EBR-II SPENT FUEL TREATMENT DEMONSTRATION PROJECT STATUS

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Summary
The application of electrometallurgical technology to spent nuclear fuel treatment is being demonstrated by treating 410 kg uranium spent driver fuel and 1200 kg uranium spent blanket fuel from the Experimental Breeder Reactor-II (EBR-II) spent driver and blanket fuel. This fuel is a metallic uranium alloy and contains elemental sodium, which is a reactive material. Since reactive material is considered hazardous by United States Environmental Protection Agency regulations, this fuel requires treatment before disposal in a geologic repository. The EBR-II spent fuel treatment demonstration conditions this fuel in an integrated process where the fuel is converted into three different products: low enriched uranium (LEU), ceramic waste and metallic waste. This demonstration was initiated in June 1996 and has treated approximately 50 percent of the driver fuel. The higher throughput equipment that will be used for blanket treatment processes has been installed in the hot cell facility and is being tested with depleted uranium. Metal waste forms have been produced from the irradiated metals from the driver fuel. Ceramic waste process equipment has been built and is currently being tested before installation in the hot cell facilities. This paper discusses the processes and the current results from the first 20 months of operation.

Introduction
For approximately ten years, Argonne National Laboratory developed a fast reactor fuel cycle based on nonaqueous processing (e.g., Hannum 1997). When the United States fast reactor program was canceled in 1994, the fuel processing technology, called the electrometallurgical technique, was adapted for treating unstable spent nuclear fuel for disposal. While this technique, which involves electrorefining fuel in a molten salt bath, is being developed for several fuel categories, its initial application is for sodium-bonded metallic fuel. This fuel is considered not suitable for repository disposal and must be treated (e.g., DOE 1997). The Department of Energy (DOE) owns approximately 60 metric tonnes of sodium-bonded fuel; EBR-II fuel comprises approximately forty percent of the inventory. Since the electrometallurgical treatment process is a possible treatment technology, DOE requested the National Research Council (NRC) to provide a technical evaluation of this approach for treating DOE spent nuclear fuel. In their initial report (e.g., National Research Council 1995), the NRC committee recommended that a demonstration of the electrometallurgical technology using sodium-bonded metallic spent fuel should meet some specific success criteria before a decision was made to implement this technology for other fuels.
In June 1996, DOE approved a hot demonstration program in which 100 spent driver assemblies and 25 spent blanket assemblies from EBR-II would be treated over a three-year period. The 100 driver assemblies (410 kg uranium) will be processed in one electrorefiner so the fission product chloride concentration in the salt will reach approximately 6 weight percent. The 25 spent blanket assemblies (1200 kg uranium) will be treated in another electrorefiner to demonstrate higher throughput rates of 150 kilograms uranium per month. In addition, the demonstration program will show the technical feasibility of the two high level waste forms that are produced. The waste form activities include the production of waste samples from the fuel processing and laboratory scale samples that encompass the full range of compositions that are expected if the process is implemented. The waste form performance will be determined by a wide variety of physical property measurements and water leaching tests. The demonstration should be completed by June 1999.

Process Operations
The fuel treatment processes are operating in the Fuel Conditioning Facility (FCF), a shielded hot cell facility at the Argonne site in Idaho. The treatment process steps, which include fuel assembly dismantling, element chopping, electrorefining, cathode processing and casting, are shown in Figure 1. Fuel assembly dismantling removes the individual stainless steel clad elements from the assembly hardware. These elements are sheared in the element chopper into fuel segments 0.5 to 1 cm in length. The driver elements (0.58 cm diameter and 75 cm length) are sheared with a small chopper while a larger blanket element chopper processes the larger blanket elements (1.25 cm diameter and 159 cm length).

The driver fuel segments are placed in anode baskets that are placed in the Mk-IV electrorefiner (Figure 2). This electrorefiner (e.g., Mariani and Vaden 1997, Goff et al. 1996, and Ackerman 1991) has a 1 m diameter vessel with a 10 cm layer of molten cadmium and a 30 cm layer of molten salt held at 450-500°C. The electrorefiner has four ports for two anodes and two cathodes that can process two 10 kg loads of uranium, simultaneously. For blanket fuel treatment, which will demonstrate higher throughput, a second (Mk-V) electrorefiner was installed in FCF in late 1997. It has an identical vessel with four ports; however, each port has a concentric anode-cathode module (ACM) (Figure 3) that holds 35 kg loads of uranium. The four ACMs (a total of 140 kg uranium) can be processed simultaneously.

After electrorefining, the cathode products are placed in a graphite crucible and transferred to the cathode processor, an induction-heated vacuum distillation furnace (e.g., Westphal et al. to be published). The residual salt and cadmium are removed from the uranium and recycled to the electrorefiner. The consolidated uranium product is placed in a different graphite crucible and transferred to the casting furnace, which is also an induction-heated device. The uranium product is mixed with depleted uranium to produce a homogeneous LEU product (< 20 percent $^{235}$U) and is sampled by injection casting.
Driver and Blanket Fuel are Treated in Parallel

Figure 1: Process Flow for Driver and Blanket Treatment

Figure 2: Mk-IV Electrorefiner Schematic
After the uranium is removed from the fuel cladding, the remaining stainless steel cladding and the undissolved fuel alloy (typically zirconium for the driver) is incorporated into a metal waste form (e.g., Abraham et al. 1996 and Westphal et al. 1994). This waste form is nominally a 15 percent zirconium – 85 percent, by weight, stainless steel and immobilizes the typical activation products and metallic fission products such as ruthenium, niobium and technetium. For the demonstration, the cladding hulls are processed through the cathode processor where the adhering electrorefiner salts are removed and recycled to the electrorefiners. The salt-free cladding hulls are mixed with zirconium and cast in the same furnace that is used for the LEU product. This casting furnace can produce up to a 6 kg metal waste ingot. In addition to the metal waste samples from the spent fuel, laboratory scale ingots (10 to 1000 g sizes) are being cast in tilt pour furnaces and muffle furnaces.

