

ELECTROMETALLURGICAL TREATMENT OF EBR-II SPENT FUEL

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ABSTRACT

This paper will detail the results of the EBR-II Spent Fuel Treatment Program at Argonne* National Laboratory. This program is responsible for the treatment of sodium-bonded spent fuel from the operation of the Experimental Breeder Reactor II. An electrometallurgical technology is used to neutralize the sodium in the fuel and produced high-level waste for geological disposal. The EBR-II Spent Fuel Treatment Program has been ongoing since FY2000 after successful completion of a three-year demonstration. In FY2003, this program was incorporated into the Advanced Fuel Cycle Initiative within the Department of Energy's Office of Nuclear Energy, Science and Technology.

INTRODUCTION

In September 2000, the production-scale treatment of sodium-bonded spent nuclear fuel from the operation of the Experimental Breeder Reactor II (EBR-II) commenced at Argonne National Laboratory-West (ANL-West). Because of the presence of elemental sodium within the individual fuel elements, this fuel is not viewed as being acceptable for direct disposal in the proposed repository at Yucca Mountain. An electrometallurgical technology is employed for the treatment operations. It results in waste forms acceptable for disposal in the repository. This treatment program is part of the Advanced Fuel Cycle Initiative (AFCI) within the Department of Energy's Office of Nuclear Energy, Science and Technology (DOE-NE). Included in this program are activities to expand the capabilities of the technology to aid in further reducing the time required to treat sodium-bonded fuel and to provide a technology for potentially treating other fuel types.

Production-scale treatment operations were initiated after the successful demonstration of the technology through the treatment of a representative quantity of EBR-II fuel between 1996 and 1999 [1], and after completion of a non-proliferation assessment [2], an independent cost study [3], and an environmental impact statement [4]. During the demonstration phase of this program the Committee on Electrometallurgical Techniques for DOE Spent Nuclear Fuel was formed within the National Research Council to evaluate the technical viability of the process. This committee reviewed the progress of the ANL activities and issued ten reports. In their final report, issued after the demonstration was completed, they included the following two findings:

“Finding: The committee finds that ANL has met all of the criteria developed for judging the success of its electrometallurgical demonstration project.

Finding: The committee finds no technical barriers to the use of electrometallurgical technology to process the remainder of the EBR-II fuel.”[1]

They also included the following recommendation:

“Recommendation: If DOE wants an additional option besides PUREX for treating uranium oxide spent nuclear fuel, it should consider continued development and implementation of the lithium reduction step as a head-end process to EMT.”[1]

The Record of Decision (ROD) authorizing continuation of the Spent Fuel Treatment Program was issued in September 2000 [5]. The scope of the program includes treatment of 24,750 kg of heavy metal in the form of EBR-II driver and blanket fuels.

Since the program was established, all annual milestones established by DOE-NE have been completed on time. More than 3 metric tons heavy metal (MTHM) of fuel has been treated in the Fuel Conditioning Facility (FCF) at ANL-West. Fuel treatment operations are on going along with process testing to further increase throughput and reduce the costs to treat the remaining inventory of EBR-II fuel. Technology development work is also being performed to complete the qualification of the two high-level waste forms resulting from electrometallurgical treatment.

In addition to treatment of EBR-II metallic fuel, a limited amount of work was also performed in FY2003 to assess the treatment of commercial oxide fuel by a pyrochemical process. A small-scale demonstration of this technology was performed with irradiated fuel in a hot cell at ANL-West.

FUEL PROCESSING

The electrometallurgical process can be divided between fuel treatment operations and high-level waste operations. Two types of sodium-bonded fuel, driver and blanket, have been treated. The driver fuel (63% U-235) was irradiated to a relatively high burnup (approximately 8 atom percent). The irradiated blanket fuel was depleted uranium with a low burnup (approximately 0.2 atom percent). Both fuel types are metallic.

