

**RERTR 2011 - INTERNATIONAL MEETING ON
REDUCED ENRICHMENT FOR RESEARCH AND TEST REACTORS**

**October 23-27, 2011
Marriott Santiago Hotel
Santiago Chile**

**GTRI PROGRESS IN DEVELOPING PYROCHEMICAL PROCESSES
FOR RECOVERY OF FABRICATION SCRAP AND REPROCESSING
OF MONOLITHIC U-MO FUEL**

J. Figueroa, M.A. Williamson, M.A. Van Kleeck, R.J. Blaskovitz,
T.A. Cruse, J.L. Willit, S. Chemerisov, and G.F. Vandegrift

Chemical Sciences and Engineering Division

Argonne National Laboratory, 9700 S. Cass Ave, Argonne, IL 60439 – USA

ABSTRACT

The NNSA Global Threat Reduction Initiative (GTRI) - Conversion program is currently engaged in the development of a nuclear fuel that would enable conversion of US high-performance research reactors (HPRRs) from high-enriched uranium to low-enriched uranium alloy fuel, monolithic uranium-10wt% molybdenum (U-10Mo). Fabrication scrap from the manufacture of the fuel plates for the five U.S. HPRRs will need to be recycled. A number of fuel scrap recovery and treatment options are being considered by GTRI. Of these options, a design concept was developed for a pyrochemical processing system to recover low enriched uranium from the U-10Mo scrap and the benefits of such an approach will be briefly described. Uranium recovery from the fabrication scrap is achieved by electrorefining. The engineering-scale electrorefining system and results from several scrap treatment tests are discussed. The applicability of the pyrochemical process to treating used HPRR fuel is also being evaluated, and results from laboratory-scale electrorefining tests with irradiated U-10Mo fuel will be described.

Introduction

A design concept was developed for a pyrochemical processing system to recover low enriched uranium from the monolithic uranium – 10wt% molybdenum fuel scrap (U-10Mo) produced during the fabrication of fuel plates for the U.S. high performance research reactors (HPRRs).[1-3] The conceptual pyroprocessing system included a process flowsheet providing a theoretical material balance for the system, a brief description of process layout and each unit operation, and the results from feasibility experiments conducted at the laboratory- and engineering-scale that demonstrated uranium recovery from U-10Mo scrap.[1-2] Laboratory-scale electrorefining validation tests successfully demonstrated the recovery of uranium from Zr-bonded U-Mo fuel scrap without significant Zr and Mo impurities being present in the product. In addition, these studies demonstrated that the noble metal fuel components remained within the anode baskets.

The submitted manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory ("Argonne"). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government. Work supported by the U.S. Department of Energy, National Nuclear Security Administration's (NNSA's) Office of Defense Nuclear

Engineering-scale experiments confirmed the laboratory-scale results and were used to begin developing a set of operating guidelines for the system.

Uranium recovery from fuel scrap by pyrochemical means has a number of benefits as compared to the traditional aqueous approach. [1-2] The pyrochemical system directly treats the fuel scrap, recovering the uranium in metal form that can be used in fuel fabrication, no oxide to metal conversion process is required as with the aqueous approach. The pyroprocesses are compact and add minimal footprint to the fuel fabrication facility. Waste produced from the process is far less than that anticipated from an aqueous process and limited to solids including waste salt and crucible materials. More importantly, the pyroprocess does not require the use of hydrofluoric acid to complex the zirconium to avoid explosive reactions or lead to corrosion of the process equipment.

Our research and development activities over the past year focused on extending engineering-scale electrorefining operations experience by treating scrap U-10Mo alloy and simulated U-10Mo fuel, preliminary product processing tests to demonstrate the removal of residual salt from the electrorefining product and uranium consolidation, and evaluating the applicability of pyrochemical processes to the treatment of irradiated U-10Mo fuel. Results from the studies will guide future tests to determine uranium recovery efficiency, define product quality, identify material loss to waste streams, and the effect of process hold-up. The results of the engineering-scale experiments will also be used in future updates to the mass balance flowsheet and increase the accuracy of a process model being developed for a prototype system. The results of our experimental studies in the past year are described in this paper.

