SPENT FUEL TREATMENT AT ANL-WEST

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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 Cycle Facility (FCF) at ANL-West to produce stable waste forms for storage and disposal. The treatment operations will employ a pyrochemical process that also has applications for treating most of the fuel types within the Department of Energy complex. The treatment equipment is in its last stage of readiness, and operations will begin in the Fall of 1994.

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 has been undergoing modifications for the demonstration of the fuel cycle for the Integral Fast Reactor (IFR), an innovative liquid-metal-cooled reactor concept. 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. The refining operation was based on a pyrochemical operation employing molten salts and liquid metals.[3] The decision has now been made to terminate the operation of EBR-II, so plans for refining and fabricating new fuel have also been terminated.

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 high enriched uranium. For the safe storage and eventual disposal of EBR-II spent fuel, treatment operations are required. The treatment operations will neutralize the reactive sodium, stabilize the fission products, and recover the actinides in a stable form. These operations will be performed in the Fuel Cycle 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 will be treated in FCF. This fuel is currently stored at ANL-West in three locations: the EBR-II reactor vessel, the Hot Fuel Examination Facility (HFEF), or the Radioactive Scrap Waste Facility (RSWF).

The driver fuel is composed primarily of four different subassembly types: Mark-IIC, Mark-IICS, Mark-III, and Mark-IIIA. The bulk of this fuel is a uranium-(10 weight percent) zirconium alloy. All four subassembly types are composed of 61 fuel elements containing high 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 subassembly hardware are made of 316, D-9, or HT-9 stainless steel.

The approximate inventory at ANL-West is 58 Mark-II types and 163 Mark-III types. These

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subassemblies contain more than 800 kg of irradiated heavy metal. The average subassembly burn-up is approximately 10 atom percent, but some elements have burn-ups as high as 20 atom percent.

In addition to the standard driver subassemblies, there is a large quantity of experimental elements and subassemblies. 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 subassemblies. There are more than 420 blanket subassemblies containing approximately 19,500 kg of uranium, primarily depleted uranium. Additionally, they contain approximately 200 kg of plutonium.

III. TREATMENT OPERATIONS

A. Processing Operations

The fuel treatment operations are based on a pyrochemical process employing molten salts and liquid metals in an electrorefining 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 nonreactive metals.

In the electrorefining operation, the uranium and transuranics (TRU) will be separated from the bulk of the fission products. Uranium that is relatively free of TRU elements will be collected on steel mandrel cathodes (solid cathodes). Cathodes that consist of a ceramic crucible filled with approximately 26 kg of liquid cadmium (liquid cathodes) will be used to collect uranium and the TRU elements simultaneously. A small fraction of the lanthanide fission products will also be collected in the liquid cathodes, making the material unattractive for diversion.

As noted, FCF and portions of the equipment fabricated for the IFR fuel cycle demonstration will be used for treating EBR-II spent fuel. FCF consists of two operating hot cells. Figure 1 depicts FCF and the positioning of the equipment in the hot cells. Spent fuel will first be transferred into a rectangular-shaped, air-filled hot cell where the fuel elements will be separated from the subassembly hardware using the vertical assembler/dismantler (VAD). Intact fuel elements will be transferred into the adjacent, annular-shaped, argon-filled hot cell.

