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INITIAL CATHODE PROCESSING EXPERIENCES AND RESULTS

FOR THE TREATMENT OF SPENT FUEL

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ABSTRACT

As part of the spent fuel treatment demonstration at Argonne National Laboratory, a vacuum distillation process is being employed for the recovery of uranium following an electrorefining process. Distillation of a salt electrolyte, primarily consisting of a eutectic mixture of lithium and potassium chlorides, from uranium is achieved by a batch operation termed "cathode processing". Cathode processing is performed in a retort furnace which enables the production of a stable uranium product that can be isotopically diluted and stored. To date, experiments have been performed with two distillation units; one for prototypical testing and the other for actual spent fuel treatment operations. The results and experiences from these initial experiments with both units will be discussed as well as problems encountered and their resolution.

I. INTRODUCTION

One of the key features for the treatment of Experimental Breeder Reactor-II (EBR-II) spent fuel at Argonne National Laboratory is the separation of fission products from the uranium fuel.¹ Metallic uranium fuel, burdened with a multitude of fission products, is initially separated by an electrorefining operation although additional recoveries of the uranium product are realized by a distillation process. In addition to the separation of the fission products, treatment of the spent fuel enables the neutralization of the elemental bond sodium which was essential for heat transfer during reactor operations.

Spent fuel treatment commences with the delivery of the fuel to the Fuel Conditioning Facility (FCF).² Currently, spent fuel is being stored in three locations:

the EBR-II reactor vessel, the Hot Fuel Examination Facility (HFEF), and the Radioactive Scrap and Waste Facility (RSWF). Depending on its origin, the fuel is either dismantled into elements from assemblies or retrieved directly as elements from storage containers. Initial fuel element handling operations are performed in the air cell of FCF.

The fuel elements are then transferred to the argon cell of FCF for element chopping. Elements are chopped into segments by a solenoid-driven punch press and loaded into perforated baskets. Four baskets make up an anode assembly which is then installed into the electrorefiner. Electrorefining is performed by placing the sheared fuel elements into a molten LiCl-KCl electrolyte for dissolution and electrotransporting actinides as a cathode product.³ The cathode product is primarily uranium and eutectic salt loaded with fission product chlorides. Final refining of the actinides in the cathode product is achieved by distilling the process fluids, salt and cadmium, in a vacuum retort. Homogenization and sampling of the uranium product is accomplished by remelting in the casting furnace. Waste products from the process are placed into appropriate waste forms before final disposal.^{2,4}

Development of a distillation process for uranium refining can be traced to Argonne's original spent fuel processing effort.^{5,6} For this process, uranium was

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recovered from a zinc-magnesium alloy by vacuum distillation. Evolution of the current cathode processor design is based on this original research.

Following the initial conceptual design of the cathode processor, it was decided to build two units of equal dimensions, the prototype and the in-cell unit. The prototype cathode processor is located in a glove box enclosure at Argonne-East in Illinois while the other unit has been placed in a shielded hot cell of FCF at Argonne-West in Idaho. Operation of the prototype cathode processor commenced a full year in advance of the in-cell unit to allow for potential design modifications. During the first year of test operations, several changes were made to the prototype cathode processor and, subsequently, to the in-cell design. To date, operation of both cathode processors has comprised of simple distillation experiments with salt and/or cadmium only as well as experiments with actual dendritic cathodes containing depleted uranium (DU). Obviously, the presence of the prototype cathode processor has alleviated additional testing of the in-cell unit while also providing invaluable dual testing capabilities.

II. EQUIPMENT

A. Layout and Features

The major components of the cathode processor are shown in Figure 1. The major features of the cathode processor are the vessel (shell plus flanges), the induction-heated furnace region inside the vessel at the top, and the condenser/collector region inside the vessel at the bottom. The induction-heated furnace region uses a passively cooled induction coil and a graphite furnace liner that acts as the susceptor. The condenser/collector region includes a condenser assembly and a receiver crucible, which is used to collect the condensed cadmium and salts.