Fuel treatment operations are performed in the Fuel Conditioning Facility (FCF), pictured in Figure 1, a hot-cell complex that consists of two adjacent hot cells. Spent fuel is first transferred into a rectangular-shaped, air-filled hot cell where the fuel elements are separated from the fuel assembly hardware using the vertical assembly dismantler. Intact fuel elements are transferred into the adjacent, annular-shaped, argon-filled hot cell. In the argon cell, fuel elements are first chopped into segments with an element chopper. These segments are then transferred to an electrorefiner in steel baskets. The electrorefiners contain a molten salt medium of LiCl-KCl eutectic and dissolved actinide chlorides, such as UCl_3 and PuCl_3 . For both electrorefining and fuel chopping, separate equipment is used for blanket and driver fuel.



Fig. 1 Fuel Conditioning Facility hot cell complex at ANL-West.

In the electrorefiners, the spent fuel is electrochemically dissolved from the anode baskets, and an equivalent amount of uranium is deposited on a cathode. The uranium is separated from the bulk of the fission products and transuranics. Most of the fission products (alkali, alkaline earth, rare earth, and

halides) and transuranics accumulate in the salt. The bond sodium is neutralized by forming non-hazardous NaCl. The cathode products from electrorefining operations are further processed to distill adhering salt and to recover uranium. These operations are performed in the cathode processor. As part of the driver fuel processing, the recovered uranium is blended with depleted uranium in the casting furnace to produce a product that is less than 20 percent enriched. The low enriched uranium product is formed into ingots and placed in interim storage in canisters at ANL-West pending a DOE decision on final disposition. The electrorefiner and cathode processor operations are described in more detail in other papers [7,8].

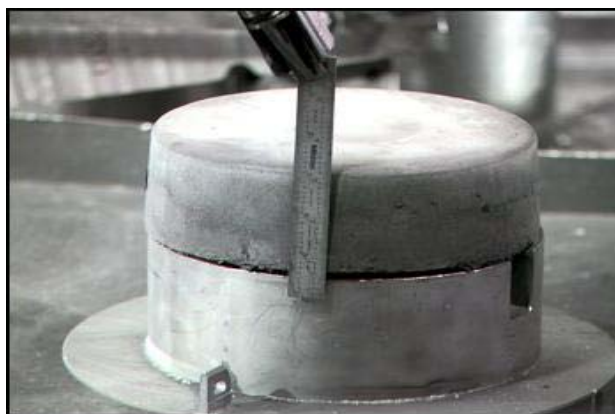


Fig. 2 Low enriched uranium product

Activities over the past two years have focused on meeting the production milestones established with DOE and on performing the research, development, and engineering required to increase process throughput. The baseline treatment rate established with DOE for both FY2001 and FY2002 was 540 kg of heavy metal (kgHM). This rate is lower than the potential throughput capacity because funding limits operations to 5 days per week and 8 hours per day. In FY2001, this milestone was exceeded when 600 kgHM were treated. In FY2002, 615 kgHM were treated.

In FY2003, the Spent Fuel Treatment Program was combined with the Advanced Accelerator Applications (AAA) program to become part of the Advanced Fuel Cycle Initiative (AFCI). Like the EBR-II Spent Fuel Treatment Program, the AAA Program had elements that employed pyrochemical processing for the treatment of spent nuclear fuel. Combining these two programs took advantage of these synergistic activities.

Once a part of AFCI, the activities of the EBR-II Spent Fuel Treatment Program expanded to support not only treatment of the EBR-II fuel, but also technology development activities associated with the treatment methods for other fuel types. The treatment of EBR-II fuel and operations established for this program were used to facilitate the development of additional technologies. Still, in support of this development work, 540 kgHM was treated in FY2003.

Part of the technology development activities is focused on increasing the process throughput for the remaining EBR-II fuel. Significant advancements have been made since starting the production program in FY2001. During the demonstration, the batch size for driver fuel was two assemblies, or approximately 8.2 kgHM. New baskets for processing driver fuel have now been designed and tested that hold 3 driver assemblies. The benefit of this change is a substantial reduction in handling operations and overall hot cell manipulator usage. Repairs of manipulators due to routine usage cause significant downtimes.

