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FUEL CONDITIONING FACILITY ELECTROREFINER START-UP RESULTS

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ABSTRACT

At Argonne National Laboratory-West* (ANL-West) there are several thousand kilograms of metallic spent nuclear fuel containing bond sodium. This fuel will be treated in the Fuel Conditioning Facility (FCF) at ANL-West to produce stable waste forms for storage and disposal. The treatment operations will make use of an electrometallurgical process employing molten salts and liquid metals. The treatment equipment is presently undergoing testing with depleted uranium. Operations with irradiated fuel will commence when the environmental evaluation for FCF is complete.

I. INTRODUCTION

The Experimental Breeder Reactor II (EBR-II) at ANL-West in Idaho went critical in 1963. It is a sodium-cooled fast reactor with a maximum power level of 62.5 MWt while generating 20 MWe. Between 1965 and 1969, EBR-II spent fuel was processed in FCF using a melt refining operation.[1] The recovered actinides were recycled as new fuel. After 1969, FCF was modified to emphasize fuel examination services, and EBR-II fuel was treated at the Idaho Chemical Processing Plant.

In recent years, FCF underwent modifications to demonstrate the fuel cycle for the Integral Fast Reactor (IFR), an innovative liquid-metal-cooled reactor concept. The IFR was being developed to take advantage of the properties of metallic fuel and liquid-metal cooling to offer significant improvements in reactor safety, operation, fuel cycle economics, environmental protection, and safeguards.[2] For the IFR fuel cycle demonstration, EBR-II spent fuel was to be refined, and the recovered actinides were to be fabricated into new fuel. During 1994, the decision was made by the Department of Energy (DOE) to terminate this program, and EBR-II was shutdown on September 30, 1994.

The EBR-II spent fuel, which is metallic fuel containing bond sodium, is unique with respect to the other fuels within the DOE complex. The sodium is fused within the fuel slug structure which swells extensively during irradiation. The presence of the sodium adds a reactive and therefore a hazardous component. Additionally, EBR-II core fuel uses highly enriched uranium. For the safe storage and eventual disposal of EBR-II spent fuel, treatment operations are required to neutralize the reactive sodium, stabilize the fission products, and recover the actinides in a stable form. These operations will be performed in the Fuel Conditioning Facility and will make use of existing equipment from the terminated IFR demonstration.

II. SPENT FUEL INVENTORY

EBR-II fuel is primarily categorized as either driver or blanket fuel. Both types are metallic fuel containing bond sodium, so both require treatment. This fuel is currently stored at ANL-West in four locations: the EBR-II reactor vessel, the Hot Fuel Examination Facility (HFEF), the Radioactive Scrap Waste Facility (RSWF), and most recently, the air cell in FCF. The fuel in the reactor vessel is presently being unloaded. Although not part of planned treatment operations, additional EBR-II metallic fuel is stored at the Idaho Chemical Processing Plant and the Savannah River Site.

The driver fuel is composed primarily of four different fuel assembly types: Mark-IIC, Mark-IICS, Mark-III, and Mark-IIIA. The Mark II materials are control and safety assemblies. Most of the driver materials are Mark III type core assemblies. The bulk of this fuel is a uranium-(10 weight percent) zirconium

alloy. All four fuel assembly types are composed of 61 fuel elements containing highly enriched uranium. The end-of-life enrichment of the Mark-II types is approximately 75 percent, and the Mark-III types are approximately 63 percent enriched. The cladding and fuel assembly hardware are made of 316, D-9, or HT-9 stainless steel. Some characteristics are given in Table 1.

Table 1 - Driver Fuel Characteristics

Fuel Type	Mark IIC	Mark IICS	Mark III & IIIA
Initial Heavy Metal (kg)	2.5	2.5	4.5
Cladding Diameter (cm)	0.44	0.44	0.58
Cladding Length (cm)	62	52	73
Fuel Slug Initial Length (cm)	34	34	34

The approximate inventory of driver fuel at ANL-West is more than 1000 kg of heavy metal. The average fuel assembly burn-up is approximately 8 atom percent, but some elements have burn-ups as high as 20 atom percent.

