Success Criteria for the Electrometallurgical Treatment Demonstration

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SUCCESS CRITERIA FOR THE ELECTROMETALLURGICAL TREATMENT DEMONSTRATION

ABSTRACT

Argonne National Laboratory is demonstrating the application of electrometallurgical treatment processes to Experimental Breeder Reactor-II spent nuclear fuel. Begun in June 1996, 100 driver fuel assemblies and 25 blanket fuel assemblies will condition be conditioned this demonstration project. In order to validate the technical and economic viability of the technology, the Department of Energy has established four success criteria with specific supporting goals. The results from both laboratory-scale and engineering-scale testing are being to evaluate the processes, products and equipment against the target goals. The interim results have provided confidence that the integrated electrometallurgical processes will prove to be a viable option for treating problematic spent nuclear fuels for geologic disposal.

BACKGROUND

The Experimental Breeder Reactor-II (EBR-II) ceased operation on September 30, 1994. When the reactor was shut down for the last time, it contained a full core loading of experimental sodium-bonded metallic fuel assemblies and an external blanket of sodium-bonded depleted-uranium assemblies. Other spent fuel and blanket assemblies from EBR-II were stored in the Hot Fuel Examination Facility (HFEF) and the Radioactive Scrap and Waste Facility (RSWF). Previously, older-type spent nuclear fuel (SNF) from EBR-II had been reprocessed at the Idaho Chemical Processing Plant (ICPP). That practice was discontinued in 1988, because the new type of zirconium-rich fuel then being demonstrated in EBR-II could not be safely processed in the ICPP using existing flowsheets and equipment. Instead, the new fuel was to be recycled into EBR-II after processing in the Fuel Conditioning Facility (FCF), which is attached to the reactor building. When the ICPP was shut down in 1992, some 2000 kilograms of the earlier-type SNF from EBR-II was left stranded in wet storage in the fuel basin at ICPP.

The shutdown of EBR-II created a spent fuel management problem. The sodium that forms the thermal bond in the EBR-II fuel is a Resource Conservation and Recovery Act (RCRA) reactive metal. Without further use in research and development, the State of Idaho threatened to regulate the SNF as a mixed hazardous/radioactive waste. With the signing of an agreement with the State of Idaho in 1995, the Department of Energy (DOE) committed to remove all SNF from Idaho by 2035. DOE's plan calls for management of its stainless-steel clad fuel at Idaho National Engineering and Environmental Laboratory (INEEL) site, rather than shipping it to another location for temporary storage or treatment. A recent INEEL Spent Nuclear Fuel Task Team has stated that the general consensus is that sodium-bonded fuel will require treatment before the geologic repository or national monitored retrievable storage facility accepts it.

The EBR-II spent fuel has a number of characteristics that place it in the “problem” category for direct disposal. The metallic sodium in the fuel has received the most attention, because of the potential regulatory issues associated with placing RCRA-regulated material in a high-level waste repository. The sodium is a highly reactive material that will react vigorously with ground water or moist air. Another problem is that at the expected temperatures inside the disposal casks, the sodium would be liquid. The Department currently expects to exclude free liquids from the repository. The residual uranium in the fuel is pyrophoric at some storage conditions, particularly after some of it has corroded and formed uranium hydride. Finally, the criticality issue associated with disposing highly enriched fuels has not been fully resolved. Because of these reasons, it is widely accepted that the EBR-II SNF will be conditioned in some manner prior to repository disposal.

The DOE decided to consider electrometallurgical technology for conditioning the EBR-II SNF for ultimate disposal and stable interim storage. Developed as part of the Integral Fast Reactor (IFR) Program, the technology, facility and equipment were available for treating the fuel. Electrometallurgical technology had been shown in the laboratory to be capable of processing uranium metal and partitioning the elements that would be highly radioactive in the spent fuel. Two waste forms were developed for stabilizing the high level waste into very compact monoliths that together require less storage volume than the unprocessed fuel. Because much of the necessary investment had already been made, electrometallurgical
technology was a promising option for treating the fuel to eliminate the RCRA and other issues.

