SPENT FUEL PYROPROCESSING DEMONSTRATION

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A major element of the shutdown of the U. S. liquid metal reactor development program is managing the sodium-bonded spent metallic fuel from the Experimental Breeder Reactor-II to meet U. S. environmental laws. Argonne National Laboratory has refurbished and equipped an existing hot cell facility for treating the spent fuel by a high-temperature electrochemical process commonly called pyroprocessing. Four products will be produced for storage and disposal. Two high-level waste forms will be produced and qualified for disposal of the fission and activation products. Uranium and transuranium alloys will be produced for storage pending a decision by the U. S. Department of Energy on the fate of its plutonium and enriched uranium. Together these activities will demonstrate a unique electrochemical treatment technology for spent nuclear fuel. This technology potentially has significant economic and technical advantages over either conventional reprocessing or direct disposal as a high-level waste option.

Introduction

Given the recent drastic changes in the U. S. advanced reactor development program, the demonstration program described in this paper is a remarkably similar, but truncated version of the program plan of a year ago. Argonne National Laboratory (ANL) is on the brink of demonstrating an innovative, unique, electrochemical technology for treating metallic spent nuclear fuel. The basic technology was developed for the Integral Fast Reactor (IFR) program¹, which, until it was recently canceled, was the U. S. nuclear research and development program for advanced liquid metal reactors (LMRs). The roots of the treatment technology lie in a primitive melt-refining metal fuel cycle demonstrated from 1965 to 1969 as part of initial operation of the Experimental Breeder Reactor-II (EBR-II).² Originally developed as a closed fuel cycle, appropriate elements of the current technology are being carried forward to treat the accumulated spent EBR-II fuel to meet existing U. S. environmental law and to process the high-level waste residue into a form suitable for storage and subsequent geologic disposal.

The metallic EBR-II fuel contains a sodium thermal bond between the fuel pin and the cladding. During irradiation, much of the sodium logs in fission gas pores within the fuel. Sodium is a reactive metal that by law cannot be buried for disposal. Further, an end-of-life enrichment exceeding 50% and a potential for long-term pyrophoric hydride formation following cladding breach eliminate the EBR-II fuel as a candidate for direct geologic disposal. By the time the last of the EBR-II spent fuel is treated, a throughput rate and unit cost should be established that will allow economic evaluation of this technology as a waste disposal option.

The metallic fuel cycle developed for the IFR, shown schematically in Fig. 1, is very compact. With exception of waste equipment that was to be added within two years, the entire recycling process required just nine machines. The key elements of the recycle technology are electrorefining to extract the uranium, plutonium and other actinides from the dissolved fuel, cathode processing to produce metal ingots from the electrorefiner products, and injection casting to fabricate new fuel pins. Although EBR-II is only a 62.5 MWt power plant, these three major pieces of equipment were to process batch sizes typical of commercial LMR operation. With termination of the LMR development program, the fuel fabrication mission has been eliminated, and added emphasis has been placed on waste form development. Most of the fission products will be immobilized in a zeolite-based mineral waste form, while converted fuel fabrication equipment will alloy the stainless steel structural material with noble metal fission products and cast the alloy into a homogeneous ingot.

The U. S. Department of Energy (DOE) has over 100 types of spent fuel to be disposed, some 2700 tonnes in all. EBR-II driver and blanket fuel assemblies comprise only 20 tonnes of the total, but the majority of the remainder, 2100 tonnes of N Reactor spent fuel, is also metallic. N Reactor fuel is therefore directly treatable by the same electrochemical technology used to reduce EBR-II fuel to a stable waste form.³ The N Reactor fuel, designed to be reprocessed within 60 days after discharge, is now suffering to varying degrees from significant corrosion after years of wet storage. Processing the fuel to stabilize it may be necessary to prevent significant pyrophoric hydride formation when the fuel is placed in dry storage.