2. Engineering-Scale Electrorefiner Experiments

2.1 Experimental Setup

The engineering-scale experiments were performed using Argonne National Laboratory's planar electrode electrorefiner (PEER), which is a prototype module for a potential commercial electrorefiner that could be used to treat U-Mo scrap. The key components of PEER, shown in Figure 1, are a stainless-steel screen anode basket (green), two sets of four stainless-steel cathode rods (white), manual scrapers (blue), and a product basket attached to the bottom of the rectangular frame to collect the uranium product (purple). The manual scrapers consist of blocks with holes (similar to dies), only slightly larger than the cathode rod, that travel the length of the rod to remove product from the cathodes. The anode basket was sized to contain approximately 10 kilograms of U-10Mo scrap. The basket incorporated an extra center rib bisecting the width of the screen to reduce bowing of the porous side panels. The entire apparatus is contained within an inert atmosphere (argon) glovebox maintained below 10 ppm oxygen with low moisture content.

The electrorefiner was operated under galvanostatic conditions throughout the experiments. A voltage limit was established to ensure that uranium was the only transport species present in the molten salt electrolyte; for example, a cut-off voltage of 0.45 V was used to limit the anodic dissolution of zirconium from the feed material. An external power supply was connected to the cell to provide the current required for the refining process. After a specified number of amp-hours passed, the cathodes were scraped to recover the uranium from the electrodes.

Occasionally, PEER was examined before scraping the cathodes to determine the morphology of the deposit on the cathodes. The processing media consisted of a ternary LiCl – KCl – UCl₃ (5-7 wt%) molten salt electrolyte maintained at approximately 500°C.



Figure 1. Sketch of ANL Planar Electrode Electrorefiner (PEER)

Feed material for the process consisted of U-10Mo scrap in the form of plates approximately 1/8" thick and simulated U-10Mo fuel. The simulated fuel was used because of the limited quantity of U-10Mo plate scrap available for the tests. Each simulated fuel was selected for its characteristic qualities such as being clad in zirconium, which simulated the Zr-bonding on the U-10Mo alloy, or consisting of a U-Zr alloy, which simulated the behavior of the U-10Mo alloy.

Chemical analysis of feed material and product is always a crucial aspect of process development but is especially important in designing a system that must meet uranium fuel-fabrication specifications. A major requirement for the uranium used in fuel fabrication is to track the equivalent boron content by testing for B, Cd, Dy, Eu, Gd, Li, and Sm that act as poisons in thermal reactors.[4] In addition to the equivalent boron elements, the standard elements of interest were K, Fe, Zr, Mo, Cd, and, of course, U. Inductively coupled plasma–mass spectrometry (ICP-MS) was used throughout the process to analyze the feed material to establish baseline values, uranium dendrites from electrorefining, molten salt composition, and finally, the uranium ingot after consolidation. Prior to the analysis of uranium dendrites from the electrorefiner, the product was washed with water and the solid metal and salt-loaded aqueous solution were analyzed independently to determine the composition of each phase.

2.2 Results and Discussion

Zr-bonded U-10Mo Scrap. The first engineering-scale electrorefining test was performed using 2.725 kg of Zr-bonded U-10Mo scrap cut into strips approximately 1" x 4". Galvanostatic operation of the system resulted in currents at the beginning of the test of approximately 20 amps followed by the slow linear decay by a total of 12 amps over the first 100 amp-hours. As the experiment continued, the current level declined until testing was stopped as the current dropped

below 5 amps. The experiment was terminated after ~720 amp-hours. An important experimental note for the test run was that the voltage limit for the test was raised from 0.45V to 0.8 V at ~ 370 amp-hours. The test was conducted to determine if even at the higher voltage limit, which was known to allow the transport of Zr in the system, would Mo be retained within the anode basket. During the test, the cathodes were intermittently scraped to harvest the uranium product, a sample of which is shown in Figure 2. The uranium product from each scraping was retained in separate containers and analyzed to determine if the uranium purity remained consistent throughout the test run.



