

**PRODUCTION ELECTROMETALLURGICAL TREATMENT
OF EBR-II SPENT FUEL**

by

K. M. Goff, R. W. Benedict, G. M. Teske and T. J. Johnson

Engineering Technology Division and Facilities Division
Argonne National Laboratory-West
P. O. Box 2528
Idaho Falls, ID 83403-2528

The submitted manuscript has been created by the University of Chicago as Operator of Argonne National Laboratory (Argonne®) 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.

To be Presented
at the American Nuclear Society Fifth Topical Meeting
DOE Spent Nuclear /Fuels and
Fissile Materials Management

Charleston, South Carolina

September 17 – 20, 2002

*Work supported by the U.S. Department of Energy, Office of Nuclear Energy, Science and Technology, and the Office of Environmental Management under Contract W-31-109-Eng-38.

PRODUCTION ELECTROMETALLURGICAL TREATMENT OF EBR-II SPENT FUEL

K. M. Goff
R. W. Benedict
G. M. Teske
T. J. Johnson

Argonne National Laboratory - West
P. O. Box 2528
Idaho Falls, ID 83403
(208) 533-7084
goff@anl.gov

ABSTRACT

Following the successful demonstration of electrometallurgical treatment, the Spent Fuel Treatment Program was established at Argonne National Laboratory* (ANL) to treat sodium-bonded spent nuclear fuel. The treatment of 24,750 kg of heavy metal is included in this program. Production treatment operations begin in September 2000. The program also includes additional research and development activities to increase process throughput and to obtain final qualification of the resulting high-level waste. Through two years, all Department of Energy (DOE) milestones established for the program have been met or exceeded.

I. INTRODUCTION

From June of 1996 through August 1999, a demonstration of electrometallurgical treatment of was performed at ANL.¹ The purpose was to demonstrate the technical viability of the process as a method for conditioning DOE spent nuclear fuel for disposal.

* 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.

Sodium-bonded fuel from the Experimental Breeder Reactor II (EBR-II) at ANL-West in Idaho was treated during the demonstration. Sodium was used within the fuel elements to provide a thermal bond between the fuel matrix and cladding. This sodium metal is highly reactive. Because of its presence, the fuel is generally believed to be unsuitable for direct disposal in a geological repository and to require treatment.^{2,3} Table 1 provides a list of the sodium-bonded spent fuel in the DOE complex.

Table 1. Sodium-Bonded Spent Nuclear Fuel

Fuel Type	Mass of Heavy Metal (MT)	Storage Location
EBR-II Driver (alloyed w/zirconium)	1.1	ANL-West
EBR-II Driver (Fissium alloy)	2.0	Idaho Nuclear Technology and Engineering Center (INTEC)
EBR-II Blanket	22	ANL-West
Fermi Blanket (alloyed with molybdenum)	34	INTEC
FFTF Test Assemblies	0.25	Hanford

During the three-year demonstration 100 EBR-II driver fuel and 13 EBR-II blanket assemblies were treated. The demonstration of waste forms for stabilizing the fission products and transuranics was part of this project.

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 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.”⁴

They also included the following recommendation:

“Recommendation: If DOE want 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.”⁴

After the successful demonstration, a non-proliferation assessment was performed on the technology,⁵ an independent cost study was completed,⁶ and an environmental impact statement was issued.⁷ Based on these reviews, DOE issued a Record of Decision (ROD) for the treatment and management of their sodium-bonded fuel in September 2000.⁸

The ROD calls for all sodium-bonded spent nuclear fuel, except for the Fermi blanket fuel, to be electrometallurgically treated at Argonne National Laboratory-West (ANL-W). Because of different characteristics of the Fermi blanket fuel, DOE has decided to continue to store this fuel while treatment options are assessed.⁸

With the issuance of the ROD, a program was established at ANL to coordinate the treatment of the 25 MTHM. The program also includes additional research and development work to increase process throughput and to obtain final qualification of the resulting high-level wastes. This program is managed by DOE's Office of

Nuclear Energy, Science, and Technology (DOE-NE) with oversight by the Chicago Operations Office (DOE-CH).

II. PROGRAM ORGANIZATION

The first steps in transitioning from the demonstration program to more production oriented operations were establishing a program organization and then developing an implementation plan. The new project was titled the Spent Fuel Treatment Program. The implementation plan was developed and approved by both ANL and DOE-NE in October 2000. The plan outlines the status of the technology now and plans for making improvements to increase the processing rates.