With headend modifications, many of the remaining DOE fuel types can also be treated in this way, always producing just the two waste forms. The advantage of selecting a minimum number of treatment technologies is the huge cost savings that can be gained over having to qualify many different fuel types or waste forms for geologic disposal. Because so much of the DOE spent fuel is reactive, corroded, or highly enriched, some type of treatment will be required before the fuels can be responsibly disposed.
Fig. 1. The IFR Fuel Cycle Demonstration Modified for EBR-II Spent Fuel Disposition

Fig. 2. EBR-II and the Fuel Cycle Facility
Facility Preparation

The IFR fuel cycle demonstration required only a small hot cell facility. Such a facility was available — the Fuel Cycle Facility (FCF), which was used for the original melt-refining process demonstrated during the first five years of EBR-II operation. However, to meet modern safety standards for facilities processing nuclear fuels, especially plutonium-bearing fuels, many improvements to the facility were mandated. These modifications principally improve confinement of radioactive particles, including any plutonium aerosols that might be generated during a severe accident.

FCF was originally constructed in the early 1960s along with EBR-II and was used from 1964 through 1969 to demonstrate an immature version of pyrometallurgical fuel processing. In subsequent years, the facility was used for examination of irradiated fuels and materials until a new hot cell facility (HFEF) began operation in 1975. FCF was partially decontaminated several years prior to commencing construction for modifications in 1987.

FCF, which is directly connected to EBR-II as shown in Fig. 2, has two separate shielded hot cells. Operations involving intact fuel elements will be carried out in the air-atmosphere cell. These processes involving exposure of heavy metals or reactive metals will be done in the argon-atmosphere cell. The air cell is a 66 m² concrete cell with eight windows, sixteen master/slave mechanical manipulators, two electromechanical manipulators, and a crane. The argon cell has 220 m² of floor space, a welded steel liner, 19 windows, 26 master/slave mechanical manipulators, four electromechanical manipulators, and two cranes. Both hot cells have remote repair/recuperation capability for all active components of the handling systems.

Extensive modifications to FCF were needed in order to carry out its programmatic mission and to fulfill regulatory requirements. Most of the facility modifications have been done as a result of changes in DOE safety requirements since FCF was originally constructed in the early 1960s. These safety modifications have included numerous confinement improvements as well as installation of a safety-class exhaust system and a 360 kW emergency diesel system. DOE review and approval of the safety documentation, including the Final Safety Analysis Report, the Technical Safety Requirements, and the Criticality Safety Hazards Report were obtained during 1993. All necessary construction will be completed in 1994. Full operation is expected by the beginning of 1995 if DOE can complete its formal review on schedule.

EBR-II Spent Fuel Inventory

The EBR-II spent fuel inventory to be processed for the IFR termination consists of approximately one tonne of driver assemblies and nineteen tonnes of blanket assemblies. The fuel is currently stored in the reactor vessel, a separate hot cell facility (HFEF), and in an underground dry storage facility (RSWF). There are several more tonnes of similar spent EBR-II fuel stored at another location at the Idaho National Engineering Laboratory (INEL) that is not currently scheduled for the demonstration. However, the programmatic environmental impact statement for DOE spent fuel management leaves open the possibility of pyroprocessing that fuel also, as well as other fuel types.

The driver assemblies contain 61 fuel elements that are still highly enriched, between 60% and 75% at end of life. Most driver assemblies have an average burnup of eight atom percent, but some have reached 20%. Some experimental assemblies contain about 20% plutonium, but in all there are only about seven kilograms in driver and experimental assemblies. The blanket assemblies have reached burnups in the 1-2% range and contain an estimated total of 200 kilograms of plutonium. Each blanket assembly is comprised of 19 fuel elements and contains approximately 10 times as much heavy metal as a driver assembly.

Spent fuel from EBR-II is washed in the FCF cask handling area to remove residual external sodium prior to storage. The fuel is then transferred to HFEF where it is packaged for storage in RSWF. Short-cooled fuel is stored in HFEF until it decays to below the maximum thermal limit for RSWF. Some structural stainless steel is removed from blanket assemblies to accommodate size restrictions in the RSWF liners. Transport between HFEF and RSWF is accomplished with a shielded cask and forklift system. Weather and ground conditions prevent transport during some months of the year. RSWF fuel destined for treatment in FCF may have to pass back through HFEF. Driver fuel from the final unloading of EBR-II in 1997 may be taken directly into the FCF cells for treatment.