The majority of the fission products and transuranium elements accumulate in the electrorefiner salt and are incorporated into a stable ceramic waste form (e.g., Pereira et al., 1996) that is suitable for a geologic waste repository. The process steps for the demonstration are shown in Figure 4. After a portion of the electrorefiner salt is removed and solidified, the salt is ground to a powder. This salt powder is mixed with zeolite in a V-mixer that can operate up to 550°C. After the salt is absorbed into the zeolite, glass materials are mixed with the salt loaded zeolite. This powder mixture will be loaded into hot isostatic press (HIP) cans that are evacuated and seal welded. The cans are transferred to the HIP that converts the material to a ceramic monolith. The demonstration scale equipment will be installed in the Hot Fuel Examination Facility, a second hot cell facility at the Argonne site in Idaho. This equipment will produce radioactive ceramic waste samples from the electrorefiner salt used to process the driver fuels. In addition, a laboratory scale HIP and two hot uniaxial presses are producing small ceramic waste samples to support waste characterization activities.

**Process Results**

Fifty-two driver assemblies have been processed and 390 kilograms of LEU product has been produced. The early operating experience has shown that the Mk-IV electrorefiner can process four driver assemblies within five days. Complete uranium dissolution can be achieved by operating at voltages between 0.8 and 1.2 volts. If zirconium retention in the cladding hulls is desired, the electrorefiner voltage needs to be limited to prevent zirconium oxidation. The demonstration is providing the data that will allow the specification of process operating conditions to meet a wide range of applications.

The cathode processor has shown that 17 kg of highly enriched uranium (HEU) cathodes can be processed with a 36 hour processing time. A minimum operating temperature of 1150°C with an operating pressure of one torr completely separates the salts from the uranium metal. Recent tests have shown that the processing time can be shortened to 24 hours by backfilling the cathode processor with argon during the cooldown portion of the process cycle.

The casting furnace has successfully converted 167 kg of cathode products into LEU product. This equipment design was based on similar equipment that has been operating since the 1970s
Figure 3: Configuration of Concentric Anode Cathode Design

Figure 4: Process Steps for Demonstration Ceramic Waste Operations
and has provided reliable operations. The only problems have occurred when cathode processor product was produced at a vacuum around 10 torr rather than one torr. These products contain trace quantities of salt that attacks the yttria coating on the graphite casting crucibles. This salt-yttria coating interaction allows the uranium product to react with the graphite and the crucibles cannot be reused in future operations.

The blanket equipment has been installed in the FCF hot cell. The blanket chopper has sheared unirradiated blanket elements that did not contain the sodium-bonded fuel. Chopping of sodium bonded blanket elements will commence when the Mk-V electrorefiner is ready to receive blanket fuel. The Mk-V electrorefiner has been loaded with the eutectic salt and uranium chloride that is required for operations. The concentric ACMs have been tested for mechanical operations and initial process testing with depleted uranium plates has started. The necessary modifications of the cathode processor and casting furnace for larger cathode products has been completed and is waiting for testing with Mk-V electrorefiner product.

A metal waste form testing matrix has been established to generate the data that will provide the waste form attributes, leaching characteristics, accelerated aging transformations and performance at different repository service conditions. Twenty-four non-radioactive fission product samples have been produced at a range of compositions. Fifty-three spiked samples have been produced with different concentrations of technetium, uranium, plutonium and neptunium. One of three planned three-kilogram metal waste ingots has been produced from irradiated cladding fuels. These samples are undergoing testing to show waste form technical viability.

A ceramic waste form testing matrix and process qualification plan has been established for the ceramic waste form. The ceramic testing matrix will produce the data as specified for the metal waste form; however, the test methods will be developed for a ceramic rather than a metal. The process qualification plan details the important process variables in each step of the process and suggests which parameters need to be controlled to produce a consistent waste form. Laboratory scale equipment will produce 20 different samples with plutonium and uranium plus 64 samples with different composition ranges. Laboratory scale samples containing $^{238}$Pu will also be produced to study the effect of alpha damage on the ceramic. The demonstration scale equipment will produce 15 non-radioactive samples and 5 radioactive samples. The demonstration scale equipment is currently being tested for establishing process parameters and initial scoping tests with non-radioactive samples.

Conclusions
The initial demonstration results indicate technical and economic feasibility of the electrometallurgical process for sodium bonded fuel. The driver fuel is approximately 50 percent complete and will continue to be treated at the rate of four assemblies per month. The 100 driver treatment operations should be completed in February 1999. The treatment of the 25 blankets should commence in May 1998 with completion in June 1999. The metal and ceramic waste forms are being produced and the initial qualification results indicate that these wastes should be suitable candidates for a geologic disposal site.
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