At the end of the demonstration, the electrorefiner rate for blanket fuel per anode-cathode assembly was less than 300 gHM per hour. Through testing this rate has been increased to more than 600 gHM per hour. These improvements were made in large part by changing the method in which the baskets containing the spent fuel were rotated in the electrorefiner with respect to the cathode in the system.

The uranium recovered from both driver and blanket treatment is processed through the same cathode processor to remove adhering salt and produce a consolidated ingot. With both process streams sharing this single piece of equipment, it has the potential to be a bottleneck in reaching target processing rates. To minimize this impact, the equipment is being modified so that the batch size for blanket material can be increased from 30 to 64 kgHM.

In addition to these changes to existing process equipment in FCF, more significant modifications to electrorefiner designs are being assessed that could potentially increase process throughput for other fuel types e.g., oxides, by orders of magnitude. These design modifications include a planar-type geometry instead of the cylindrical designs used in existing equipment. The application of these designs to the treatment of EBR-II fuel in FCF is being examined to determine if the treatment duration can be reduced significantly. In FY2004, treatment operations are focused on driver fuel while these higher-throughput options are assessed for blankets.

HIGH-LEVEL WASTE

Electrometallurgical treatment of spent nuclear fuel for disposition results in two high-level waste (HLW) forms, the ceramic waste form and the metal waste form. The operations for the production of the HLW occur in the Hot Fuel Examination Facility (HFEF) hot cell complex adjacent to FCF.

The ceramic waste form, which stabilizes the electrorefiner salts, is a glass-bonded sodalite produced from the thermal conversion of zeolite A. The salts are occluded into the zeolite structure in a heated V-mixer. The capacity of the operating V-mixer in HFEF is 112 kg. It rotates at 17 rpm and can be heated to more than 500°C, which facilitates the salt occlusion process. After the salt is occluded in the V-mixer, the salt-loaded zeolite is mixed with 25% glass frit. This mixture is loaded into a canister and then consolidated into a monolithic waste form in a furnace at 915°C.

The metal waste form consists of metallic ingots that are used to stabilize noble metal fission products, non-actinide fuel matrix and cladding materials. Minor amounts of actinides that remain in the cladding hulls after dissolution are also present. Zirconium metal is added to improve performance properties and to produce a lower melting point alloy. The typical composition is stainless steel and 15 weight percent zirconium. It is produced in a casting operation at 1600°C. Waste operations are described in more detail in other papers [9,10].

During the demonstration program, the production of HLW forms with irradiated materials was demonstrated in HFEF. Equipment for full-scale production operations is now being developed as part of the on-going program.

For the ceramic waste, zeolite A is obtained from a commercial vendor, but it must first go through a sizing process and be dried before use in the ceramic waste. Equipment has been placed into operation for these tasks which occur outside the hot cells. The zeolite is sized using a mill/classifier from Prater Industries, Inc. It is then sieved. The sized zeolite is dried using a rotating furnace fabricated at ANL with a capacity of 160 kg.

A prototype production furnace for the ceramic waste is being procured in FY2004. This furnace will be capable of operations up to 1000 °C and will be used to produce ceramic waste ingots up to 400 kg in

mass. This furnace will be used to produce surrogate waste form samples starting in FY2005 for qualification of the production process. Simultaneous with this testing program, an in-cell furnace will be prepared based on the testing results.

During the demonstration, eight metal waste form ingots were produced from irradiated materials for subsequent characterization. These ingots were all smaller than 10 kg, and they were produced using the cathode processor and casting furnace in FCF. For production operations a dedicated piece of equipment is required. A prototype production metal waste form furnace was built and placed into operation in FY2002. The production ingot size is approximately 60 kg. This equipment is presently undergoing process testing in a uranium glovebox while the in-cell equipment is being readied for installation in HFEF. The check out of the production furnace, shown in Figure 3, should be completed in FY2004.