In addition to the standard driver fuel assemblies, there is a large quantity of experimental elements and fuel assemblies. Approximately 400 of these elements contain a uranium-plutonium-zirconium alloy.

The largest fraction of heavy metal in spent fuel at ANL-West is contained in the blanket fuel assemblies. There are almost 23,000 kg of heavy metal in the blanket fuel at ANL-West. The bulk of this mass is depleted uranium, but there are also more than 100 kg of plutonium.

III. TREATMENT OPERATIONS

FCF consists of two operating hot cells. Figure 1 depicts FCF and the positioning of the equipment in the hot cells. The fuel fabrication equipment pictured been removed from the cell as part of the EBR-II shut down. Spent fuel will first be transferred into a rectangular-shaped, air-filled hot cell where the fuel elements will be separated from the fuel assembly hardware using the vertical assemble dismantler (VAD). Intact fuel elements will be transferred into the adjacent, annular-shaped, argon-filled hot cell.

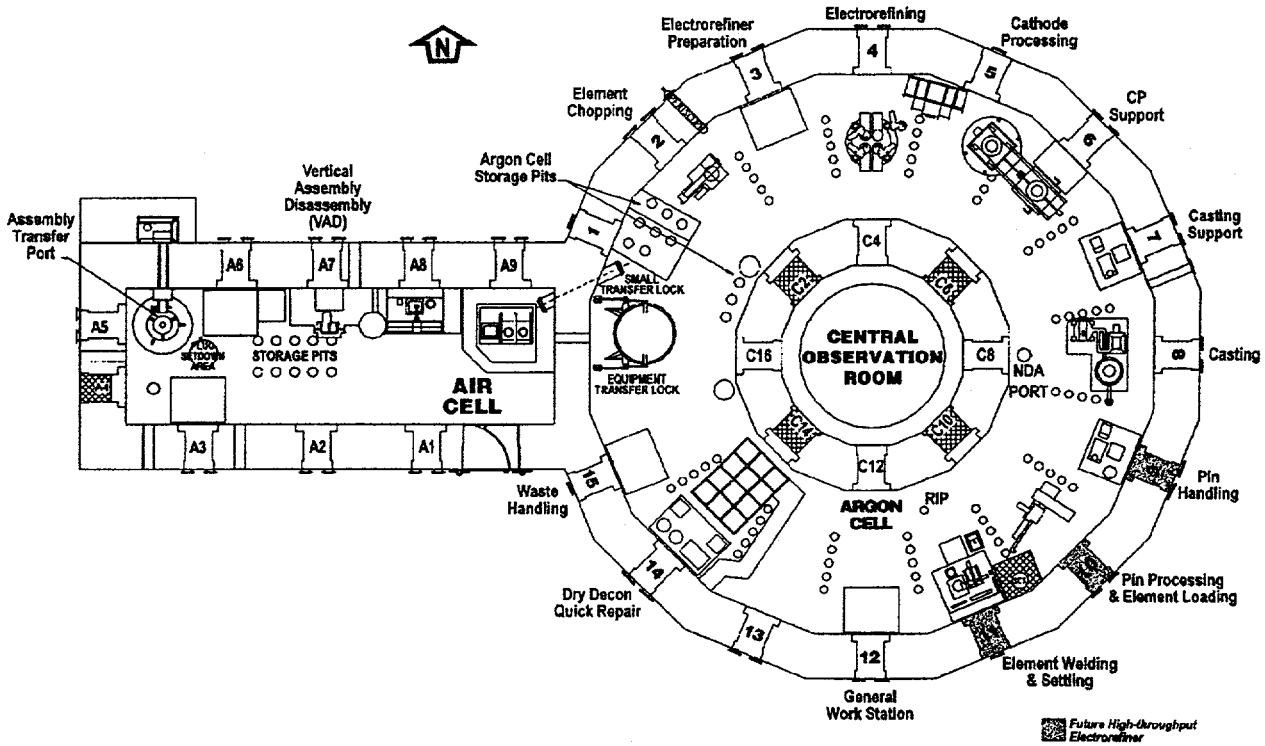

Figure 1. Fuel Conditioning Facility Hot Cells

In the argon cell, the fuel elements are first chopped into segments ranging from 0.64 cm (0.25 inch) to 1.9 cm (0.75 inch) with the element chopper. The chopped segments are then transferred to the electrorefiner in steel baskets (anode baskets).