Figure 2. U-10Mo Scrap Uranium Deposit Adhering to Cathode Rods

The ICP-MS results in Table 1 show the purity of the uranium products recovered throughout the test. Four separate uranium products were collected with U1 being the first harvested to U4 at the end of the run. Samples U1 and U2 were collected with a voltage limit of 0.45V and U3 and U4 were collected at the higher voltage limit setting. The uranium products were washed with water to analyze the salt constituents separately from the metal. The uranium metal samples all showed impurity levels for Mo and Fe that were below detection limits. The Zr content in U1 was 0.01 wt% (~70 ppm) while interestingly the Zr content in sample U2 was reported as 1.74 wt% (~17000 ppm), similar to the levels seen in the subsequent products where the voltage limit allowed the anodic dissolution of Zr. Another uranium sample from U2 was submitted for analysis to better understand if the high Zr content was due to a sampling error or the result of Zr dissolution and transport to the cathode. Specifications for the uranium metal used in fuel for the Oak Ridge High Flux Isotope Reactor list that Zr and Mo values must be below 250 and 100 ppm, respectively.[4] Thus, it is essential to develop a sound operating protocol for the refiner.

Table 1. ICP-MS Results for the Zr-bonded U-10Mo Scrap Test (wt%)

Sample ID	Sample wt. (g)	Li	K	Fe	Zr	Mo	Cd	U
U1-Solids	0.7627	0.21	0.95	< 0.18	0.01	< 0.001	0.003	91.54
U2-Solids	0.5191	0.02	< 1.45	< 0.37	1.74	< 0.003	0.02	73.24
U3-Solids	0.1898	0.04	< 3.55	< 0.90	2.20	< 0.007	0.02	101.75
U4-Solids	0.2168	0.03	< 2.62	< 0.67	1.71	< 0.005	0.04	100.23

Samples of the anode residue obtained from the basket at the conclusion of the test revealed that complete uranium recovery had not been achieved. Clearly, the low surface area of the fuel is such that it limited the exposure of uranium to the salt phase, an observation that is consistent with the low currents observed throughout the test. In future tests, the feed material needs to be conditioned (e.g., shredding the fuel plates) to provide a higher surface area of alloy exposed to

the salt phase thereby increasing the current and recovery efficiency. A comparison of the composition of initial feed to the two anode-residual samples confirmed that Mo content in the basket increased on average from 9 wt % to 64 wt%, which is consistent with limited or no dissolution of Mo. Conversely, Zr values declined in the anode residue from 6 wt% to 2 wt% because the voltage limit was set above the Zr dissolution threshold. This result correlates with the increased Zr content found in the uranium products U3 and U4.

Simulated Fuel Tests. The first simulated fuel electrorefining test was conducted using segmented N-reactor fuel. This fuel is composed of depleted uranium with Zircaloy cladding. The constitution of the Zircaloy cladding is primarily Zr with the alloying agents Sn, Fe, Cr, and Ni.[5] In this test, the Zircaloy cladding simulates the Zr-bonding on the U-10Mo scrap. The anode basket was loaded with ~6.5 kg of fuel and allowed to equilibrate overnight prior to experimentation. Currents above 20 amps were achieved throughout the 2000 amp-hour run and were very stable between each of the ~200 amp-hour depositions. The test was stopped as the resistance in the system increased dramatically and theoretical calculations predicted that the uranium had been fully anodically dissolved. ICP-MS results confirmed that the cathode deposit was composed of uranium with very low impurity levels for Fe, Zr, Mo, and Cd, shown in Table 2 below as NR product. The salt wash from the cathode product had a uranium content that was 9.6 wt%, slightly less than twice the content shown in the filtered bulk salt electrolyte sample that was determined to be 5.9 wt% U. This trend is reproduced in every analytical result of uranium product regardless of the feed type. These findings may be attributed to the naturally occurring concentration gradient at the deposition site. A future test to investigate this phenomenon is planned to provide additional insight into the deposition mechanism.