The cathode processor occupies two stations in the FCF argon cell. The vessel assembly is supported 191 cm above the floor of the argon cell. The elevated support allows bottom-loading of the cathode processor. The bottom loaded configuration requires a dedicated loading mechanism which consists of a lifting mechanism and a trolley-rail system. The lifting mechanism raises and lowers the crucible assembly (the process crucible, the radiation shield assembly, and the condenser/receiver crucible assembly) between the shell and rail. The rail system moves the crucible assembly from the lift position to the loading/unloading position.

B. Enhancements

During initial testing of the prototype cathode processor, two major problems were encountered. The first problem was that a design operating temperature of 1370°C could not be achieved in the furnace region. The second problem was twofold: 1) cadmium was migrating out of the inner confinement and condensing in undesirable locations; and 2) during the processing of large amounts of cadmium, the cadmium was condensing and collecting in the condenser and not in the receiver crucible.

Thermal losses from the furnace region to the condenser and furnace liner regions prevented the prototype cathode processor from reaching the design operating temperature of 1370°C. A major contributor was the thermal radiation loss down the annulus around the radiation shield assembly into the condenser region. Larger thermal radiation shields were added thus greatly reducing the loss from the furnace region. Furnace components were modified to provide an improved inductive match between the induction coil and the susceptor. To further reduce thermal losses to the furnace liner region, the outer diameter of the furnace liner assembly was reduced and additional insulation installed between the furnace liner and coil assemblies. Additional testing of the furnace performance was conducted with these modifications by bringing the furnace up to various prescribed temperatures, holding at that temperature, and then raising the furnace up to the next prescribed temperature until the design temperature of 1370°C was reached. These runs did not contain any process materials. Tests of this type verified that the furnace design temperature could be obtained and that the balance of the cathode processor was staying sufficiently cool.

The condenser assembly was redesigned to address the second problem. A new seal design was employed between the upper surface of the condenser and the lower surface of the furnace liner assembly base. The condenser walls were thermally uncoupled from the support can/baffle assembly and the horizontal shelves in the condenser were removed. Because of these changes, the migration of cadmium out of the inner confinement was eliminated and the intended collection of distillate in the receiver crucible was achieved.

In addition to the modifications to improve the vacuum boundary leak tightness, modifications were made to eliminate component failure during operations. Components which had an early history of failure were;