Driver Fuel Treatment

The EBR-II driver fuel treatment campaign will begin in 1995 and take approximately three years to complete. There are 240 driver assemblies to be processed; FCF has an environmental air permit to process a maximum of 90 assemblies per year. Equipment will be tested during startup by processing unirradiated depleted uranium fuel elements.

Driver assemblies will be transferred by cask from HFEF to FCF, where they will be inserted into the air cell. The hex cans will be cut away and the 61 fuel elements will be separated by a single machine called the vertical assembler/disassembler (VAD). The VAD, a 30 year old piece of equipment that was to be used in the IFR fuel cycle demonstration, is also capable of assembling 61 fresh fuel elements into a new assembly. The stainless steel hardware will be stored until it receives further processing to make one of the waste products.

The individual fuel elements, 750 mm long by 5.8 mm diameter, will be transferred into the FCF argon cell. Here they will be sliced into six millimeter segments and collected in fuel dissolution baskets in 20 kg batches. Each element contains a 339 mm, 82 gram fuel slug. These cruciform-shaped stainless baskets become the anodes in the next piece of process equipment, the electrorefiner. This first significant test of chopping sodium-bonded metal alloy will answer technical questions about the effect of sodium on the shearing operation and the lifetime of the dies.

The heart of the fuel treatment is a 500°C electrochemical process employing molten salts and liquid metals in an electrorefining operation. The molten salt is a LiCl-KCl eutectic containing actinide chlorides and
fission product chlorides. One meter in diameter, the FCF electrorefiner contains 427 kilograms of LiCl-KCl salt with a depth of 333 mm, as shown in Fig. 3. Below the salt phase, the electrorefiner also contains a 102-mm deep, 533-kg pool of molten cadmium that can serve as an anode, cathode, or a collector of noble metals.

Uranium and transuranics (TRU) will be separated from the majority of the fission products in separate electrorefiner operations. The fuel segments in their baskets will be loaded in the electrorefiner anodes and directly electrotransported for collection of relatively pure uranium on solid mandrel cathodes. The active fission products, sodium, and any residual heavy metals will form chlorides in the salt eutectic. Noble metal fission products are expected to be left behind along with the cladding hulls in fine mesh screens within the anode baskets. The metal screens and their contents will become one of the two high-level waste streams.

![Fig. 3. Schematic of the FCF Electrorefiner](image)

The transuranics, principally plutonium, can be collected after their concentration in the salt builds up sufficiently. They will be deposited in a cathode crucible containing 26 kilograms of liquid cadmium. Uranium and some rare earth fission products will be co-deposited with the TRU. The heavy metal in the cathode product will be no more than about 75% plutonium. Each liquid cathode could contain up to four kilograms of plutonium. The technical uncertainty in this operation is that plutonium collection at this scale or in the presence of a high concentration of fission products has never before been attempted in an electrorefiner of this type. One-hundred gram quantities have been collected in liquid cathodes in clean salt environments with simulated fission products. Additional cold plutonium may be added to the electrorefiner after startup so the performance of the liquid cadmium cathode can be tested early in the demonstration, allowing time for possible modifications.

The cathode products will be removed from the electrorefiner, placed in a crucible, and inserted into the cathode processor. This piece of equipment will evaporate the adhering salt or the cadmium and consolidate the metal product into a segmented ingot that will later be broken into wedge-shaped pieces. This particular retort application has never been done at such large scale for actinide products. The FCF cathode processor, adequate for the demonstration, is not expected to bear much resemblance to an eventual production machine. The goals of the demonstration are to minimize product loss during the retort and to recycle the condensed cadmium and salt back into the electrorefiner.