DEMONSTRATION PROJECT

In 1995, electrometallurgical technology had been applied to irradiated fuel only on a very small scale. Since more development and testing were required before it could be considered a fully viable technique for treating the entire inventory of EBR-II spent fuel, DOE decided on a two-phase approach. The first phase would be a technology demonstration conducted on a fraction of the SNF sufficient to examine issues of processing fuels of varying burnup, production of acceptable waste forms, and the economics of feasible throughput. If no technical showstoppers were identified in the demonstration, the Department would propose to treat the balance of the inventory in the second phase. The demonstration would provide the data required to support an environmental impact statement for application of the technology the remaining EBR-II SNF.

DOE prepared and approved an environmental assessment for a demonstration of the technology at Argonne National Laboratory-West. The EBR-II Spent Nuclear Fuel Treatment Demonstration Project started in June 1996 and is expected to end in June 1999. During the testing program, 100 EBR-II driver (core) fuel assemblies (410 kgs highly enriched uranium) and 25 EBR-II blanket assemblies (1200 kgs depleted uranium) will be processed. Waste form samples will be prepared and tested. Sufficient data for evaluating the safety, environmental impact, and economics of the technology will be generated. Criteria have been established against which the success of the demonstration will be measured.

SUCCESS CRITERIA

Criterion 1: Demonstration that 125 EBR-II assemblies can be treated within three years, with a throughput rate of 16 kg/month for driver assemblies sustained for a minimum of three months and a blanket throughput rate of 150 kg for one month.

Specific goals to meet criterion 1

1. Freeze process modifications and operating parameters while demonstrating a continuous throughput of 16 kg of driver uranium for three months.
2. Demonstrate the capability to electrorefine approximately 150 kg of blanket spent fuel in one month.
3. Distill the electrolyte from electrorefiner cathode products through the cathode processor in FCF and blend the resulting ingot with depleted uranium in the casting furnace to produce a low-enriched uranium storage ingot.
4. Specify acceptable operating parameters and throughput for the cathode processor to meet uranium product specifications and electrorefiner production rates of 16 kg of driver uranium for three months.
5. Specify acceptable casting furnace operating parameters for producing low enriched uranium from 16-kg driver uranium per month for three months.
6. Cast three batches of irradiated cladding hulls (2 driver assemblies per batch) into a typical metal waste form (stainless steel - 15% zirconium).
7. Process three kilograms of salt containing approximately 6 weight % fission products into 10 ceramic waste samples.

Criterion 2: Quantification (for both composition and mass) of recycle, waste, and product streams that demonstrate projected material balance with no significant deviations.

Specific goals to meet criterion 2:

1. Develop uranium product specifications with range of acceptable impurities: plutonium, neptunium, technetium-99 and ruthenium-106. Specify process-operating parameters for uranium ingots that meet uranium specifications.
2. Develop metal waste specifications that are based on performance characterization results of small samples with variations in the principal constituents: zirconium, uranium, technetium, plutonium, neptunium, and noble metals. Determine performance characterization with electrochemical technique, corrosion tests, vapor hydration tests, and attribute tests.
3. Develop metal waste process specifications for major process variables: operating temperatures, hold time, and cooling rate.
4. Develop ceramic waste specifications that are based on performance characterization results of samples with principal constituent variations: glass, fission products, uranium, and plutonium. Determine performance characteristics with
attribute, characterization, accelerated, and service condition tests.

5. Develop ceramic waste process specifications for major process variables: free chloride, zeolite moisture content, and chloride per unit cell.

6. Quantify volume of low-level and transuranic waste generation under standard operating conditions.

7. Return the cathode processor condensate to the individual electrorefiners during the 16-kg driver/month for three months and 150 kg blanket/month operations.

8. Specify unit process operations for metal spent fuel treatment, uranium ingot production, and waste form production.

9. Estimate mass balances for uranium, transuranics, sodium, and key fission products for overall process.

10. Prepare the flowsheet and develop process specifications for the subsequent inventory operation.

**Criterion 3: Demonstration of an overall dependable and predictable process, considering uptime, repair and maintenance, and operability of linked process steps.**

**Specific goals to meet criterion 3:**

1. Record facility and equipment availability for process operations during the three month 16 kg per month driver demonstration.

2. Record process interruption for chemistry results during the three-month operation at 16 kg per month.

3. Develop quantitative process models for each key step in the treatment process.

4. Develop a process model that estimates throughputs as a function of equipment availability, maintenance requirements, and individual process times.