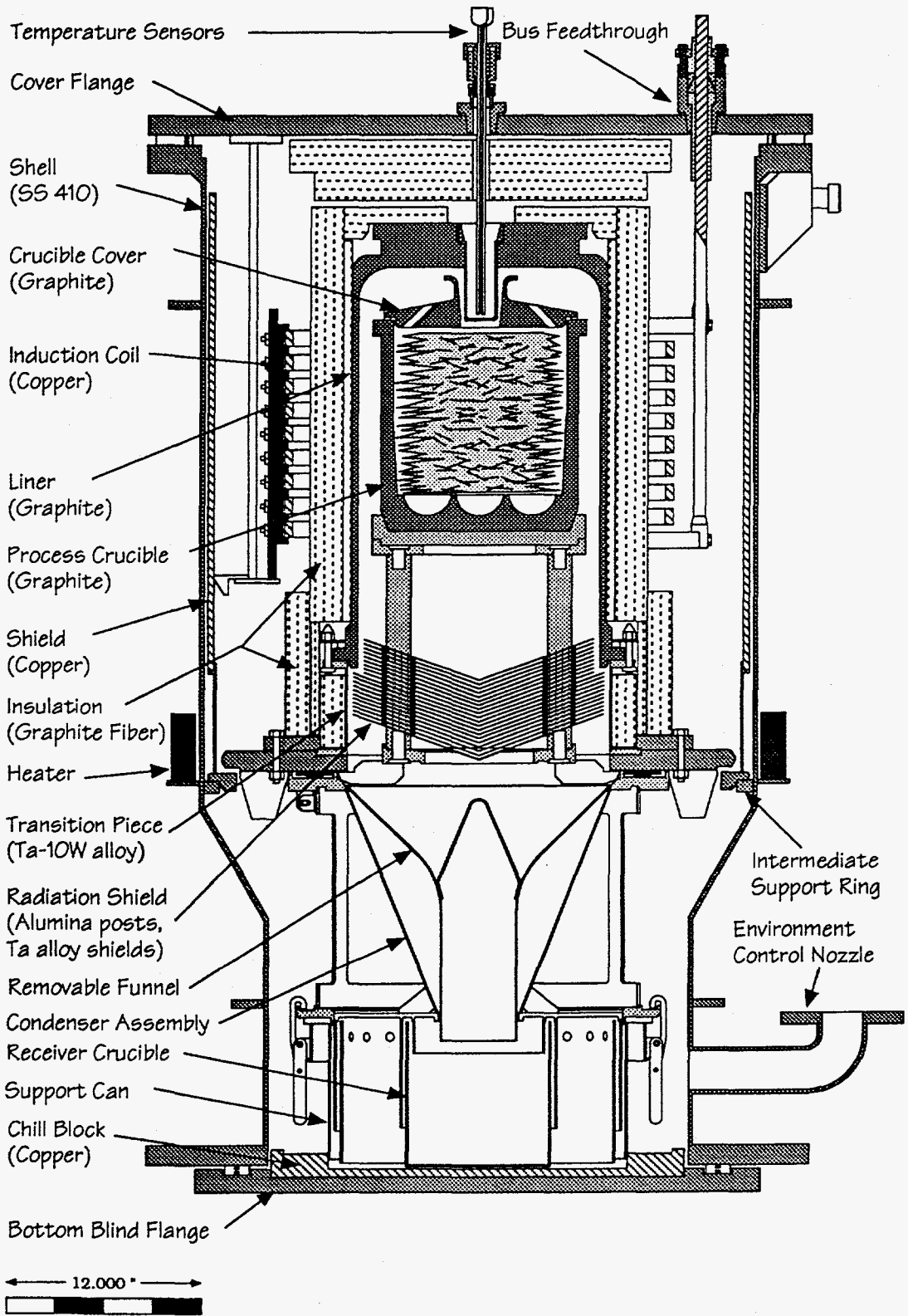


Figure 1. Schematic of Cathode Processor Equipment

the induction coil power feed through seals, the furnace liner instrument well, the thermal radiation shield structural members, and the vacuum pump. All of these components have been modified successfully to eliminate the earlier frequent failures.

C. Process Sequence

The overall process sequence involves: 1) process crucible coating; 2) process crucible loading; 3) cathode processor loading; 4) processing; 5) cathode processor unloading; and 6) receiver and process crucible unloading. The processing sequence described is for the in-cell cathode processor but similar operations are performed for the prototype. The following paragraphs discuss this sequence.

The process crucible is transferred out of the argon cell to a coating station in the basement of the FCF. The graphite crucible is cleaned, weighed, and coated. The coated crucible is weighed and either baked-out in a furnace out-of-cell or transferred back into the argon cell and baked-out in the cathode processor vessel.

The process crucible is loaded at a worktable near the electrorefiner. The crucible assembly is lowered from the vessel and translated on the rail system to the loading position. The radiation shield and condenser assemblies are removed, and the receiver crucible is installed in the support can/baffle assembly. The condenser and radiation shield assemblies are then re-installed. The process crucible is placed on top of the radiation shield assembly, and a splatter shield installed on the crucible to prevent splattering of distillate during processing. The entire assembly is then translated under and raised into the vessel. The charge is then processed, with the cadmium and salt being vaporized and collected in the receiver crucible while the uranium remains in the process crucible.

Upon completion of the processing operation, which includes an adequate cooldown period, the crucible assembly is lowered out of the vessel and translated on the rail system to the unloading position. The splatter shield is removed, and the process crucible is removed and weighed. The radiation shield and condenser assemblies are removed to allow access to the receiver crucible. The receiver crucible is removed and weighed. The condenser and radiation shield assemblies are then reinstalled onto the support can/baffle assembly. The assembly is translated under and raised into the vessel. A mass balance is performed for material accountability purposes.

The receiver crucible, containing the salt and cadmium distillate, is then unloaded and returned to the electrorefiner. The process crucible is unloaded by placing the crucible into a dumping apparatus and inverting the process crucible to release the uranium ingot(s).