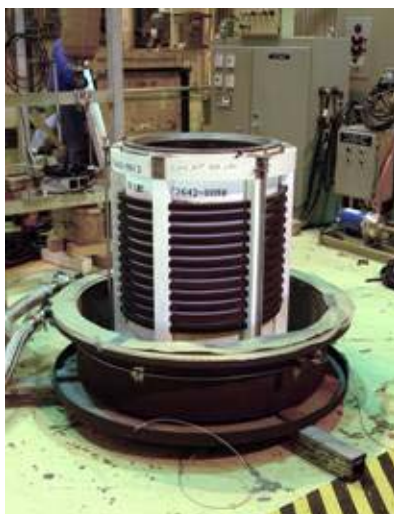


Fig. 3. Production Metal Waste Form Furnace during mock-up testing.

Establishment of the disposal path for both HLW waste forms was an FY2001 milestone. In the House of Representatives Report 106-693, accompanying House Resolution 4733, the Energy and Water Development Appropriations Bill, 2001, the House Committee on Appropriations requested DOE to prepare a report on the disposition of the waste streams [11].

This report was issued in March 2001 [12]. In it the ceramic and metal waste forms were both formally identified as HLW that will be sent to a geological repository for disposal. The report was signed by DOE's Director of Nuclear Energy, Science, and Technology, Director of Civilian and Radioactive Waste Management, and the Assistant Secretary for Environmental Management. For the program as defined in the report, 59 DOE standardized canisters will be sent to a repository.

TREATMENT OF OXIDE FUEL

In addition to increasing pyrochemical process throughput, FY2003 APCI direction supported laboratory-scale demonstrations of head-end processes to expand the application of pyrochemical fuel treatment to commercial light water reactor oxide fuel. This demonstration employed actual spent fuel. The development of an oxide reduction process whereby spent oxide fuel could be converted to a metallic form suitable for pyrochemical treatment has been ongoing at ANL since the 1990s. [13, 14, 15]

The oxide reduction process can involve reduction of uranium oxide by either chemical or electrochemical means. In the case of chemical reduction, a Li metal reductant may be used, while electrochemical treatment employs an applied voltage. Laboratory-scale testing performed at ANL in FY2003 utilized electrochemical reduction to convert oxide fuel from the Belgium Reactor 3 (BR3) to a metallic form.

The test with spent fuel was performed in the hot fuel dissolution apparatus (HFDA), housed in the inert environment HFEF hot cell. The test apparatus consisted of a liter-scale molten salt bath equipped with appropriate power supplies, electrodes, sampling ports and data acquisition systems to support a variety of test configurations.

The tests were performed in a 650° C molten LiCl salt containing 1 wt% Li₂O. A total of 5 tests were performed using both depleted uranium oxide (DUO₂) and spent BR-3 fuel. In both cases, approximately 50 g of DUO₂ or crushed fuel were loaded into a permeable basket that served as the cathode, while a platinum wire served as the anode. Upon application of the appropriate voltage between the anode and cathode, uranium oxide present in the fuel was converted to a metal at the cathode, while oxygen gas was liberated at the anode. After sufficient current was passed through the cell, the basket was recovered from the molten salt and the resulting material was visually examined then subjected to chemical analysis. The sectioned basket of the fuel is pictured in Figure 4. Visually, the reduced fuel samples showed a metallic appearance. Chemical analysis indicated up to 98% reduction of the fuel and depleted uranium oxide samples. These test results provide key evidence that pyrometallurgical treatment is applicable to both metal and oxide fuel types.



Fig. 4 Sectioned basket of spent oxide fuel reduced to metal.

CONCLUSIONS

Electrometallurgical treatment has progressed from a demonstration technology to a production process for treating sodium-bonded spent nuclear fuel. Fuel treatment rates are being maintained while research and development activities are supported to further increase process throughputs and to expand the applicability of pyrochemical treatment to other fuel types. Additionally, HLW form production processes are being designed, tested, and implemented.

