no oxides
    - Uranium retention in baskets estimated at 0.5 wt%

- Mo and Zr completely retained within baskets
- Complete recovery of uranium metal from cathode
- Residual salt on products estimated at 20 wt%
- o Uranium Consolidation
  - Complete separation of uranium metal from salt
  - Uranium retention in crucibles estimated at 0.1 wt%
- o Salt Distillation
  - Complete separation of U-metal and Mo/Zr metal from salt



Figure 1: Mass balance flowsheet for uranium recovery process.

### **Feed Materials**

The monolithic fuel fabrication process produces two distinct feed streams with respect to the pyrochemical recycling process: aluminum-free scrap and aluminum-clad foils. These two streams are composed of residuals from the down blending and casting process, shearing shards from foil sizing, Zr coated foils, failed fuel plates, rejected fuel elements, and material hold-up from contaminated equipment. The distinctive characteristics, specific quality assurance failures, and any pre-conditioning steps required prior to pyrochemical processing are described for both streams. Overall 85 wt% of the material available for recycle is assumed to be treated by pyroprocessing. Uranium could be recovered from the remaining material but the material would need to be treated by other methods prior to pyrochemical methods.

Stream	Description	LEU Available for Recycle	LEU Treated by Pyro
Al-Free Scrap	Casting Hold-up	446	89
	Rolling / Shearing	735	735
	Zr-Coated	566	566
	Al-Free Total	1747	1390
Al Clad Foils	Plates	321	321
	Elements	289	289
	Al Clad Total	610	610
Combined	Total	2357	2000

Table 1. Feed streams to pyrochemical uranium recovery process.

## <u>Aluminum-Free Scrap</u>

The aluminum-free scrap feed stream is composed of down blending, casting, rolling, shearing, and Zr-coated scrap that are recovered prior to the aluminum cladding step used to create fuel plates. Overall, the aluminum-free scrap feed stream makes up 59 wt% of the material available for recycle. Rolling and shearing scrap make up the largest fraction of the aluminum-free scrap and has visible surface deformities or is leftover shearing pieces that do not meet foil Zr-coated scrap has irregular boundary diffusion layers that failed quality specifications. assurance inspections. Down blending and casting scrap is the hold-up that is strongly bonded to process crucibles or has failed to meet feed specifications. The hold-up must be treated by physical methods to release the alloy from the crucibles. Due to the complexity of harvesting the down blending and casting scrap an extremely conservative estimate was made that only 20 wt% of this stream will be recovered from the crucibles and treated by the proposed pyrochemical process. Investigation into the strength of the bond between the alloy and the crucibles along with exploration of mechanical techniques required to harvest the hold-up is needed to determine the amount of casting scrap that could be recovered for subsequent pyrochemical treatment. Further research and development of harvesting techniques could lead to significantly higher uranium-recovery fractions.

#### Aluminum Clad Foils

The aluminum-clad foils are a combination of the plates and elements that have failed a variety of quality assurance inspections such as the presence of surface defects, dimensional irregularities, or abnormalities in the aluminum clad. This stream accounts for 26 wt% of the overall material available for recycle and will first require disassembly of the element hardware. Following disassembly, the plates will undergo a chemical-decladding treatment to remove the Al before being sent to the chopping or shredding head-end process.

#### **Head-End Process**

#### Chopping or Shredding

Chopping or shredding mechanical manipulations are used on all the feed material to meet the size requirements of the electrorefiner fuel baskets and to expose the inner U-10Mo fuel meat within Zr-coated foils. The maximum fuel meat thickness from the five U.S. research reactors is anticipated to be 760  $\mu$ m (0.03 inches),[1] which would be easily converted into acceptable feed by a commercial chopper. The chopping or shredding equipment would be operated within an inert atmosphere to eliminate any possibility of oxide formation on the freshly exposed metal.

#### Chemical Decladding

Aluminum-clad foils are treated with a chemical decladding process that uses an alkaline solution of 10% NaOH to dissolve the Al.[2] The aluminum clad is converted into a NaOH/NaAlO<sub>2</sub> solution and collected as aqueous waste. The aluminum must be removed from the feed stream to avoid competing electrochemical reactions within the uranium electrorefiner.