The fuel treatment operations in the electrorefiner are based on a process employing molten salts and liquid metals in an electrochemical operation. The molten salt medium in the electrorefiner is a mixture of LiCl-KCl eutectic and actinide chlorides. Below the salt phase is a pool of molten cadmium that can serve as an anode, cathode, or just a collector for less reactive metals.

In the electrorefiner, the spent fuel can be electrotransported out of the anode baskets and an equivalent amount of material deposited either in the cadmium pool (anodic dissolution) or directly to a solid cathode (direct transport). Material deposited in the cadmium pool can later be electrotransported to a solid cathode. The uranium will be separated from the bulk of the fission products and transuranics using a steel mandrel cathode (solid cathode). Most of the fission products and transuranics will become concentrated in the salt and cadmium phases in the electrorefiner. The transuranics and alkali, alkaline earth, rare earth, and halide fission products will be primarily in the salt phase. The sodium will be neutralized by forming non-hazardous NaCl. The more noble metal fission products and fuel alloy zirconium will be in the cadmium phase primarily as insolubles or retained in the chopped cladding segments in the anode baskets.

The electrorefining operations have been demonstrated on laboratory and engineering scales in the Chemical Technology Division at Argonne in Illinois.[3] Those initial demonstrations used simulated fission products and small amounts of plutonium.

The cathode products from electrorefining operations will be further processed in a separate operation to distill any adhering salt or cadmium and recover the uranium which will be sampled for material control and accountability. These operations will be performed in the cathode processor and casting furnace. The solid cathode products will consist of uranium that is relatively free of transuranics. As part of the driver fuel processing, the solid cathode will contain highly enriched uranium. Therefore, the recovered uranium metal will be blended with depleted uranium to produce a product that is less than 20 percent enriched. The low enriched uranium products will be placed in interim storage on the ANL-West site as ingots in canisters pending a DOE decision on ultimate disposition.

As part of the treatment operations, the cadmium and salt will periodically be pumped through a metal sintered filter to remove any insolubles and most of the noble metal fission products. These materials will be combined with the stainless steel from the fuel elements and zirconium from the fuel matrix to produce a metallic waste form containing approximately 3 weight percent fission products, 15 weight percent zirconium, and stainless steel. The cadmium will be recycled throughout the treatment operations.

After filtration, the salt will be contacted with zeolite to remove the more active metal fission products and transuranics including plutonium. The resulting zeolite will be processed with additional zeolite and glass additives into a ceramic waste form which will be qualified for disposal as high level waste. Portions of the salt will be recycled.

IV. Equipment Status and Operation

The fuel treatment systems have been developed and installed in parallel with refurbishment of the facility. Operations in both cells are being performed using remote methods. The major processing components are now in the hot cells.

A. Vertical Assemble Dismantler

In the air cell, a vertical assemble dismantler (VAD) removes the fuel elements from the fuel assemblies. In a Mark III fuel assembly, the fuel assembly hardware is more than 66 percent of the assembly mass. This material will be treated as greater than class C waste.

The VAD is the only processing system retained from earlier fuel cycle work. Its design is based on a nearly identical piece of operating equipment in the Hot Fuel Examination Facility (HFEF) at ANL-West. This system has been cleaned and reassembled with new electrical drive and control components. The operation of this piece of equipment has been verified by dismantling mock fuel assemblies.

B. Element Chopper

In the argon cell, the element chopper is used to segment fuel elements in preparation for dissolution in the electrorefiner. It processes elements one at a time by removing the spacer wire from the fueled section and chopping the fueled section into segments of certain lengths. These segments fall into fuel-dissolution baskets (anode baskets) mounted in movable receptacles positioned on a turntable below the shearing blades. The non-fueled portion of the elements, or plenum section, is dropped into a separate container. Element segmentation is done with a punch press fitted with shearing tools. The L12000 press used is a Lourdes

Figure 2. Element Chopper in the FCF Argon Cell

Systems, Inc. solenoid-driven die set. The element chopper installed in the argon cell is pictured in Figure 2.