III. EXPERIMENTATION

A. Prototype Cathode Processor

Most of the experimental testing has been completed in the prototype cathode processor at Argonne-East. The test program began with assembly and control system check-out tests, continued with hot operational tests without process materials, and has proceeded to hot operational testing with process materials. As previously described, the hot operational testing of the prototype cathode processor is accomplished in an argon atmosphere provided by a large test enclosure.

The hot operational testing runs with process materials began with cadmium, then cadmium and salt, DU, and finally cadmium, salt, and DU.

Early distillation tests using cadmium and salt indicated that the original condenser drainage features were inadequate as described earlier. Typical tests distilled 500 grams of cadmium with 1500 grams of salt and 33,000 grams of cadmium with 500 grams of salt. The tests with the original condenser design resulted with as much as 67 wt. % of the distillate being held up in the condenser, depending on the operating conditions and history. The present condenser design with improved drainage features consistently results in less than 1 wt. % hold-up.

Baseline tests have been completed which melt DU without cadmium or salt present. These tests used graphite crucibles with various wash coats and included a melt of about 11,000 grams of DU in a full size graphite crucible coated with a yttria (Y_2O_3) wash. The test with yttria coating ran to a maximum temperature of 1370°C with a hold time of 30 minutes and resulted in well consolidated ingots with a very light scale on the surface, no loose dross, and excellent mold release. More recent baseline melt tests were operated at a maximum temperature of 1200°C with a hold time of 30 minutes to melt 0.6 cm thick by 5.1 cm diameter DU disks. These more recent tests utilized small graphite test crucibles which were coated with wash coats of yttria, alumina (Al_2O_3), or zirconia (ZrO_2). In each case

the DU melted to conform to the shape of the crucible mold. No loose dross was formed but the ingots were covered with a very thin, tenacious scale.

When the testing progressed to the processing of salt and cadmium with the DU, a major change in the quality of the DU melt was observed. The addition of salt to the process resulted in the salt reacting with the yttria coating and the formation of loose dross, a heavier scale on the ingot surface, and very poor mold release. The yttria wash coat essentially disappears in the presence of the salts. This led to a search for an alternative coating for graphite crucibles or an alternative crucible material.

Prior to processing with DU, simulated solid cathode runs were performed which contained approximately 500 grams of cadmium and 1500 grams of salt in the process crucible. A maximum process crucible temperature set point of 1300°C was programmed with a hold time at 1300°C of about 60 minutes. All the cadmium and salt were distilled out of the process crucible and collected in the condenser/receiver assembly. Tests of this type verified that cadmium and salt were efficiently transported downward into the receiver crucible as expected. Inspection of the process crucible after these runs show that all of the cadmium and salt had been vaporized out of the crucible.

A subsequent simulated solid cathode test included approximately 10,000 grams of DU in addition to the 500 grams of cadmium and 1500 grams of salt. A full size graphite crucible coated with a yttria wash coat was used. Loose dross was formed and the ingots had to be mechanically removed from the crucible molds, ruining the mold pocket surfaces in the process.

Following the simulated solid cathode experiments, a solid cathode product from the Chemical Technology Division (CMT) electrorefiner located at the Argonne-East site was processed and contained 9228 grams of dendrite material. The dendrites consisted of approximately 90 wt. % DU, 10 wt. % salt, and negligible amounts of cadmium. This test run brought the process crucible up to 1300°C for 30 minutes, distilling off the salt and consolidating the uranium into seven ingots that formed in the seven hemispherical pockets at the bottom of the process crucible. All the salt was vaporized and collected in the condenser/receiver assembly during the test. The uranium was fully consolidated into seven ingots of about equal mass. In this case, the graphite crucible was coated with the zirconia coating and the ingots were easily removed by inverting the process crucible. Subsequent testing has

continued to demonstrate that excellent mold release is obtained using a zirconia wash coat of graphite crucibles. However, as compared to processing DU without salt present, the ingots are typically covered with a heavier scale on the surface and loose dross is formed. Careful attention to the application of the coating and a high temperature bake-out of the crucible before processing reduces the amount of dross formed but does not eliminate it.