## **Pyrochemical Operations**

#### **Electrorefining**

Uranium electrorefining, and the associated process chemistry needed to yield a purified uranium product, is an established technology that has been demonstrated for a variety of programs including the Integral Fast Reactor Program and treatment of spent fuel from the Experimental Breeder Reactor II.[3, 4] Electrorefiners, such as the planar electrorefiner being developed at Argonne, have demonstrated capability to meet the annual throughput requirements of the U-10Mo scrap recovery process.

Electrorefining is an electrochemical process used to produce a purified metal from an impure feed material. In the case of uranium, it is performed in a molten salt such as LiCl-KCl eutectic at 500°C. The operating conditions of the electrochemical cell are controlled to only allow the transport of uranium from the anode baskets to the cathode. As electrical current is applied to the cell, uranium in the anode fuel baskets is oxidized to trivalent uranium ions that dissolve into the molten salt. Simultaneously, trivalent uranium ions in the molten salt are reduced at the cathode yielding a metallic uranium deposit. The uranium metal that is deposited on the cathode is harvested and sent to a salt distillation and consolidation process. The zirconium and

molybdenum present in the fuel are electrochemically less reactive than the uranium and remain in the fuel baskets. They are mechanically recovered from the fuel baskets after the electrorefining process and sent to waste after residual salt is removed by distillation. Previous experimental work with U-10 wt% Zr fuel coupled with the knowledge that Mo is nobler than Zr, suggest that higher operating potentials can be used for the U-10Mo scrap, which will lead to better separations. All product transfers from the electrorefiner can contain up to 20 wt% residual chloride salts; hence the need for distillation. The recovered salt is recycled to the electrorefiner.

The electrorefining process is performed in a furnace well contained within an inert atmosphere glovebox. The system is sized to accommodate approximately 55 kg of scrap alloy per loading. An operations schedule that would meet the desired throughput requirement of 10 kg of LEU treatment per day is: one day for fuel basket loading, three days for U electrotransport, and one day for product recovery plus cell clean-up. However, the electrorefiner design can be easily modified to accommodate different batch sizes and operational schedules.

The fuel baskets would be constructed of stainless steel sized to contain approximately 23 liters of material (assuming a conservative bulk density of 2.5 kg per liter). The original monolithic U-10Mo fuel matrix can have uranium densities up to 16.5 kg U per liter but the packing density within the fuel baskets will be considerably lower.[5] A set of six baskets with a cross section of 24" wide x 1" thick x 10" (61.0 cm x 2.5 cm x 25.4 cm) tall would contain this material. The basket dimensions are limited to 10" (25.4 cm) tall to simplify the product loading and the harvesting process. The six-basket set has an estimated empty mass of 55 kg and a fully loaded mass of 110 kg with the U-10Mo alloy scrap. It is estimated that the fuel baskets will require annual replacement. The overall basket assembly footprint would be approximately 2 ft x 2 ft (0.6 m x 0.6 m) and fit within the electrorefiner vessel opening, which is sized to be 3 ft x 3 ft (0.9 m x 0.9 m).

## Uranium Consolidation

Uranium-product consolidation, also known as cathode processing, is required to remove residual salt that adheres to the uranium dendrites and consolidate the uranium into an ingot.[6, 7] In current practice, the uranium metal coated with residual salt is recovered from the cathode of the electrorefiner cell and placed into a crucible that is installed in an induction furnace. The furnace is first evacuated to a slight vacuum (e.g., 50 torr) and then heated to 850°C to allow the salt to distill from the uranium dendrites. This salt is collected in a condenser and eventually recycled to the electrorefiner system. The crucible is then heated to 1200°C to melt the uranium product into an ingot. Melting the uranium into an ingot is required to ensure that any residual salt within the dendrites is released so that purity specifications are met for fuel fabrication. After cool-down, the uranium ingot is removed from the crucible and recycled to fuel fabrication.

The principle components of the cathode processor include a stainless-steel vessel, the loading/unloading trolley, an induction furnace, vacuum pumps, and the condenser assembly. Many of these components are commercially available and require few modifications before installation. The equipment is sized to process up to 50 kg of U per batch. The primary crucible is constructed of hafnium nitride coated niobium (empty mass of approximately 90 kg).