The second simulated fuel electrorefining test was conducted with chopped stainless-steel pins containing U-10wt% Zr alloy. The purpose of using this fuel type was to simulate the separation process with respect to uranium alloys (as opposed to bonding or cladding) as well as to continue to evaluate operating parameters for the refining system. The anode basket was loaded with ~7 kg of fuel and current was passed after the system was allowed to equilibrate. The currents for the test progressed as usual with higher currents seen in the beginning of the test that steadily declined as a function of total amp-hours passed. The current was interrupted every 200 amp-hours in order to perform an in-situ scrape of the cathodes. After ~800 amp-hours, the cathode assembly was raised from the well to manually remove the dendrites from the cathode rods and to unload the product collector. The appearance of the recovered uranium was similar to that recovered in previous tests, shiny needle-like dendrites. Analytical results revealed the product to be high purity uranium metal (shown as UZr product in Table 2) that was coated with only 3.5 wt % residual salt.

Table 2. ICP-MS Results for the Simulated Fuel Tests (wt%)

Sample ID	Sample wt (g)	Li	K	Fe	Zr	Mo	Cd	U
NR Product	0.3123	0.12	1.13	< 0.65	1.E-03	< 4E-03	< 0.01	82.0
UZr Product	0.9495	0.10	2.81	< 0.61	0.05	< 4E-03	< 0.01	87.0

3. Uranium Consolidation Experiments

Uranium consolidation is the process of removing the salt adhering to the uranium dendrites, collected during the electrorefining process, by distillation and melting the uranium into a homogeneous ingot. The uranium metal is recycled to fuel fabrication while the salt that was collected in a condenser is recycled to the electrorefiner. Experiments were designed to demonstrate the feasibility of the process and to determine if the final uranium product meets fuel specifications. Coupled testing of the electrorefining and uranium consolidation processes helps to define the amount of salt retained in the system and provides insight into the amount of waste generated throughout the overall process.

3.1 Experimental Setup

The uranium consolidation apparatus is housed within the same inert atmosphere (argon) glovebox as PEER. Uranium dendrites were transferred directly from the electrorefiner to the consolidation experiment. The dendrites selected from each electrorefining test were harvested from the top of the product collector and therefore might contain a lower amount of residual salt than material harvested via the automated product removal system. Dendrites were loaded into a hafnium-nitride coated niobium crucible. The salt condenser consisted of a long alumina tube that extended beyond the furnace's heating zone with a porous alumina plate that sat above the processing crucible. Loosely packed alumina wool inside the condenser tube captured the distilled salt from the process. The condenser assembly was placed over the crucible and the entire set-up placed in a vertical, clam-shell tube furnace. A temperature controller was used to ramp temperatures to levels sufficient to distill the residual salt and melt the uranium product into an ingot. Melting the uranium into an ingot is desired to guarantee that any salt entrapped within the dendrites is released. After the maximum temperature was held for a specified period of time, the uranium ingot and salt-loaded alumina wool were cooled and sampled for analysis.

3.2 Results and Discussion

U Consolidation of Zr-bonded U-10Mo Electrorefining Product. The HfN coated Nb crucible was loaded with 247 grams of the U1 uranium dendrites and inserted into the furnace with the salt condenser assembly described above. The system was heated at 1°C per minute, held at 300°C for 30 minutes and then taken up to the maximum temperature of 1230°C at a heating rate of 1°C per minute. The maximum temperature was held for 3 hours before the test was terminated and the system was allowed to cool. A cooling rate of 1°C per minute was set but below 500°C the cooling rate was probably much less due to the poor heat conduction properties of argon gas in the glovebox. The system was cooled to room temperature before the clam-shell heater was opened to unload the products.

Inspection of the processing crucible revealed that the uranium dendrites were successfully consolidated into an ingot that was free of salt. The salt was observed as a thin coat of material on all the upper surfaces of the salt condenser that included the alumina tube, dispersion plate, and packing wool. Manual manipulations were required to release the metal ingot from the processing crucible. A photograph of the top and bottom of the recovered uranium ingot is shown in Figure 3.



Figure 3. Uranium Consolidation Ingot

The uranium ingot had a final mass of 208 grams that correlated to a salt mass loss of 15.8 wt%. In order to investigate the uniformity of the sample, the ingot was removed from the glovebox and bisected in air on a SiC cutting wheel. The cross-section of the ingot was found to be visually free of gas pockets and appeared to be consistent across the sample. A thin cut of the material was obtained and segmented into four equal sections for analysis. The alumina wool from the test was completely immersed into an aqueous solution and the solution was analyzed by ICP-MS.