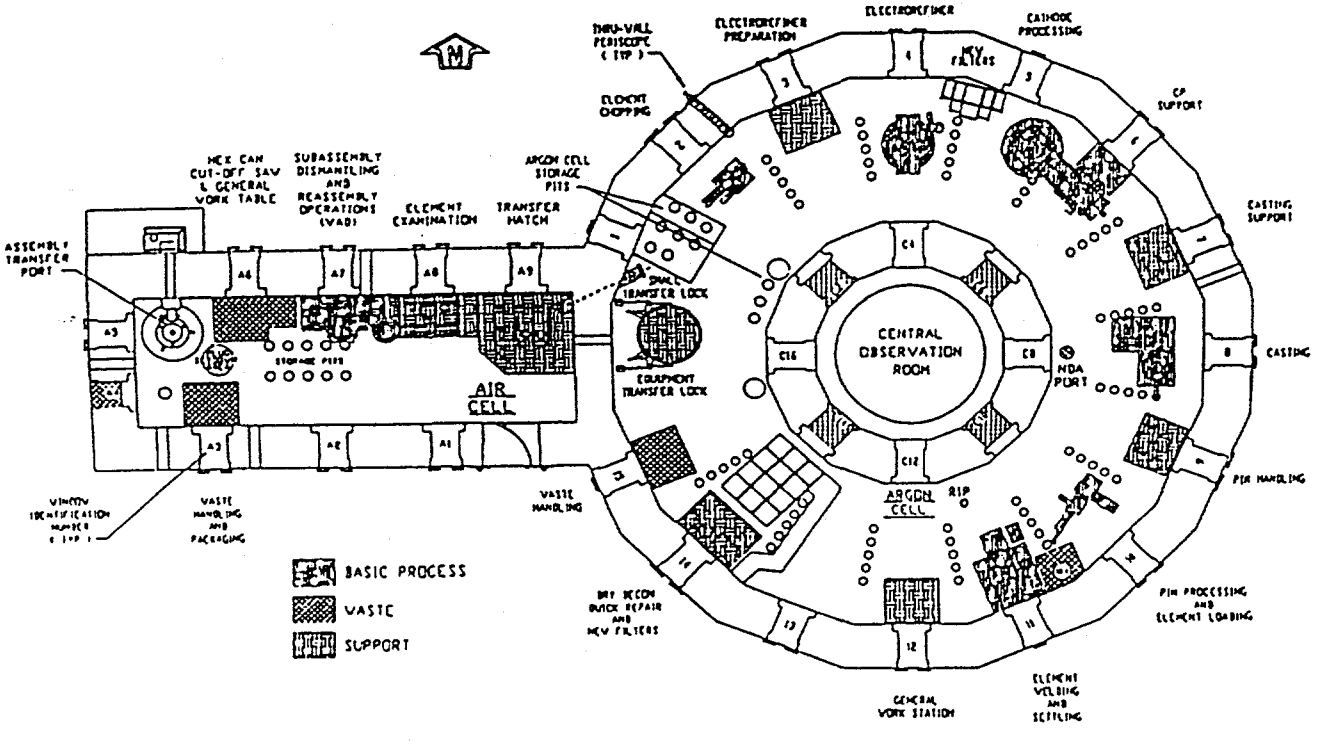

Figure 1 Air and Argon Hot Cells in the Fuel Cycle Facility

In the argon cell, the fuel elements are first chopped into 0.64 mm (0.25 inch) segments with the element chopper. The chopped segments are then transferred to the electrorefiner in steel baskets (anode baskets). 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 either a solid or liquid cathode (direct transport). Material deposited in the cadmium pool can later be electrotransported to a solid or liquid cathode.

The cathode products from electrorefining operations will be further processed in a separate operation to distill any adhering salt or cadmium and consolidate the remaining actinides which will be sampled for material control and accountability. These operations will be performed in the cathode processor and casting furnace.

The electrorefining operations have been demonstrated on laboratory and engineering scales in the Chemical Technology Division at Argonne in Illinois.[4] Those initial demonstrations used simulated fission products and small amounts of plutonium.

B. Waste Operations

In the electrorefining operations, most of the fission products will become concentrated in the salt and cadmium phases in the electrorefiner. The alkali, alkaline earth, rare earth, and halide fission products will be primarily in the salt phase. The sodium will be neutralized by forming non-reactive NaCl. The nobler metal fission products will be in the cadmium phase primarily as insolubles.

The electrorefiner is capable of passively dissipating approximately 6 kW of fission product heat. Once this limit is reached, the salt and cadmium will be treated to lessen the heat content. As part of the treatment operations, the cadmium and salt will first be pumped through a metal sintered filter to remove any insolubles and most of the noble metal fission products. 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. The resulting zeolite will be processed with additional zeolite and glass additives into a mineral waste form. Portions of the salt will be recycled.

The metal filter containing the noble metal fission products will be combined with the element and subassembly hardware and processed into a stainless steel based waste form. Zirconium recovered from the fuel alloy will also be added as a stabilizer.

In the treatment operations, the uranium and plutonium will be recovered as cathode products which are eventually consolidated into metal ingots in the cathode processor. The solid cathode products will consist of uranium that is relatively free of TRU. As part of the driver processing, the solid cathode will contain highly enriched uranium. Therefore, as part of the cathode processing operation, the recovered uranium metal will be blended with depleted uranium to produce a product that is less than 20 percent enriched. Both the uranium and the TRU products will be placed in interim storage as ingots in canisters pending a DOE decision on ultimate disposal.

IV. Equipment Status and Operation

The fuel treatment systems have been developed and installed in parallel with refurbishment of the facility. All of the major processing components are now in the hot cells. Operations in both cells are being performed using remote methods, and cell support and facility systems are in the final stages of installation or testing.

Subassembly handling will occur in the 65 square meter rectangular air cell. This cell was stripped of most equipment from previous missions and partially decontaminated. Refurbishment included servicing the lead-glass shield windows, replacing all the master-slave manipulators, and reconditioning the overhead crane and both bridge type manipulators.

