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IFR FUEL CYCLE - PYROPROCESS DEVELOPMENT*

by

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The Integral Fast Reactor (IFR) fuel cycle is based on the use of a metallic fuel alloy, with nominal composition U-20Pu-10Zr. In its present state of development, this fuel system offers excellent high-burnup capabilities. Test fuel has been carried to burnups in excess of 20 atom % in EBR-II irradiations, and to peak burnups over 15 atom % in FFTF. The metallic fuel possesses physical characteristics, in particular very high thermal conductivity, that facilitate a high degree of passive inherent safety in the IFR design. The fuel has been shown to provide very large margins to failure in overpower transient events. Rapid overpower transient tests carried out in the TREAT reactor have shown the capability to withstand up to 400% overpower conditions before failing. An operational transient test conducted in EBR-II at a power ramp rate of 0.1% per second reached its termination point of 130% of normal power without any fuel failures. The IFR metallic fuel also exhibits superior compatibility with the liquid sodium coolant; intentionally-defected fuel pins have been operated in EBR-II for over 230 days beyond the point of cladding breach without the release of fuel material or solid fission products into the primary coolant.

Equally as important as the performance advantages offered by the use of metallic fuel is the fact that this fuel system permits the use of an innovative reprocessing method, known as "pyroprocessing," featuring fused-salt electrorefining of the spent fuel. Development of the IFR pyroprocess has been underway at the Argonne National Laboratory for over five years, and great progress has been made toward establishing a commercially-viable process. Pyroprocessing offers a simple, compact means for closure of the fuel cycle, with anticipated significant savings in fuel cycle costs.

Pyroprocessing of IFR spent fuel begins with the dismantling of irradiated fuel assemblies and chopping of the fuel pins into short segments. The fuel pin segments, including the cladding, are placed in a perforated metal basket and inserted into the IFR electrorefining cell. The electrorefining cell is a low-alloy steel vessel, on the order of 1-m diameter and 1-m high in its present configuration, that contains an electrolyte salt (eutectic LiCl-KCl mixture) above a pool of liquid cadmium. The cell is operated in a temperature range of 700-775K. The basket containing the chopped fuel pin segments is made the anode, and the fuel material is anodically dissolved. Because of the favorable thermodynamic properties of the relevant chlorides, the actinide elements (U, Pu, Np, Am, Cm) distribute between the salt and the cadmium pool. The alkali metals and alkaline earth elements form stable chlorides and reside only in the salt phase. The rare earth fission products reside predominantly in the salt phase. The more noble metals do not form chlorides under the conditions that exist in the electrorefiner and hence remain as metallic particulates. Uranium is electrotransported to a solid steel cathode, forming a dendritic deposit containing about 85-90 wt.% uranium and the balance salt, with minor amounts of fuel alloy zirconium and cadmium. Typical batch sizes are 10 kg heavy metal per electrode. The relative free energies of formation of the chlorides of uranium and the transuranic (TRU) elements and their relative concentrations in the salt preclude deposition of plutonium and the minor actinides on the solid cathode. A liquid cadmium cathode located in the salt phase is utilized for recovering the TRU elements. The deposition of Pu, Am, Np, and Cm takes place at the liquid cadmium cathode in the form of cadmium intermetallic compounds (e.g., $PuCd_6$), and uranium deposits as the pure metal after saturation of the cadmium is reached. A typical liquid cadmium cathode deposit contains 3-4 kg of plutonium. A small amount of rare earth fission products deposit together with the heavy metals at both the solid and liquid cadmium cathodes, providing a significant degree of self-protection against diversion of the product materials. The codeposition of uranium and the minor actinides with the plutonium cathode product also serves to make the product less desirable for use as a weapons material.

The cathode products from the electrorefining cell are transferred to the cathode processing unit operation, where they are melted to vaporize any occluded salt or cadmium, forming an ingot of heavy metal. The cathode processor ingots are sampled and sent to the injection casting furnace, where the composition of the batch is adjusted prior to injecting the fuel alloy into fuel slug molds. The fuel slugs are then cut to the appropriate length, and inserted into fuel pin cladding tubes along with heat transfer bond sodium. After fuel pin closure and inspection, the pins are assembled into bundles and loaded into new assembly hardware for return to the reactor.