The principal purpose of the element processing/element chopper qualification done to date has been to prove that element segmentation can be performed remotely in an argon cell environment and to provide feedstock for electrorefiner qualification and testing. As part of qualifying the chopper for operation, stainless steel slugs were first processed. A total of 82 stainless steel slugs with the same dimension as Mark III driver fuel elements were chopped into 1.9 cm (0.75 inch) segments.

The material from the equivalent of four simulated Mark III fuel assemblies was chopped next. The fuel slug material was depleted uranium with 10 weight percent zirconium. It was clad in stainless steel. Bond sodium was also present. This material will be used as part of cold testing for the electrorefiner. A total of 237 simulated fuel pins were processed. This material fills eight fuel dissolution baskets, or two anode assemblies. One anode (four baskets) was prepared with 0.64 cm (0.25 inch) segments. Another anode was prepared with 1.9 cm (0.75 inch) segments. The different sizes will be used to determine how segment length affects fuel dissolution in the electrorefiner. The loaded anode baskets are pictured in Figure 3.

This unirradiated fuel chopping test provided a conservative qualification of the element chopper. Because depleted uranium is not as brittle as irradiated fuel, chopping it requires about three times the force as

Figure 3. Fuel Dissolution Baskets (Anode Baskets) Loaded with Simulated Chopped Fuel

irradiated material. Additionally, sodium is not logged in the fuel matrix in the simulated fuel. With this segregated sodium phase, more gumming of the chopper blades occurred than is expected with irradiated fuel.

With the successful qualification of the element chopper, efforts are now being spent trying to reduce processing time. In processing a batch of material, 42 percent of the time is due to set-up including loading containers and preparing the machine, 38 percent is actual processing time, and 20 percent is associated with maintenance. The processing time is set by the design of the element chopper. The only real improvements in this process would involve changing the method of removing the spacer wire from the fueled section of the element and increasing the number of elements which can be processed at one time. These changes are being examined. The time required to set up the machine is primarily determined by the realities of working in a hot-cell environment, tracking material content, and remote handling. Optimization of this portion of the process will come naturally as the operators gain experience with the operating procedures. Additionally, some optimization will occur as some of the containers used in the element chopping process are redesigned to make them more operator-friendly.

The main thrust of element chopper operations for reducing batch process time has been to lessen the amount of routine maintenance required during processing. Ninety percent of this maintenance is a result of press vibration and component wear. Process vibration has caused problems with electrical connections and limit switches. Modifications to these components have been successfully made to lessen down time. Another maintenance item has been the replacement of the shear blades. Tool steels of various hardness are being examined for use as chopper blades to optimize their lifetimes.

C. Electrorefiner

The FCF electrorefiner was set up and qualified for operation during the fall of 1994. In December 1994 and January 1995, 431 kg of LiCl-KCl and 531 kg of cadmium were loaded in the vessel. The system then was heated to 500°C.

As increments of salt and cadmium were loaded, level measurements and volume calibration operations were performed.[4] These measurements are used as part of the materials control and accountability (MC&A) plan for the electrorefiner. During irradiated fuel operations, the inventory and associated uncertainties of special nuclear material in the electrorefiner must be established using both the analytical samples of salt and cadmium and the volume of these liquids as calculated from level measurements. The uncertainties associated with the liquid volumes were determined from the calibration operations. The volume of the salt can be determined with a 0.26% two sigma relative uncertainty.

During March 1995, depleted uranium was placed in the electrorefiner for the first time. Fuel dissolution baskets with 20 kg of uranium were loaded into the electrorefiner salt. Cadmium chloride was added to the salt, and the uranium was oxidized as follows:

$$2U + 3CdCl_2 \longrightarrow 2UCl_3 + 3Cd.$$

The presence of UCl_3 facilitates the electrochemical transport of actinides in the salt medium.

After charging the salt with uranium, seven salt samples were taken over a two-month period to determine the sampling and analytical uncertainty for uranium and to verify that uranium concentration in the salt was stable and not affected by system impurities. The relative sample standard deviation for the uranium concentration in the salt samples, given in Table 2, is 0.21%, or 0.42% at two standard deviations. These results indicate that the uranium concentration in the salt is stable and that any error associated with the sampling and analysis is small.