In the air cell, a vertical assembler-dismantler (VAD) removes the fuel elements from the subassemblies. The VAD is the only processing system retained from earlier fuel cycle work. This system has been cleaned and reassembled with new electrical drive and control components. Also in the air cell, a work table has been installed at the east wall to support element handling and transfers between the cells. Here fuel elements are loaded into magazines before movement to the argon cell.

Material is moved between cells via a small transfer lock, which can accommodate items of mass less than 500 kg with dimensions to 2.8 m long by 0.3 m diameter. Larger items, of mass up to 4536 kg and dimensions to 1.83 m diameter by 2.44 m height, can be moved to the argon cell through the Equipment Transfer Lock, which is connected to the air cell and a component repair area via a tunnel in the facility basement. Testing of the transfer and air cell systems was completed in September 1994.

The annular argon cell has a work area of approximately 185 square meters and is served by 15 workstations, 14 of which are equipped with various

master-slave manipulator configurations. Fuel and equipment movements utilize four bridge-type manipulators and two overhead cranes. This cell was stripped of equipment, partially decontaminated, the shield windows serviced, and all remote handling equipment replaced or refurbished. Work in the argon cell has been accomplished entirely by remote means since September 1993.

Prior to installation in the hot cells, equipment and systems go through three phases of qualification. After a standard design process and parts fabrication, phase 1 activities assemble components by hand and functionally test the system by the responsible engineering group. The second phase includes assembly, operation with a surrogate material, maintenance, and disassembly, all by remote methods in a full scale out of cell mock-up facility. The phase 2 checks are conducted by experienced remote handling personnel and typically result in many small but important modifications to make the system more operator friendly in-cell. Here also, the operations technicians receive their first introduction to the new systems. Phase 3 is conducted in-cell by the operations crews. After assembly and control system check out, the final qualifications conclude with the processing of depleted uranium.

Fuel elements arrive in the argon cell in cylindrical magazines which mount on the element chopper, where fuel bearing regions of individual elements are converted to 0.64 mm segments by a solenoid driven punch press. Remaining element hardware is segregated for later processing into the metal waste form. The fuel segments are collected in perforated baskets, four of which will be combined to form an anode assembly for the electrorefiner.

In-cell qualification of the element chopper is complete to the point of processing the depleted uranium elements for the first electrorefiner run, which is expected to occur in November 1994. Early experience has shown that a chopping rate of 200 elements a week can be expected. Currently, the element chopper is being run for operator training using surrogate materials.

Perforated steel baskets containing chopped element segments are combined to form an electrorefiner anode assembly at the Electrode Assembly/Disassembly Machine (EADM), located at the argon cell workstation between the element chopper and the electrorefiner. Here also, cladding hulls left after electrorefining will be removed from the anode baskets for processing into the metal waste form. Additionally, both solid and liquid cathodes will be collected and prepared for further processing at this station. In-cell assembly of the EADM is complete and check out of the controls should be finished by mid October 1994.

The electrorefiner supports four electrode assemblies (EA). Full operation will consist of two anode assemblies, each with up to 10 kg of heavy metal, transporting to two cathode assemblies. The fuel and process chemicals are isolated from the cell atmosphere by a pure argon cover gas. The ER completed out of cell qualification in May 1994, and is now assembled in the argon cell.

After processing at the EADM, recovered uranium and TRU products from the electrorefiner are transferred to the cathode processor for purification. The cathode product is placed into a graphite crucible and inductively heated in a vacuum to drive off residual salts and cadmium. The cathode processor was installed in the argon cell April 1994. Hot operation tests (empty) were successfully conducted through August 1994. Hot tests on depleted uranium are scheduled for October 1994, and processing of the first ER cathodes should occur in early December 1994.

Cathode ingots are melted once more during sample casting operations. The molten material is forced into an evacuated quartz mold and cooled to produce a pin that will be used as a representative sample of the ingot. The casting furnace has been operational in-cell since October 1993, and has successfully produced four castings of depleted uranium. The pins produced in these operations have been used to conduct the in cell qualification of the material control and accountability systems and general nuclear material handling systems. They will also be used for the depleted uranium elements to qualify the element chopper and the electrorefiner.

V. SCHEDULE

A. Processing Operations

Start-up of the spent fuel treatment operations is on-going. The first material to be processed through the system, as part of start-up operations, will be simulated fuel made with depleted uranium. It will be sodium bonded and contain the uranium-zirconium alloy. The fuel slugs for this simulated fuel have already been cast using equipment in FCF and the Fuel Manufacturing Facility (FMF) at ANL-West. The fabrication of the fuel slugs into fuel elements will begin at the end of September 1994.