The test results to date have led to the search for a better wash coat for graphite crucibles or an alternative crucible material. In addition to yttria and zirconia coatings, an alumina wash coat has been tested. The experience with alumina to date has been that we have been unable to get the coating to adhere to the crucible without cracking and/or flaking off. All of the wash coatings tested so far have been supplied by Zyp Coatings, Inc., Oak Ridge, TN.

Graphite crucibles, which have been plasma-spray coated with tungsten and zirconia using a multi-layer proprietary procedures of Applied Coatings, Inc., Columbus, OH, have also been tested. The plasma-spray coatings were coated with the zirconia wash coats prior to use. The plasma coating was very easy to wash coat and resulted in what appeared to be a superior coating from visual examination. The initial test results were promising; however, after about six test runs the plasma coatings had appeared to have disappeared from the mold region of the crucibles. This may have been a result of vigorous mechanical cleaning and recoating of the zirconia wash after each run. The cleaning was performed in an effort to examine the condition of the plasma coating after each run.

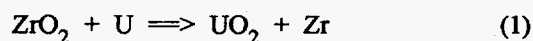
Alumina crucibles and beryllia (BeO) crucibles have also been tested with solid cathode materials. The test with the alumina crucible was not acceptable but the alumina was of unknown purity. High purity alumina crucibles will be tested in the future. The experience with small beryllia crucibles has been that the initial results from a new crucible are excellent. However, after five or six runs poor mold release is experienced and the scale on the ingot appears to be getting heavier. When poor mold release is experienced, the result is that the ingot sticks to the beryllia and, when removed, mechanically pulls small amounts of the crucible material from the mold region. One test was made with a large beryllia crucible which mechanically failed apparently from thermal stresses. The beryllia crucibles were manufactured by Brush Wellman, Inc., Tucson, AZ. At this point beryllia crucibles do not appear

promising for use in this process.

B. In-Cell Cathode Processor

As mentioned, testing with the in-cell unit started a full year later than with the prototype. Thus, the new condenser and other modifications were in place for the initial experiments with the in-cell unit. The first test with the in-cell cathode processor utilized a simulated solid cathode product and completed the qualification of the equipment. The simulated experiment consisted of approximately 6000 grams of DU, 800 grams of eutectic LiCl-KCl salt, and 40 grams of cadmium loaded separately but in the same crucible, see Table 1. Other operating variables as well as conditions for Test 1 are also given in Table 1. Sampling at the casting furnace was not performed for this experiment.

Probably the most important result from Test 1 was the formation of a significant amount of a black dross layer on top of the ingots. Although observed during prototype testing, the quantification of this material was not performed. The dross formation is caused by the reaction of the uranium melt with the zirconia crucible coating according to equation (1). The zirconia coating is used on the graphite process crucible to minimize the reaction of the uranium with the graphite and for ease of ingot removal from the crucible. Loss of uranium to the dross would affect the overall efficiency of the spent fuel treatment process and thus, should be minimized.



As shown in Table 1, the dross and coating material have been combined following a run and weighed since the chemical composition of the two streams shows little variance. If the amount of zirconia coating applied to the crucible is subtracted from the dross and coating stream weight, an estimated amount of uranium can be assigned as lost to this stream. Estimation of uranium loss in this way is not entirely rigorous considering the stoichiometric implications of the equation and possible external sources of oxygen but represents a simple method for calculation of the uranium loss. A value of 6.6 wt. % is estimated for the uranium loss to the dross and coating stream for Test 1 in terms of the final ingot weight.

Typical chemical analyses of the dross and coating stream show a composition of between 60-90 wt. % U, 5-20 wt. % Zr, and the balance being primarily oxygen although other minor impurities (carbon and nitrogen)

have been observed. X-ray diffraction results have confirmed the following species in the dross and coating samples: ZrO_2 , $\text{ZrO}_{0.35}$, UO_2 , UO, UC, and U. The occurrence of substoichiometric species of both uranium and zirconium oxides (UO and $\text{ZrO}_{0.35}$, respectively) is rare but not undocumented, particularly under vacuum conditions and high temperatures.^{7, 8, 9}

The process crucible for Test 1, and Tests 2 and 3, had seven hemispherical molds machined symmetrically into the bottom of the crucible to contain the consolidated uranium product. The coating was applied by brushing the zirconia slurry onto a heated crucible. Application of the coating for subsequent tests was by a spray gun for uniformity. In addition, a bake-out, at approximately 300°C, of the crucible and coating was performed as suggested by the coating vendor for succeeding tests. A slight adherence of the ingots to the crucible during the dumping operations for Test 1 was observed and probably caused by the absence of the bake-out step.