Although it is estimated the crucible will be replaced annually, process experience may show that it can be used for longer periods. The overall footprint of the cathode processor is approximately 8 ft x 16 ft (2.4 m x 4.9 m). Due to this large footprint and the desire to maintain low glovebox capital costs, only the loading and unloading of the cathode processor would be performed in the inert glovebox. A trolley rail system would be designed to deliver the crucible to the consolidation vessel while maintaining an inert atmosphere. The consolidation equipment would be located next to the glovebox to minimize the distance that the crucible must travel.

### Salt Distillation

Removal of residual salt from the molybdenum and zirconium is accomplished by the same method as that for uranium. The salt-coated molybdenum and zirconium scrap is harvested from the fuel baskets and transferred into a crucible, which is designed for use in an induction furnace. The furnace is evacuated to a slight vacuum (e.g., 50 torr) and the crucible is heated to 850°C to allow the salt to distill. The salt is collected in a condenser assembly and eventually recycled to the electrorefiner system. After the salt is distilled, the remaining Mo/Zr scrap is cooled to room temperature, collected from the crucible, and packaged for disposal.

The principle components used to distill the chloride salt are identical to the equipment required for the uranium product consolidation. The difference between these two operations is shorter processing time, as the Mo/Zr scrap does not require consolidation into an ingot. Operational scheduling would be designed to perform both the salt distillation and the uranium consolidation operations within the same piece of equipment. Sharing the equipment greatly reduces the facility footprint and capital cost. The use of alternative technologies, such as a commercially available rotary kiln, could distill the chloride salts from the Mo/Zr scrap. Rotary kilns are continuous operations that can treat smaller amounts of material per unit time while still achieving the desired throughput and potentially have an advantage over batch processors. However, use of a rotary kiln for the Mo/Zr would increase the facility footprint.

## **Facility Description**

The recovery facility would be composed of a scrap-preparation area, a pyrochemical glovebox/ uranium consolidation area, and a storage area. The size of the overall facility would depend on storage requirements as well as the design of scrap-preparation area. Figure 2 shows a conceptual facility design that accommodates all functions and is 36 ft x 44 ft (11 m x 13.5 m)--1584 ft<sup>2</sup> (147 m<sup>2</sup>). An overhead bridge crane would be shared across the facility and equipped to handle equipment installations and product transfers.

## Feed Preparation Area

The feed-preparation section of the facility is 28 ft x 17 ft (8.5 m x 5.2 m). The preparation area has three stations - crucible hold-up recovery (8 ft x 10 ft—2.3 m x 3. m)), chemical decladding (10 ft x 10 ft—3 m x 3 m) and plate / element disassembly (10 ft x 10 ft-3 m x 3 m) operations. The area also includes two hoods that are available for analytical or chemistry support work. Future research and development on scrap preparation specifically U-10Mo recovery from crucibles would help to provide a more detailed design of this area.



Figure 2: Conceptual facility layout for uranium recovery process.

## Pyrochemical Glovebox and Consolidation Area

The pyrochemical glovebox and consolidation area is compactly contained with a 24 ft x 20 ft (7.3 m x 6.1 m) area (Figure 3). This area contains an inert glovebox, consolidation processor, glovebox atmosphere purification system, and system control panel. Additional equipment located in this area includes: power supplies, material transfer systems, and vacuum components. The glovebox is fitted with a specialized trolley system designed to deliver the U dendrites or Mo/Zr crucible to the consolidation vessel while maintaining an inert atmosphere. Due to the large footprint of the consolidation system and the desire to reduce the glovebox manufacturing

costs, this design would only house the loading and unloading of crucibles within the glovebox. The trolley would utilize glovebox transfer locks to transfer the loaded crucible to the induction furnace and return the uranium ingot or cleaned Mo/Zr scrap along with the salt condenser after the cycle is completed.