To evaluate production throughput a process model was developed and validated during the demonstration program. With the model, the impact of process improvements were assessed and potential bottlenecks identified. At the end of the demonstration, the operating system in FCF was capable of treating 2200 kg of heavy metal per year at a staffing level to support operations at 24 hours per day and seven days per week. The goal of the program was to increase the rate to 5000 kg of heavy metal per year. Details for making these improvements as outlined in the plan are discussed later.

The Implementation Plans also details milestones for the program, various reporting methods to DOE, and performance measures. The milestones included goal processing rates on an annual basis. With these established processing rates and anticipated improvements, total treatment times were determined based on guidance from DOE concerning staffing levels and anticipated funding. Initial staffing levels allow operations at 8 hours per day and 5 days per week that support a treatment rate of 540 kg of heavy metal per year. A transition will gradually occur to 12 hours per day 7 days per week and then again to 24 hours per day. With these data, treatment times and costs were established. These costs included research and development costs and waste qualification costs. The total duration of the program as initially established is 13 years.

After the ROD was issued in September 2000, the Spent Fuel Treatment Program was organized and production treatment operations resumed the same month. The program is now in the second year. Though the first year of operations all milestones were met.

III. FUEL TREATMENT STATUS

A. Process Description

The 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).

Fuel treatment operations are performed in the FCF hot-cell complex that consists of two 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.

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 and casting furnace, respectively. As part of the driver fuel processing, the recovered uranium is blended with depleted uranium 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.

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. The electrorefiner, cathode processor, and waste operations are described in more detail in other papers.^{9,10, 11, 12}

B. Treatment Results

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. Based on the treatment rate through April 2002, the milestone is again expected to be exceeded for FY2002. From both the demonstration and production program more than 2 MTHM has been electrometallurgically treated in FCF.

In developing the implementation plan, a process to increase process throughput focused on the following areas:

- (1) Increasing the electrorefiner batch size for driver fuel processing,
- (2) Increasing the electrorefining rate for blanket fuel,
- (3) Increasing the cathode processor batch size for blanket fuel,
- (4) Increasing the electrorefiner batch size for blanket fuel processing,
- (5) Decreasing handling operations for the cathode processor crucibles, and
- (6) Increasing the recovered uranium product density from blanket fuel processing.

Substantial advances were made over the last two years on the first three tasks.

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 baskets are pictured in Figure 1. The benefit of this

changes is a substantial reduction in handling operations and overall hot cell manipulator usage. Repairs of manipulators due to routine usage causes significant downtimes.

At the end of the demonstration, the electrorefiner rate for blanket fuel 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

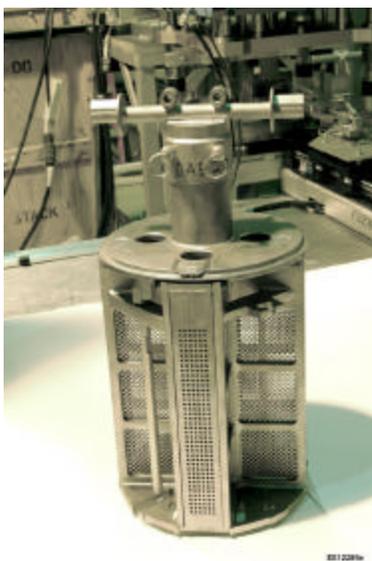


Figure 1. New Fuel Dissolution Baskets for Driver Fuel.

cathode in the system. These electrorefiner improvements are discussed in another paper.⁹

The uranium recovered from both driver and blanket processing are 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 the goal 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. Figure 2 shows the new and old crucible sizes for comparison. Making this modification alone will allow the process throughput to potential increase by 1000 kgHM per year. The equipment for this change is being fabricated and is expected to be placed into service in FY03.



Figure 2. New and Old Crucible for Uranium Processing
C. High-Level Waste Results

During the demonstration program, the production of HLW forms with irradiated materials was demonstrated in the Hot Fuel Examination Facility (HFEF) at ANL-West. 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. Its batch size is 160 kg. The mill/classifier and zeolite dryer are pictured in figures 3 and 4, respectively.