For Test 2, approximately 3800 grams of dendritic material from the FCF electrorefiner was processed in the cathode processor to gain experience with actual cathode products. Again, the seven-ingot crucible was used with a sprayed zirconia coating and bake-out step. The zirconium content was found as a result of sampling at the casting furnace and is due to zirconium carry-over from the cathode processor crucible coating. Operating conditions mimicked those of the first test except for an extended hold time. As can be seen in Table 1, the uranium loss more than doubled (6.6 wt. % to 13.8 wt. %) from the first test and is most likely caused by the smaller batch size rather than the increased hold time.

Based on the results from Tests 1 and 2, it appears that the formation of dross poses a significant problem to the process. The alternative material testing to date at the prototype has not found a better performer thus, subsequent tests with the in-cell cathode processor will attempt to ameliorate this problem with regard to zirconia coating.

Test 3 was performed at a reduced maximum operating temperature of 1225°C in an attempt to minimize the effects of equation (1). Dendrites for this run were provided by the CMT electrorefiner since the FCF dendrites were not available. The higher zirconium content of the dendrites can be attributed to the electrorefining of typical metallic fuel with a composition of ninety percent uranium and ten percent zirconium (U-10Zr) by weight. Because of the

zirconium and its effect on the melting point of the melt, the ingots bridged together and appeared partially consolidated. In addition, an imbalance of material at the casting furnace led to the conclusion that not all of the distillate had volatilized at the cathode processor. Hence, other parameter adjustments must be made for future tests at 1225°C to ensure complete distillation. An appreciable reduction in uranium loss was achieved for this test by decreasing the maximum temperature.

Dendritic material from CMT was once more processed in the in-cell cathode processor for Test 4. Included in the salt of this material were minor amounts of several rare earth chlorides as noted on Table 1. The major change for this experiment was the use of a new crucible configuration which both decreases the available coating/melt reaction surface area, by almost a factor of two, and eliminates bridging caused by the flat surfaces of the original design. As evident, the loss of uranium to the dross was further reduced from 2.3 wt. % to 1.7 wt. % for Test 4. Again, the ingot appeared partially consolidated due to the high zirconium content of the dendrites.

The dendrites for Test 5 were from the FCF electrorefiner and therefore contained no zirconium as indicated in Table 1. Operation of the cathode processor

was performed at 1260°C. Uranium loss for this run was comparable to the previous test showing that zirconium content and a reduction of 40°C in the maximum temperature have essentially no effect on the loss.

As referred to earlier, operation of the cathode processor at temperatures of 1225°C or lower requires the assurance that all of the distillables be vaporized. For Test 6, the initial operating pressure was reduced from 10 Torr to 1 Torr to accommodate the operating temperature of 1225°C. This pressure reduction decreases the estimated boiling point of the salt by 150°C to approximately 800°C. Even though the vaporization of the distilland is initiated at 800°C and 1 Torr, complete distillation occurs differentially with increasing temperatures. The dendrites for Test 6 were from the FCF electrorefiner and contained no zirconium as shown in Table 1. A slight reduction in the uranium loss values is realized for this run based on the combined effects of temperature, pressure, and crucible design.