The second test with the U-Mo uranium product utilized the same HfN-coated Nb primary crucible and was loaded with 234 grams of the U2 uranium dendrites. The condenser assembly and the heating profile for the test were identical to the one described for the first consolidation test. After the system had cooled below 150°C, the clam-shell heater was opened to the glovebox and the processing crucible was unloaded from the furnace. The final product was a metal ingot that had adhered to the processing crucible. Manual manipulations were required to release the material from the crucible. In the process of removing the ingot from the crucible, it began to show signs that the HfN coating had chipped as a result of the manual manipulations. The uranium ingot had a final mass of 220 grams that correlated to a salt content of only 6 wt%. The ingot was processed outside the glovebox in air using a SiC cutting wheel and the entire slice of material was sent as one sample for analysis along with the alumina wool.

The ICP-MS data for the Zr-bonded U-Mo consolidation run with the U1 electrorefining dendrites showed very little variation in the purity values among the different locations within the ingot itself and confirmed visual inspections that suggested the sample was homogeneous. The ingot consisted of high-purity uranium metal with lower than detection limit values for Li, K, Fe, Mo, Cd, B, Sm, Eu, Gd, and Dy. The average Zr content of the U1 ingot sample was ~25 ppm, which is a tenth of the allowed 250 ppm level set as the standard for fuel fabrication.[4] The ICP-MS results for the consolidation run with the U2 electrorefining dendrites showed similar results. The U2 ingot had lower than detection impurity values for the boron equivalent elements with the exception of Li, which was reported at 0.14 wt%. The Zr content within the U2 ingot, 0.05 wt%, was found to be much lower than expected given that the feed material was analyzed as having a Zr content of 1.74 wt%. (Recall that an additional sample of U2 dendrites was sent for chemical analysis due to uncertainty in Zr content.) Zirconium metal has a melting point (1855 °C) that is ~700 °C higher than uranium (1132 °C) and would remain within the metal phase if present in the consolidation process. Samples of the alumina wool coated with salt revealed that the uranium content in the salt phase was much less than 1 wt%, which is contradictory to the uranium concentration data obtained for the bulk salt and the salt collected from the dendrites.

4. Pyroprocess Development with Irradiated U-Mo Fuel

The applicability of the pyrochemical process for treating used HPRR fuel was evaluated at the laboratory-scale using electrorefining tests conducted with irradiated U-10Mo fuel. The study utilized the capabilities of the Argonne LINAC to irradiate Zr-bonded U-Mo foils using the γ /fission reaction on ^{238}U . The laboratory-scale electrorefining tests focused on investigating the recovery and decontamination of uranium from the used fuel, and elucidating the behavior of iodine and other fission products in the pyroprocess. In addition, operating parameters for a used-fuel electrorefining system were explored.

4.1 Experimental Setup

The laboratory-scale electrorefining experiments were performed in a furnace well contained within an inert atmosphere (argon) glovebox maintained below 10 ppm oxygen and having a low moisture content. The key components of the electrorefining apparatus, shown in Figure 4, used with irradiated Zr-bonded U-10Mo foil are a stainless-steel screen anode basket, threaded stainless-steel cathode rod, stainless-steel process crucible, and a mullite closed-end tube with a silver wire immersed in a LiCl-KCl-1wt%AgCl salt as the reference electrode. A threaded rod was chosen as the cathode over a smooth rod to promote better retention of the metallic deposit. The loaded anode basket was sized to contain up to 10 grams of U-10Mo feed. A 250 ml stainless-steel beaker was selected as the processing crucible and it contained ~200 grams of LiCl-KCl-5wt% UCl_3 molten salt electrolyte. An external power supply was connected to the cell to provide the current required for the refining process. Ceramic oxides were used on the main process crucible and the electrodes to maintain electrical isolation from the glovebox. 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.

The feed material was prepared by irradiating Zr-bonded U-Mo scrap with a 200 μA beam current and 18 MeV beam energy for six hours at the Argonne LINAC facility using the γ /fission reaction on ^{238}U . After irradiation of Zr-bonded U-Mo foils, samples were sent for analysis to determine the starting composition prior to the pyrochemical processing.