![Fig. 4. Schematic of the FCF Cathode Processor](image)

The driver uranium product will be blended with depleted uranium down to less than 20% enrichment. One option for doing this is in the cathode processor; another is to take it to a higher temperature furnace, which also would ensure better homogeneity of the product ingot. The uranium may be transferred to one of the site's safeguarded vaults for long-term storage, depending on its radioactivity level.

The TRU product will be taken to the FCF casting furnace to cast a single pin for sample analysis. The product will be highly radioactive due to the minor actinides and the rare earth fission products present. A decision will be made on whether to alloy the product with other metals such as depleted uranium and zirconium prior to storage. The product ingot will require heavy shielding if it is to be stored in one of the plutonium vaults on site. Storage locations are also available within both hot cell facilities. There is also an option for storing very highly radioactive products in the underground storage facility, RSWF.

Waste Operations

After approximately a year of operation, the fission product heating in the salt and cadmium phases will
approach the six kilowatt thermal limit for passive dissipation of heat by the electrorefiner. Before this limit is reached, the fission product concentration must be reduced before more fuel processing takes place. Alkali, alkaline earth, rare earth, and halide fission products will accumulate primarily in the salt phase. Less chemically active metal fission products will be in the cadmium phase primarily as insolubles. The bond sodium will be neutralized in the salt as sodium chloride.

Waste processing equipment is currently being designed, prototype-tested, and fabricated, but no waste equipment is operating or installed in FCF. The first two waste operations for the electrorefiner will therefore involve "drawing down" the actinide concentration in the salt to a very low level, followed by removal of most of the salt. This bulk salt will be stored in cans in the FCF cell until it can be processed to separate the remaining actinides and fission products.

Most of the mineral waste form samples made from EBR-II driver fuel processing will be required for testing leading to eventual qualification for geologic disposal.

The FCF casting furnace that was to fabricate the metal fuel for future EBR-II operation will be converted to produce a metal alloy ingot that will be the metal waste form. Cladding hulls and mesh screens from the anode baskets, filters from the bulk fluid handling operation, and possibly cut up fuel assembly hardware will go into the alloy. The decision on the assembly hardware will depend on whether it can be disposed as low level waste, or whether it is more cost-effective to make it part of the high-level waste form.

The waste samples prepared from these operations will be subjected to extensive testing: product consistency test (PCT), materials characterization center (MCC-1), toxicity characteristic leachate procedure (TCLP), and corrosion tests. The MCC-1 and the PCT tests are standard tests for qualifying vitrified high-level waste. The TCLP test is required to demonstrate that the waste will not be classified as chemically hazardous.

Blanket Fuel Processing

Much general process knowledge will be gained in the three years of driver fuel treatment that will precede the blanket processing. However, several differences require that certain changes be made in the process and the equipment before the blanket processing begins. First, the heavy metal mass to be processed is approximately 20 times greater, and treatment must be completed in less than two years to meet the established DOE schedule for termination. Second, the blanket fuel assemblies have very light burnup, only on the order of one to two percent. Third, each assembly contains 19 rather than 61 elements, but each element is thicker, denser, and longer.

The key to meeting the process schedule is developing a high-throughput electrorefiner. The goal of a high-throughput electrorefiner for FCF is to be able to process 1000 kg of heavy metal per month, rather than the currently licensed 400 kg per year, and to demonstrate the capability to process 1000 kilograms per day. Several designs are being considered, but all have the capability of electrotransporting from 200 to 400 kilograms per batch. Transport operations typically take less than 24 hours. The conceptual designs fit within the vessel of the current FCF electrorefiner. To meet the schedule, it is expected that an existing spare vessel will be fitted with a new top and internals. One major change in the electrorefiner design will be to eliminate the cadmium pool, which is an ultimate disposal concern because cadmium is a listed hazardous waste. To cut down on the materials handling time between electrotransport operations, current plans call for a commercial gantry robot to be incorporated into the high-throughput electrorefiner design.

The larger blanket fuel, 1.5 m long with a 12.5 mm diameter, contains 2.5 kg of heavy metal. Chopping this fuel to the desired segment length of 12 mm will require extensive modifications to the present FCF element chopper, or more likely, a new machine. Technologies under consideration include a commercial rod shear, a punch press, a sheet nibbler, and a scrap shredder.