### Pyrochemical Glovebox

The inert atmosphere glovebox shown in Figure 4 is 16 ft long x 8 ft wide and contains four distinct areas; (1) chop/shred, (2) electrorefining, (3) crucible preparation for product consolidation, and (4) storage. The chop and shred area is one station, 4 ft x 4 ft (1.2 m x 1.2 m), that is used to cut the feed alloy into a size suitable to fill the electrorefining fuel baskets. The chopping operation is performed in an inert atmosphere to eliminate any possibility of oxide formation on the metal scrap. The electrorefining area requires four separate window stations, each 4 ft x 4 ft (1.2 m x 1.2 m). These stations comprise the heated salt well, fuel-basket loading, basket harvesting/cleaning, and uranium-dendrite harvesting. The product consolidation area is made up of two stations used for crucible preparation and salt harvesting. The crucible preparation station is used to load uranium dendrites into the consolidation trolley and to harvest uranium ingots. Additionally, this station is used for Mo/Zr crucible loading and unloading. The salt harvesting station is used to recover the electrorefiner salt, distilled from uranium or Mo/Zr, from the induction furnace condenser. The final station in the glovebox is dedicated for shared storage of fuel baskets, crucibles, electrorefiner salt, uranium ingots, and tools. Ingots formed in the cathode processor are stored in wells within this station prior to release to fuel fabrication. An overhead bridge crane is shared across the glovebox stations and equipped to handle both equipment installations and product transfers. The glovebox would be maintained at negative pressure relative to the facility to protect workers from possible uranium dust releases. The glovebox atmosphere would be maintained at low (ppm) oxygen, moisture and nitrogen levels using a dedicated purification system.

	Contro			
	16' 0''	Consolidation Equipment Uranium Consolidation and		
Pyrochemica & Consolida 24' 0'' x	al Glovebox tion Area 20' 0"			
				Salt Distillation
Chop / Shred Foils	Storage U-Mo Salt	Basket Harvesting / Cleaning	Crucible Preparation	8' 0" x 16' 0"
Basket Loading	Electro- refiner	Uranium Dendrite Harvesting	Salt Harvesting	Purification System 8' 0'' x 4' 0''

Figure 3: Pyrochemical glovebox and consolidation area within recovery facility.

Chop / Shred Scraps	Image Nb   Storage   U-10Mo   Salt	Basket Harvesting / Cleaning	Nb Crucible Preparation C
Basket Loading	Electrorefiner	Uranium Dendrite Harvesting	Salt Harvesting

Figure 4: Glovebox layout for pyroprocessing equipment.

### 3. Process Feasibility Tests

Research and development studies at the laboratory- and engineering-scale provide valuable data that are used to develop detailed equipment designs and optimize the facility layout. Laboratory-scale electrorefining experiments using U-10Mo fuel are necessary to confirm process viability and provide preliminary information that can be used to refine throughput calculations and determine equipment size.

## **Experimental Setup**

The electrorefining experiments were performed in a furnace well contained within an inert atmosphere (helium) glovebox maintained below 5 ppm oxygen. The laboratory-scale system, shown in Figure 5, was sized to accommodate 200 ml of LiCl-KCl-5wt%UCl<sub>3</sub> molten salt in the primary crucible. Anode baskets were constructed of stainless steel and sized to contain up to 10 grams of U-10Mo feed. The tests were performed on uncoated U-10Mo foil and also on a Zr-bonded foil. A stainless steel rod served as the cathode on which the metallic deposit collected. A new anode and cathode were used for each test to reduce the risk of cross contamination between tests. An external power supply was connected to the cell to provide the current required for the refining process. Ceramic oxide tubes were used on the main process crucible and the electrodes to maintain electrical isolation from the glovebox. A Ag/AgCl reference electrode was used prior to experimentation to verify cell conditions prior to testing. A K-type thermocouple was used to record the bulk salt temperature. The cell components and electrolyte were allowed to equilibrate at temperature prior to passing current.



Figure 5. Laboratory-scale electrorefining test cell.

#### **Results and Discussion**

Test 1 was performed at  $510^{\circ}$ C on a 1 gram U-10Mo sample using potentiostatic control for the cell. The potential was maintained at -1 V cathode versus anode for the duration of the two-day test. Tests performed on uncoated U-10Mo foils maintained a cathode versus anode potential more positive than -1.2 V to ensure that the Mo remained within the anode basket. The average current observed for the first day of testing was 100 mA and for the second day the current dropped to an average of 85 mA. After the test was completed, the anode basket and the cathode rod were raised from the salt into the heated space (480°C) above the process crucible to allow the electrolyte to drain. Figure 6 displays the anode basket and the cathode products collected from Test 1.