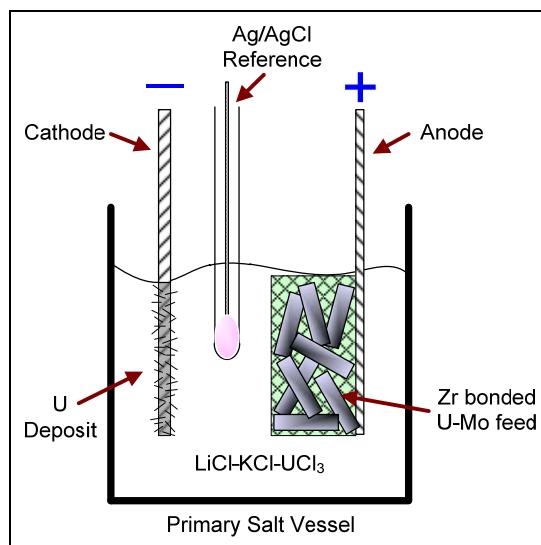


Figure 4. Schematic of the Laboratory-scale Electrorefining Apparatus

4.2 Results and Discussion

Half of the irradiated Zr-bonded U-10Mo foil was used for experimentation and chopped into smaller sections to increase the exposed uranium surface area. The resulting pieces were loaded into the stainless-steel screen anode basket and lowered into the molten-salt electrolyte maintained at 480°C. The tests were conducted using a power supply with potentiostatic control of the cell. The potential was set below -0.45 V cathode versus anode to ensure that the Zr and Mo impurities remained within the anode basket and were not anodically dissolved. The current observed during the beginning of testing was slightly above 200 mA but quickly decayed to an average of 85 mA. As testing progressed and less uranium was available at the anode, the current declined until the test was stopped as the current dropped below 10 mA. The anode basket and the cathode rod were raised from the salt into the heated space above the process crucible to allow the electrolyte to drain. Figure 5 shows the starting anode basket and one of the cathode deposits collected after processing.



Figure 5. Loaded Anode Basket (Left) and Cathode Deposit (Right) from Irradiated Fuel Test

Samples of the irradiated feed, anode residue, cathode deposit, and salt were collected and analyzed by ICP-MS and gamma spectroscopy. Preparation of the anode residue and the cathode deposit for analysis involved washing the metal with water to remove the soluble components. The remaining solid anode residue, metallic dendrites, and their respective water washes were analyzed to identify the composition of the samples.

Analysis of the washed cathode product revealed a high-purity uranium metal deposit with an impurity level for Zr at 25 ppm, which is indicative of a well-controlled cut-off potential. The Mo content in the washed uranium product was reduced from ~9 wt% in the original irradiated feed to 0.03 wt% (305 ppm). The Mo content is higher than expected given that the analytical results for the electrolyte salt show only 1 ppm Mo and 16 ppm Zr content proving that the metals were not oxidized at the anode. An additional uranium product sample was submitted for analysis to verify the results. Analysis of the washed metal residue recovered from the anode basket revealed that the uranium content in the final composition of the feed materials was 36wt% lower than the starting foil but complete recovery of uranium had not been achieved. Again this result is likely due to the limited active surface area of the foil thus limiting the anodic dissolution of the uranium. Feed material preparation such as shredding will eliminate this effect.

Gamma spectroscopy of the initial starting foil and the final products was conducted and compared to the standard peaks for 14 isotopes. The isotopes investigated were Ba-140, Ce-141, Cr-51, I-131, I-132, I-133, La-140, Mo-99, Nb-95, Nd-147, Sb-127, Tc-99M, Te-132, and Zr-95. The isotopes were chosen to predict the path of the active and noble-metal fission products within the pyrochemical process. Active-metal fission products accompanied by any transuranic elements would partition to the molten salt and the noble metal fission products would remain in the fuel basket (anode). The gamma spectroscopy results were interpreted as having the isotope present in the particular component of the cell (anode or salt) when the value was greater than sum of 3-sigma.