REFERENCES

- 1 National Research Council, "Electrometallurgical Techniques for DOE Spent Fuel Treatment: Final Report," National Academy Press, Washington, DC (2000).
- 2 U.S. Department of Energy, Office of Arms Control and Nonproliferation, "Nonproliferation Impacts Assessment for the Treatment and Management of Sodium-Bonded Spent Nuclear Fuel" (July 1999).
- 3 "Cost Study of Alternatives Presented in the Draft Environmental Impact Statement for the Treatment and Management of Sodium-bonded Spent Nuclear Fuel" (August 1999).
- 4 U. S. Department of Energy, "Final Environmental Impact Statement for the Treatment and Management of Sodium Bonded Spent Nuclear Fuel," DOE/EIS-0306 (July 2000).
- 5 Federal Register, Vol. 65, No. 182/56565 (September 19, 2000).
- 6 U. S. Department of Energy, Office of Nuclear Energy, Science and Technology, "A Report to Congress on Electrometallurgical Treatment Waste Forms," (March 2001).
- 7 D. Vaden, S. X. Li, and T. A. Johnson, "Electrometallurgical Processing Of Experimental Breeder Reactor-II Fuel," Proceedings of the Fifth Topical Meeting on DOE Spent Nuclear Fuel and Fissile Materials Management, Charleston, SC, September 17-20, 2002.
- 8 B. R. Westpahl, D. Vaden, T. Q. Hua, J. L. Willit, and D. V. Laug, "Recent Developments at the Cathode Processor for Spent Fuel Treatment," Proceedings of the Fifth Topical Meeting on DOE Spent Nuclear Fuel and Fissile Materials Management, Charleston, SC, September 17-20, 2002.
- 9 M. F. Simpson, K. M. Goff, S. G. Johnson, K. J. Bateman, T. J. Battisti, K. L. Toews, S. M. Frank, T. L. Moschetti, T. P. O'Holleran, and W. Sinkler, "A Description of the Ceramic Waste Form Production Process from the Demonstration Phase of the Electrometallurgical Treatment of EBR-II Spent Fuel," Nuclear Technology, Volume 134, pp. 263-277 (June 2001).
- 10 D. D. Keiser, S. G. Johnson, and W. L. Ebert, "Monitoring the Consistency of the Metallic Waste Form Derived from Electrometallurgical Processing," Proceedings of the Fifth Topical Meeting on DOE Spent Nuclear Fuel and Fissile Materials Management, Charleston, SC, September 17-20, 2002.
- 11 Conference Report, Energy and Water Appropriations FY2001.
- 12 U. S. Department of Energy, Office of Nuclear Energy, Science and Technology, "A Report to Congress on Electrometallurgical Treatment Waste Forms," (March 2001).
- 13 E.J. Karell, K.V. Gourishankar, J.L. Smith, L.S. Chow, L. Redey, "Separation of Actinides from LWR Spent Fuel using Molten-Salt Based Electrochemical Processes," Nuclear Technology, Vol. 136, pp. 342-353, (Dec. 2001).
- 14 K. Gourishankar, L. Redey, and M. Williamson, "Electrochemical Reduction of Metal Oxides in Molten Salts", Light Metals, ed., W. A. Schneider, (Warrendale, PA: The Minerals, Metals, and Materials Society, 2002), 1075-1082.

- 15 S.X. Li, S.D. Herrmann, M.F. Simpson, D.R. Wahlquist, "Electrochemical Reduction of Uranium Oxide Fuel in a Molten LiCl/Li₂O System," Proceedings of GLOBAL 2003, New Orleans, LA, November 16-20, 2003.

FOOTNOTES

- * The submitted manuscript has been created by the University of Chicago as Operator of Argonne National Laboratory under contract No. W-31-109-ENG-38 with the U.S. Department of Energy. 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.