Figure 6. Anode basket (left) and cathode (right) after electrorefining test.

Test 2 was run on a 1.1 gram U-10Mo Zr-coated sample using an experimental configuration identical to Test 1 and completed at  $510^{\circ}$ C. The average potential was maintained at -0.4 V cathode versus anode for the duration of the two-day test. Tests performed on the Zr-coated foils were maintained at a potential more positive than -0.5 V to keep Zr from being oxidized into the molten salt and deposited on the cathode. The average current observed for the first day of testing was 100 mA and for the second day the current dropped to 20 mA.

Test 3 was performed at the slightly higher operating temperature of  $561^{\circ}$ C. The higher temperature was used to determine if the anode basket contained less salt, after draining above the process crucible, at the end of the test. A 1.1 gram U-10Mo Zr-coated sample contained in a new anode basket was used as the feed material. The average potential was maintained at -0.4 V cathode vs. anode, which yielded an average current of 60 mA. The samples collected at the end of the run appeared to retain similar amounts of salt as compared with the first two tests performed at  $510^{\circ}$ C.

Test 4 was also run at  $561^{\circ}$ C with a 1 gram U-10Mo sample, but this test was conducted under galvanostatic control of the cell. The average current was 125 mA and that current maintained an average cathode vs. anode potential of -0.33 V.

Samples of the dendrites, the entire anode basket, and the salt electrolyte were collected for chemical analysis. Preparation of the samples for analysis involved washing the dendrites and the anode basket with water to remove the salt soluble components. The remaining solid anode residues and the metallic dendrites were analyzed using ICP/OES to identify the composition of the samples.

Analysis of the dendrites revealed that only uranium metal deposited on the cathode rods in each of the tests. The Mo and Zr weight fractions in four of the five samples analyzed had values below detection limits for the analytical technique (calibrated to <0.05%). The only exception to these data was in Test 2 where the first dendrite sample submitted from the test at the end of the first day showed a slight amount of Mo present in the deposit. The second dendrite sample submitted at the end of the test from the same cathode however, did not indicate any presence of Mo in the deposit. Furthermore in order to have deposited any Mo at the cathode, potentials would have been such as to have first deposited the Zr from the foils as well. Clearly, the analytical results from this particular test are spurious, since no Zr was detected.

Analysis of the residue recovered from the anode revealed that the final composition of the feed materials ranged greatly from complete removal of the uranium leaving only Mo and Zr to only partial anodic dissolution of the uranium. These results are not surprising because the tests were run for different periods of time and they were not designed to evaluate process efficiency. Engineering-scale tests, that use larger samples, will be conducted to evaluate process efficiency as well as product quality.

Analysis of the electrolyte salt samples submitted for Test 2-4 revealed that only U was present in the salt. This result confirms that the Mo and Zr in the foils were not oxidized at the anode and dissolved into the molten salt. If the cathode vs. anode voltage was not properly controlled in each of the tests, first Zr and then Mo would have dissolved into the electrolyte and deposited at the cathode.

It is important to note that the laboratory-scale tests were performed to prove the feasibility of the process hence, current density and current efficiency estimates were not calculated from the test data. Electrode spacing, cut-off voltages, and product recovery methods were not optimized in these tests. The data from the laboratory-scale tests along with previous electrorefining knowledge will ensure that the engineering-scale experiments are designed to evaluate these questions.

## 4. Summary

The facility envisioned for pyrochemical treatment of U-10 wt% Mo fuel scrap would recover 2000 kg of U annually from the scrap material. Electrorefining is the main pyroprocess needed to recover the uranium for recycle to fuel fabrication. Experience in recovering uranium from U-10wt% Zr fuel indicates process viability for U-10Mo fuel treatment. In fact, we expect a higher

degree of U recovery from the U-10Mo fuel because of the difference in electrochemical properties between Mo and Zr. The uranium product from the refining process is consolidated into an ingot, after residual salt is removed, and recycled to fuel fabrication. Mo/Zr scrap recovered from the fuel baskets is prepared for disposal after residual salt is removed from the surface by distillation. The electrorefining and salt distillation / uranium consolidation processes have been developed to an engineering-scale for a similar application but they require development at the scale proposed by this study.