Figure 3. Mill/Classifier and Sieve for Zeolite Processing

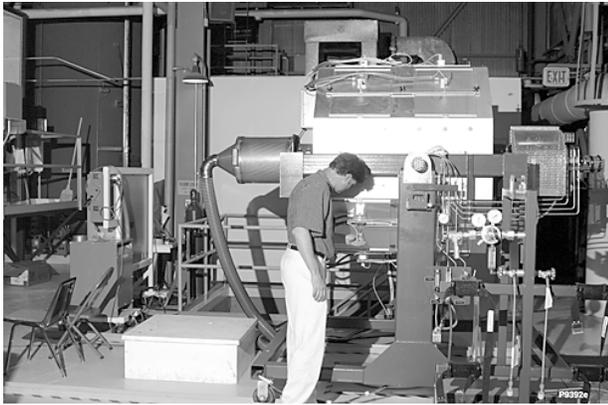


Figure 4. Production-Scale Zeolite Dryer

Two DOE milestones associated with the ceramic waste that were completed over the last two years concerned the method of consolidation. During the demonstration program, a hot isostatic press (HIP) was used to consolidate the ceramic powders into a monolithic material. This operation occurred at 850°C and under 100 MPa of pressure in the HIP. Late in the demonstration program an alternative consolidate process was developed in the Chemical Technology Division of ANL in Illinois. This method did not require increased pressures and was therefore termed pressureless consolidation. In the final report for the Committee on Electrometallurgical Techniques for DOE Spent Nuclear Fuel, they recommended pursuing this process improvement.⁴ With the establishment of the production program, DOE set a milestone in 2000 to evaluate the two consolidation options and make a recommendation on the reference. They also set another milestone in 2002 to confirm that choice based on further testing. Both of these milestones were completed on time. Pressureless consolidation was chosen as the reference production method. The main reason for this choice was to eliminate the engineering challenge of installing a production-scale HIP in an existing hot cell. The cranes in HFEF are not sized for the massive equipment associated with large pressure vessels.

During the demonstration, eight metal waste form ingots were produced from irradiated materials for 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 needed for metal waste. A prototype production metal waste form furnace, Figure 5, was built and placed into operation. 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 fabricated for installation in HFEF.



Figure 5. Prototype Metal Waste Form Furnace

Establishing the disposal path of both HLW waste forms was the source of one of the FY2001 milestones. 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.¹³ Total life-cycle costs were also requested.

This report was issued in March 2001.¹⁴ 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 presently defined, 59 DOE standardized canisters will be sent to a repository. The estimated present value of the total life-cycle cost for the program including waste disposal costs was \$423 million.

IV. 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 on-going to further increase process throughputs. Additionally, HLW form production processes are being designed, tested, and implemented.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy, Nuclear Energy Research and Development Program, under Contract W-31-109-Eng-38.

REFERENCES

1. K. M. Goff, R. W. Benedict, S. G. Johnson, R. D. Mariani, M. F. Simpson, and B. R. Westphal, "Electrometallurgical Treatment Demonstration at ANL-West," Proceedings of the ANS Embedded Topical Meeting DOE Spent Nuclear Fuel and Fissile Material Management (June 4-8, 2000).
2. *Technical Strategy for the Management of INEEL Spent Nuclear Fuel, Volume 1*, A report of the INEEL Spent Nuclear Fuel Task Team, prepared for DOE Office of Spent Fuel Management (March 1997).
3. U.S. Department of Energy, "Draft Environmental Impact Statement for the Treatment and Management of Sodium-Bonded Spent Nuclear Fuel," DOE/EIS-0306D, July 1999.
4. National Research Council, "Electrometallurgical Techniques for DOE Spent Fuel Treatment: Final Report," National Academy Press, Washington, DC (2000).
5. 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).
6. "Cost Study of Alternatives Presented in the Draft Environmental Impact Statement for the Treatment and Management of Sodium-bonded Spent Nuclear Fuel" (August 1999).
7. 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).
8. Federal Register, Vol. 65, No. 182/56565 (September 19, 2000).
9. 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.
10. 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.
11. 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).
12. 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.
13. Conference Report, Energy and Water Appropriations FY2001.
14. U. S. Department of Energy, Office of Nuclear Energy, Science and Technology, "A Report to Congress on Electrometallurgical Treatment Waste Forms," (March 2001).