The noble-metal fission products were predicted to remain within the anode, and gamma spectroscopy data showed that Cr-51, Nb-95, Tc-99M, and Zr-95 did not partition to the bulk salt electrolyte phase, thus confirming the prediction. The active-metal fission products predicted to be found in the bulk salt electrolyte are Ba-140, Ce-141, I-131, I-132, I-133, La-140, Nd-147, Sb-127, and Te-132. Using the 3-sigma methodology to determine if the isotope exists in the component, the results showed that Ba-140, Ce-141, I-131, I-133, La-140, Mo-99, and Nd-147 were present in the salt. Several of the active metal fission products, however, were also found on the uranium product including Ba-140, I-131, I-132, La-140, Nd-147, and Te-132. Although the uranium product was washed with water to remove the salt soluble species, lithium is still present in the sample. With its only source being the electrolyte salt, it is clear that the washing process to remove the residual salt from the uranium product was incomplete. In fact, it is known that salt is entrapped within the uranium dendrites and is only removed by consolidation of the dendrites into an ingot.

5. Summary

Engineering-scale electrorefining experiments successfully treated scrap U-10Mo alloy and two types of simulated U-10Mo fuel demonstrating the recovery of uranium metal without significant Zr and Mo impurities being present in the product. The studies also verified that the noble-metal fuel components within the scrap would remain within the anode and be contained for eventual waste processing. The tests investigated a variety of operating conditions for the refiner to develop a set of operating guidelines for the treatment of U-10Mo fuel scrap. Test results were extremely promising as the first uranium product (U1) recovered was determined to meet all the purity requirements for fuel fabrication. Subsequent tests with the U-10Mo and simulated fuel scrap examined the effect of changing operating parameters such as voltage limit during electrorefining, which affected the uranium purity. Future electrorefiner tests will be used to determine uranium recovery efficiency, define product quality, identify material loss to waste streams, and the effect of process hold-up.

The uranium consolidation tests established that uranium dendrites could be treated with distillation to create a homogeneous consolidated ingot. Several areas of future research were identified that included improving crucible performance, developing an optimal heating profile, standardizing the sampling techniques, and designing an improved salt condenser. Additional tests are required to develop a system that will be efficient and can meet the through-put requirements for the scrap recovery system.

The applicability of the pyrochemical process for treating used HPRR fuel was evaluated at the laboratory-scale with irradiated U-10Mo fuel. The electrorefining test definitively proved that the process could recover uranium metal without significant Zr and Mo impurities. The laboratory-scale electrorefining test was able to elucidate the behavior of multiple active and noble metal fission products in the pyroprocess. Gamma spectroscopy results were found to be in general agreement with the predicted paths for most fission products, but additional tests are required to develop a robust used-fuel electrorefining model.

6. Acknowledgements

The submitted manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.

Work supported by the U.S. Department of Energy, National Nuclear Security Administration's (NNSA's) Office of Defense Nuclear Nonproliferation, under Contract DE-AC02-06CH11357.

7. References

- [1] J. Figueroa, M. Williamson, M. Van Kleeck, R. Blaskovitz, and G. Vandegrift, “Pyrochemical Scrap Recovery in the Fabrication of LEU Monolithic U-Mo Fuel for High Performance Research Reactors,” RERTR 2010 – 32nd International Meeting on Reduced Enrichment for Research and Test Reactors, Lisbon (Portugal), Oct 10-14, 2010.
- [2] J. Figueroa, M. Williamson, and G. Vandegrift, “Pyrochemical Recovery of Uranium from Monolithic U-10Mo Fuel scrap,” RERTR 2010 – 31st International Meeting on Reduced Enrichment for Research and Test Reactors, Beijing (China), November 1-5, 2009.
- [3] 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.
- [4] R.T. Primm III, R.J. Ellis, J.C. Gehin, D.L. Moses, J.L. Binder, and N. Xoubi, “Assumptions and Criteria for Performing a Feasibility Study of the Conversion of the High Flux Isotope Reactor Core to use Low-enriched Uranium Fuel”, CD Proceedings, PHYSOR 2006, pp 22-23.
- [5] C. L. Whitmarsh, “Review of Zircaloy-2 and Zircaloy-4 Properties Relevant to N. S. Savannah Reactor Design”, Oak Ridge National Laboratory Report No. ORNL-3281, UC-80-Reactor Technology, TID-4500 (17th ed.).