A theoretical mass balance flowsheet was developed for the pyrochemical treatment process. Uranium recovery was calculated to be at >99% efficiency for the proposed process. The waste streams created in this process consist of approximately 29 kg of Al in NaOH/NaAlO<sub>2</sub> waste and 356 kg Mo/Zr scrap per year. Additionally, stainless-steel fuel baskets (55 kg) and Nb crucibles (180 kg) that are estimated to require annual replacement, will contribute to the waste generated. The amount of uranium retained on the discarded baskets and crucibles is conservatively estimated to be 12 kg annually. All uranium-bearing waste streams generated could be accumulated with the remaining down blending and casting scrap and be recycled by other separation technologies. Investigation of the harvesting techniques could lead to significantly higher percentages of feed materials that could be sent to pyrochemical treatment, however, the capital and operating cost of the recovery process would have to be less than the economic discard limit for the uranium.

A conceptual facility-layout was developed for the scrap recycling process. The overall facility area was approximately 1600 ft<sup>2</sup> (150 m<sup>2</sup>). Refinement of equipment design and placement would optimize the facility space and could lead to a reduced facility footprint. The pyrochemical glovebox and consolidation area is compactly enclosed within a 500 ft<sup>2</sup> (46 m<sup>2</sup>) area. Sharing the processing equipment for both the uranium consolidation and salt distillation operations was key to maintaining a small footprint. The use of alternative technologies, such as a commercially available rotary kiln that could distill the chloride salts from the electrorefiner products, may be able to treat the material and still achieve throughput requirements but would come at added facility footprint.

Electrorefining feasibility studies demonstrated that uranium could be recovered from the U-10Mo fuel scrap and the zirconium-bonded U-10Mo fuel scrap. Chemical analysis of the samples confirmed that the cathode deposits were entirely composed of uranium metal. Additionally, analysis of the salt samples revealed no Mo and Zr impurities were present in the molten salt electrolyte. Analysis of samples removed from the anode revealed that the Mo and Zr were contained within the anode basket. Engineering-scale electrorefining experiments are planned to investigate and optimize uranium recovery and provide design data for a prototype system. Coupled testing of the electrorefining and distillation/consolidation processes would define the amount of uranium retained in the system and provide additional insight into the amount of waste generated by the process. Validated electrorefining and uranium consolidation experiments would also provide impurity and chloride content data for the uranium ingots, which is important for fuel fabrication.

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#### 6. References

- [1] Daniel M. Wachs, Curtis R. Clark, and Randall J. Dunavent, "Conceptual Process Description for the Manufacture of Low-Enriched Uranium-Molybdenum Fuel," INL/EXT-08-1340, Feb 2008.
- [2] Moore et al., "Monolithic Fuel Fabrication Process Development at Idaho National Laboratory," 30<sup>th</sup> International Meetings on RERTR, Washington, D.C., October 5-9, 2008.
- [3] J. E. Battles, W. E. Miller, E. C. Gay, "Pyrometallurgical Processing of Integral Fast Reactor Metal Fuels," 3rd International Conference on Nuclear Fuel Reprocessing and Waste Management, Sendai (Japan), Apr 15-18, 1991.
- [4] E. C. Gay, W. E. Miller, J. J. Laidler, "Proposed High Throughput Electrorefining Treatment for Spent N-Reactor Fuel," DOE Spent Nuclear Fuel & Fissile Material Management, Embedded Topical Meeting, Reno, NV, June 16-20, 1996.
- [5] P. Lemoine, Daniel M. Wachs, "High Density Fuel Development for Research Reactors," Proceedings of GLOBAL 2007 - Advanced Nuclear Fuel Cycles and Systems, Boise, ID, Sept 9-13, 2007, page 8.
- [6] B. R. Westphal, J. R. Krsul, and D. W. Maddison, "Molten Salt Separation form Uranium During the Processing of Spent Nuclear Fuel," TMS 2002: 131st Annual Meeting and Exhibition, Seattle, WA, 2001.
- [7] A. Brunsvold, P. Roach, B. Westphal, "Design and Development of a Cathode Processor for Electrometallurgical Treatment of Spent Nuclear Fuel," Proceedings of ICONE 8: 8<sup>th</sup> International Conference on Nuclear Engineering, Baltimore, MD, 2000.