IV. DEVELOPMENTAL ACTIVITIES

Following the DU operations mode for spent fuel treatment, processing of irradiated fuel will begin at the FCF electrorefiner and subsequently at the in-cell cathode processor. Thus, the elimination or at least the

Table 1. Summary of In-Cell Cathode Processor Experiments

	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
Material (g)	5996 DU, 795 LiCl-KCl, 41 Cd	3820 FCF Dendrites	7014 CMT Dendrites	7018 CMT Dendrites	5989 FCF Dendrites	6200 FCF Dendrites
Distillate (wt.%) [1]	12.5	20.7	11.7 [2]	16.9 [3]	17.0	16.0
Zr (wt.%)	Unknown	0.1	9.1	9.8	ND [4]	ND [4]
Process Crucible	Seven-Ingot	Seven-Ingot	Seven-Ingot	Flat-Bottom	Flat-Bottom	Flat-Bottom
Coat Application	Brush, No Bake	Spray, Bake	Spray, Bake	Spray, Bake	Spray, Bake	Spray, Bake
Maximum Temp. (C)	1300	1300	1225	1300	1260	1225
Hold Time (min.)	30	60	30	30	30	30
Initial Press. (Torr)	10	10	10	10	10	1
Dross & Coat (g)	471	434	245	181	162	143
Zirconia Coating (g)	100 [5]	74	106	83	78	80
Ingot (g)	5629	2602	6028	5646	4800	5069
U in Dross & Coat (g)	371	360	139	98	84	63
U Loss/Ingot (wt.%)	6.6	13.8	2.3	1.7	1.8	1.2

[1] Includes LiCl-KCl, UCl₃, and Cd

[2] Includes 58 grams lost at casting furnace

[3] Includes NdCl₃, CeCl₃, and YCl₃

[4] Not Detected

[5] Estimated based on subsequent coatings

minimization of the coating/melt interaction must be accomplished while still in the DU mode. A value of less than 1 wt. % for the uranium loss seems like a reasonable goal given the experimentation with both cathode processors to date.

Additional testing with the zirconia crucible coating includes high temperature (> 600°C) bake-out to increase the coating hardness and optimization of the current operating conditions. The objective of both paths is to reduce the uranium losses with respect to the ingot size to less than 1 wt. %.

As stated, the development of a better coating or crucible material is currently being pursued. Potential coatings to be tested in the prototype cathode processor are calcia (CaO), zirconium diboride (ZrB₂), and magnesia (MgO). Crucible materials under consideration include the aforementioned alumina, tribocor (nitrided Nb-Ti-W alloy), and possibly magnesia or erbia (Er₂O₃). The most promising approach, to date, is the use of a sacrificial DU crucible liner inside of a yttria-coated graphite crucible. The liner would contain the dendritic material during salt distillation and be incorporated into the melt after the salt has been distilled.

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REFERENCES

1. J.E. Battles, J.J. Laidler, C.C. McPheeters, and W.E. Miller, "Pyrometallurgical Processes for Recovery of Actinide Elements," *Actinide Processing: Methods and Materials*, ed. B. Mishra and W.A. Averill, 135-151, The Minerals, Metals, & Materials Society, Warrendale, PA, 1994.
2. K.M. Goff, R.W. Benedict, and D. Levinskas, "Spent Fuel Treatment at ANL-West," *Proceedings of DOE Spent Nuclear Fuel - Challenges and Initiatives*, 282-287, American Nuclear Society, La Grange Park, IL, 1994.
3. K.M. Goff et al., "Fuel Conditioning Facility Electrorefiner Start-Up Results," this proceedings.
4. B.R. Westphal et al., "Production of Metal Waste Forms from Spent Fuel Treatment," *Proceedings of DOE Spent Nuclear Fuel - Challenges and Initiatives*, 288-294, American Nuclear Society, La Grange Park, IL, 1994.
5. J.F. Lenc et al., "Retorting Unit for Recovery of Uranium from Zinc-Magnesium Solutions," Report ANL-7503, Argonne National Laboratory, 1969.
6. C.E. Stevenson, *The EBR-II Fuel Cycle Story*, 85-119, American Nuclear Society, La Grange Park, IL, 1987.
7. J.J. Katz, G.T. Seaborg, and L.R. Morss, *The Chemistry of the Actinide Elements*, 256-260, Chapman and Hill Ltd., New York, NY, 1986.
8. J.J. Katz and E. Rabinowitch, *The Chemistry of Uranium*, 244-302, McGraw-Hill Book Co. Inc., New York, NY, 1951.
9. Holmberg and Dagerhamn, *Acta Chem. Scand.*, 15, 919, 1961.

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