Table 2. Sample Results from the Electrorefiner Stability Test

Sample Description	Uranium Concentration (weight percent)
1st stability sample	4.27
2nd stability sample	4.28
3rd stability sample	4.28
4th stability sample	4.28
5th stability sample	4.26
6th stability sample	4.27
7th stability sample	4.26

During June 1995, the first electrotransport operations were performed. The feed was 14 kg of depleted uranium loaded in the fuel dissolution baskets. Three uranium solid cathodes were recovered. The first, provided in Figure 4, was a small product to verify that the system was functioning properly. After this product was viewed, it was placed back into the electrorefiner and electrochemically dissolved.

Figure 4. The First Solid Cathode Product Recovered from the FCF Electrorefiner

During July 1995, the third cathode recovered was removed from the electrorefiner and harvested from the solid cathode mandrel. This product was run through the cathode processor to remove adhering salt and then sampled in the casting furnace. The results from these samples combined with electrorefiner salt and cadmium samples were used to demonstrate close-out operations for a batch of treated fuel. These data were provided to DOE as part of a safeguards audit to determine if the facility was ready for operations with enriched irradiated fuel. The safeguards audit was successfully completed in August 1995.

Since the DOE audit, another 25 kg of depleted uranium have been processed through the electrorefiner. Most of the cathodes recovered from these materials have been recycled back to the electrorefiner. A total of 24 depleted uranium solid cathodes were recovered between June 1995 and March 1996. The largest of these cathodes, pictured in Figure 5, was 8.2 kg. Collection efficiencies greater than 40 percent have been obtained for some cathodes. The following processing parameters have been examined as part of these runs: salt mixing, cadmium mixing, cathode rotation rate, cell current/voltage, uranium salt concentration, cathode dimensions, and electrotransport configurations.

Figure 5. An 8.2 kg Depleted Uranium Solid Cathode Product

In March 1996, electrotransport with a depleted uranium-zirconium (10 weight percent) feed was started. More than 10 kg of unclad uranium-zirconium rods were added to the electrorefiner. To date one cathode has been recovered from this material. Chopped simulated fuel containing bond sodium will next be processed.

D. Cathode Processor - Casting Furnace

The operation of the cathode processor and casting furnace for distillation of adhering salt, consolidation of heavy metal ingots, and sampling has also been demonstrated using depleted uranium. The results from these tests are presented in other papers.[5]

V. SCHEDULE

An Operational Readiness Review for FCF was completed by DOE during the summer of 1995. In August 1995, the DOE Material Control and Accountability Audit was completed.

During the summer of 1995, several non-government organizations questioned the adequacy of the FCF environmental assessment. The resolution of environmental issues is all that is needed before operations with irradiated fuel can begin. Under the IFR program, an environmental assessment (EA) was

prepared for the operation of FCF.[6] During the fall of 1995, the decision was made to prepare a new environmental assessment to specifically address the spent fuel treatment operations in FCF.[7] For the spent fuel demonstration program that the EA addresses, 100 driver and 25 blanket fuel assemblies from EBR-II will be processed to demonstrate the electrometallurgical technique. This document was issued by DOE during late January 1996. Public hearings addressing the document were held during February in Idaho Falls, Idaho and Washington, DC. The public comment period closes in late March 1996, and the environmental evaluation is expected to be complete during spring 1996.

VI. CONCLUSIONS

In the Fuel Conditioning Facility, hot cell operations are being successfully demonstrated with the process equipment required for the treatment of EBR-II spent fuel. Simulated spent fuel containing depleted uranium-zirconium and bond sodium has been processed through the vertical assemble dismantler and the element chopper. Depleted uranium has been processed through the electrorefiner to produce 24 solid cathodes. Operations with depleted uranium-zirconium have also begun. Simulated spent fuel is all that remains to be processed in the electrorefiner before proceeding to irradiated fuel. Operations with irradiated fuel will commence upon resolution of the environmental evaluation for the facility.

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