In early October 1994, the FCF electrorefiner will be loaded with 427 kg of LiCl-KCl eutectic salt and 533 kg of cadmium. During loading, an in-cell volume calibration will be performed for materials control and accountancy. Loading and hot operations testing of the equipment will be completed during October and electrorefiner salt will be charged with depleted uranium

metal to form UCl_3 in the salt phase, which is required for electrotransport. The depleted uranium operations are scheduled to begin in November 1994.

For the start-up operations of the electrorefiner, at least 20 kg of simulated fuel will be processed. This material will have been chopped earlier as part of start-up for the element chopper.

The start-up operations for the cathode processor and the casting furnace will consist of processing the cathodes recovered as part of the electrorefiner start-up. These operations are scheduled for December 1994. Hot tests in the cathode processor with depleted uranium will occur earlier in October.

Because the present equipment was designed primarily for processing driver subassemblies, they will be processed before the blanket subassemblies. Once start-up operations are completed and DOE grants approval for operations with irradiated fuel, driver subassemblies will be brought into the hot cells and spent fuel treatment operations will begin. The current air permit granted by the state of Idaho for the operation of FCF limits processing operations to 90 driver subassemblies per year. Concurrent with driver processing, advanced equipment will be designed and fabricated for use in treating the larger mass of blanket subassemblies.

B. Waste Operations

The design and fabrication of equipment for producing hot mineral and metal waste forms samples is in progress. The Bulk Fluid Handling System, which will be used to pump and filter salt and cadmium and to contact the salt with zeolites, is in the final stages of design. The zeolite products will be converted into waste form samples which are solid monoliths using a hot isostatic press (HIP). The specifications for this piece of equipment have been determined and a model has been found. Space for the HIP is being readied in the Hot Fuel Examination Facility, a hot-cell complex next to FCF at ANL-West. Hot metal waste form samples will be fabricated using the casting furnace in FCF. The required furnace modifications are currently being developed.[5]

Hot materials will be available for fabricating waste forms samples by the end of the first year of operations.[6] A number of waste form samples will be prepared and subjected to the following tests: PCT (Product Consistency Test), MCC-1 (Material Characterization Center), TCLP (Toxicity Characteristic Leachate Procedure), and corrosion tests. The MCC-1 and the PCT tests are standard tests developed for leaching studies of vitrified high-level waste. The TCLP test is used to determine if a waste stream is classified as a hazardous waste. The results of these tests will be used in the design of full-scale waste form equipment.

VI. APPLICATION TO OTHER FUEL

The primary purpose of electrorefining spent EBR-II fuel is to neutralize the reactive sodium that is bound in the fuel matrix. The treatment operation provides a number of additional benefits with respect to the disposal of high level waste. In the electrorefining operation, the fission products are separated from the long lived TRU material and uranium and placed in a waste form suitable for disposal as high level waste. For the EBR-II driver fuel, avoiding direct disposal of the actinides is crucial because of potential criticality concerns associated with highly enriched spent fuel.

A number of other spent fuels within the DOE complex have similar problems with respect to direct disposal. Several reactors use high enriched uranium. Spent fuel from the Fermi reactor and EBR-I contain bond sodium, and there is additional EBR-II fuel stored at sites other than ANL-West. These fuel types will also need to be treated for a safe final disposition.

The result of the treatment operations for EBR-II spent fuel treatment will be two waste forms that will be licensed for disposal. At present, glass and commercial light water reactor fuels are the only waste forms in the advanced stages of disposal qualification; this process has been quite expensive. If direct disposal of the many other types of DOE spent fuel is chosen, then a number of disposal forms will need to be qualified. To eliminate this expensive undertaking, most of the DOE spent fuel could be pyroprocessed into the same mineral and metal waste forms that already must be produced.

Spent fuel from the N-reactor comprises the largest mass of the DOE inventory. Like EBR-II, its spent fuel is metallic and can be directly electrorefined.[7] The quantity of high level waste can be greatly reduced by separating the uranium for disposal as low level waste.

Oxide fuels, which comprise a large fraction of the remaining DOE inventory and virtually all of the commercial spent fuel inventory, can be electrorefined after the addition of a front-end reduction operation.[8] The demonstration of this operation has already been performed at Argonne's Chemical Technology Division in Illinois.[9] Use of this single treatment method could greatly simplify the disposal process for DOE spent nuclear fuel since it would result in just two high level waste forms instead of one for each type of spent fuel.

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