**Criterion 4: Demonstration that safety risks, environmental impacts, and nuclear materials accountancy are quantified and acceptable within regulatory limits.**

**Specific goals to meet criterion 4:**

1. Demonstrate that the FCF air emissions result in an effective dose equivalent to the public less than 10 mrem per year, which is the limit in DOE 5400.5 and is less than the 25 mrem per year limit in the State of Idaho Permit to Construct Air Pollution Emitting Source.

2. Show that FCF personnel exposure is less than 0.5 rem per year average and 1.5 rem per year for the maximum individual exposure, which is a factor of three less than the DOE Occupational Radiation Protection Final Rule 10CFR835 limit that is 5 rem per year.

3. Demonstrate a material control and accountability system that shows the historical inventory difference for uranium and plutonium are within control limits based on variance propagation of measurement and sampling errors, as specified in DOE Order 5633.3B.

4. Record any unlikely and extremely unlikely accident (as defined in FSAR) during the Demonstration.

5. Estimate the safety risks, environmental impacts, and material accountancy for the inventory operations.

**EVALUATION PROCESS**

The DOE has several sources of information by which to judge the success of the demonstration program. The principal source is the set of detailed technical reports and summary status reports that Argonne publishes. The project provides DOE with a monthly status report that summarizes progress in each of the key areas of the demonstration and related electrometallurgical technology development. That report is followed by a detailed technical report that describes all the monthly activities and results from each work breakdown structure elements. Topical reports are also issued to fulfill milestones or to provide a more comprehensive status of specific aspects of the project. However, the key reports for DOE will be two status reports in which Argonne will assess the technical progress of the demonstration project against the success criteria. The first of these is a mid-point report, in which Argonne will assess whether the project is on track for a successful demonstration. The experience gained by this time should be adequate to establish the scientific feasibility of processing the irradiated fuel. The final assessment report will be issued at the end of the demonstration.
In making its evaluation of the success of the project, DOE is supported by two independent review committees comprised of experts in the foundations of electrometallurgical technology. The first is the National Research Council’s (NRC) Committee on Electrometallurgical Techniques for DOE Spent Fuel Treatment, which the NRC formed at the Department’s request. The second is the University of Chicago’s Special Advisory Committee for the [Argonne National Laboratory] Nuclear Technology Program. Neither of the two committees has the responsibility for making the decision on whether the demonstration project has been success, but rather they each provide DOE with an independent assessment of the status of the technology. During future meetings of these two peer review committees, the members will be asked to focus on the progress achieved by the demonstration project in meeting the established success criteria. These evaluations of the technology’s maturity will be provided to DOE through the committees’ reports. Committee meetings and ensuing reports will be scheduled to support the Department’s decision on the success of the demonstration project.

DEMONSTRATION INTERIM RESULTS

Fifty-two driver assemblies have been treated including six months operations at a processing rate of four assemblies per month. The driver treatment will continue through February 1999 when 100 drivers will be treated. The necessary equipment for treating the larger-size blanket assemblies is installed in the hot cell and depleted uranium testing of this high throughput equipment has started. Blanket operations are expected to start in May 1998 and the 25 assemblies should be processed by June 1999. One of the three demonstration-size metal waste samples has been cast from cladding hulls that were removed from the electrorefiner. Laboratory-scale samples have been cast with variations in metallic fission products, actinide spikes and technetium spikes. These metal waste samples are being characterized in support of waste form characterization and specification development. The ceramic waste equipment that will be used to produce sample waste forms from the electrorefiner salts is being tested with non-radioactive fission product elements and process variables are being tested. Laboratory-scale ceramic waste samples are being produced at various uranium, plutonium, non-radioactive fission product and primary constituent compositions. The ceramic waste specifications will be developed from the characteristics of these samples and results with the demonstration-scale equipment. These demonstration results indicate that the specific goals that support the success criteria should be met by June 1999.

CONCLUSIONS

The EBR-II Spent Fuel Treatment Demonstration has been treating irradiated fuel for twenty months. Four success criteria and specific goals have been established for judging the success of the technical and economic viability of the electrometallurgical treatment process. Approximately 50 percent of the driver fuel has been processed and the treatment equipment for the blanket fuel has been installed in the hot cell. Samples of the two waste forms have been produced and the demonstration scale equipment is currently being tested. The interim results indicate that electrometallurgical treatment process is a viable method to prepare problem fuels for a geologic repository.

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REFERENCES