Production of the mineral waste for at the volume needed for blanket processing will require scale-up of the
HIP pressure vessel used for sample testing. A commercial HIP machine with a maximum working zone of 254 mm (diameter) by 762 mm and a capacity of 225 kg is under consideration. The capacity of the sample preparation HIP is 35 kg.

A new furnace for metal waste form production will be required. The new inductively heated furnace will resemble the current CFC casting furnace, but will have several significant changes. It will use a larger crucible and have tilt-pour or bottom-pour capability to allow chill casting of metal waste ingots. Replacing the graphite susceptors with direct coupling to the metal waste charge material will be investigated in the conceptual design phase.

Basic technology development to support processes and equipment for the advanced treatment needed for the EBR-II spent blanket fuel is being carried out at the main Argonne site near Chicago. Electrorefining development will continue using simulated fission products in electorefiners operating under controlled conditions. These activities will support development of advanced anode/cathode pairs and product collection systems. A multistage pyrocontactor is being developed that will reduce the amount of TRU in the waste streams to very low levels, and will speed up removal of fission products from the electorefiner. Development and testing of candidate waste form compositions will continue. Tests on a tilt-pour casting furnace for metal waste form development are scheduled in the near future. A hot isostatic press is being installed to continue development of converting the fission-product-loaded zeolite to a stable mineral waste form.

In EBR-II, the blanket fuel has not been used as classic breeder blankets, but rather as additional shielding outside the radial stainless steel reflector, i.e., well separated from the reactor core. Consequently, there is considerable uncertainty in the calculated burnup and plutonium buildup in the blanket assemblies. It is assumed that this uncertainty will be unacceptably large for material control and accountability purposes, and must be supplemented by measurements to determine the fissile mass input to the process.

An active neutron assay system has been under development for nondestructive analysis (NDA) of waste cans that will shipped from FCF. This development is being expanded to include assay of the blanket fuel elements, or segments of elements. The NDA system will be installed in the basement of FCF, accessible through the argon cell floor, when development and testing in HPEF is completed.

Summary

Argonne National Laboratory will treat approximately one percent of the DOE-owned spent fuel inventory to produce stable waste forms for geologic disposal. Uranium ingots and plutonium alloy ingots will be produced as byproducts for storage pending a national decision on DOE-owned highly enriched uranium and plutonium. Although driven primarily by a legal requirement to neutralize the sodium bond in the unique EBR-II metallic alloy fuel, pyroprocessing will also obviate other potential disposal problems such as high enrichment and pyrophoricity.

The spent fuel treatment demonstration will begin early in 1995 and run for at least four years. Significant waste process operations are at least two years away. New, more advanced equipment will be added as it becomes available during the demonstration.

If successful, Argonne's pyroprocessing demonstration for spent fuel management could mark one of the few major breakthroughs in nuclear fuel cycle technology in the past three decades. Processing EBR-II spent fuel with second generation equipment will allow evaluation of extrapolating this technology to other fuel types, including over 80% of the spent DOE fuel and even to commercial light water reactor (LWR) fuel. Developed as a proliferation-resistant fast reactor fuel recycle technology, pyroprocessing of the type employed at Argonne has no potential application for MOX recycle to LWRs or for military purposes. Although it may be possible under carefully controlled conditions to electrotransport relatively pure uranium, it is impossible to achieve a plutonium product that is not heavily diluted with uranium, all minor actinides present, and a small fraction of fission products. As a waste disposal option, this is not a problem, because the relatively small quantity of recovered plutonium can be alloyed with the metal waste form to produce suitable candidate material for geologic disposal.

The goal of demonstrating 1000 kg/day throughput potential with relatively small-size equipment in a small facility seems feasible at this stage of development. If the actual demonstrated throughput potential is anywhere in the ballpark, the economic ramifications of broader application of this technology will assure its consideration as a viable fuel cycle option.

Acknowledgment


References