Laboratory development of the electrorefining process has been carried out with unirradiated fuel containing simulated fission product elements. Both solid cathode deposition and liquid cadmium cathode deposition processes have been proven. The next step in development of the technology is the demonstration of the IFR fuel cycle at the refurbished Fuel Cycle Facility (FCF) at the Argonne National Laboratory site in Idaho. The FCF was used in the 1960s to pyroprocess low-burnup EBR-II uranium-fissium fuel. The refurbished FCF is nearly ready for hot operations, and will be used to process EBR-II spent fuel and provide a supply of recycled fuel for sustained operation of EBR-II. The fuel cycle demonstration will extend over the next several years, with the intent of proving the technical and economic advantages of the IFR fuel cycle.

An integral part of IFR pyroprocess development is the treatment and packaging of high-level waste materials arising from the pyroprocess operations, and the qualification of these wastes for disposal in a geologic repository. As spent fuel batches are processed in the electrorefiner, fission products accumulate in the vessel. Fission products of the alkaline earth, alkali metal, and rare earth groups build up in the electrolyte salt phase. The transition metals and platinum group metals tend to concentrate in the cadmium pool, remain as a sludge in the anode basket, or remain with the cladding hulls. As these fission products accumulate, the heat load due to their radioactive decay processes increases until it exceeds facility or equipment design limits. At that point, it is necessary to remove the heat generating elements. First, the heavy metals present in the salt phase are recovered in a form suitable for subsequent reintroduction to the electrorefiner, by a process known as "drawdown." After the drawdown operation, which reduces the heavy metal content in the salt to less than 0.01 weight percent, the salt and metal phases are removed for treatment to recover the remaining TRU elements and remove a sufficient quantity of fission products that the salt and cadmium can be recycled.

The spent salt (the salt phase after drawdown), containing fission products such as Cs, Sr, I, and the rare earth elements, all in the form of chlorides, is first sent to a salt extraction step where the molten salt is reacted with a liquid U-Cd alloy. The extraction of the TRU elements is carried out in a multi-stage centrifugal contactor, at a temperature of 773K. The (depleted) uranium reduces the chlorides of the transuranic elements, which are present at low concentrations in the salt, with the TRU elements partitioning into the metal phase in metallic form. The TRU-bearing cadmium is returned to the electrorefiner, where the TRU elements are subsequently recovered by electrotransport from the cadmium pool. Initial experiments to characterize the separation efficiency of a single stage contactor are to commence in the near future.

After extraction of the transuranics, the spent salt is sent to a stripping operation in which the salt is reacted with a liquid Cd-Li alloy, again at a temperature of 773K. The lithium is a strong reductant and acts to reduce all of the rare earth chlorides present in the salt; the rare earths concentrate in the cadmium phase, which becomes a waste material, and most of the salt can be recycled to the electrorefiner until the decay heat load builds up to such a level that the alkaline earth and alkali metal fission products must also be removed. The removal of the rare earth elements from the salt is necessary to reduce the heat load in the electrorefiner; because they can be electrotransported with the actinide elements, it is also necessary to keep their concentration in the salt at comparatively low levels to avoid excessive contamination of the heavy metal product. Experiments with the salt stripper have been recently initiated.

The stripped salt is next sent to an immobilization step, where the molten salt is infiltrated through a zeolite column. The zeolite sorbs the fission products by two processes: ion exchange and occlusion of salt molecules in the molecular cage of the zeolite structure. The effluent salt is virtually free of fission products and can be recycled to the electrorefiner. Initial measurements have shown that high fission product loadings in the zeolite can be obtained. The release of fission products from the zeolite-based waste under conditions of groundwater impingement appears to be acceptably small.

The spent cadmium from the electrorefiner and from the salt stripping step is also treated in the IFR pyroprocess, with these two streams combined in a partitioning process where the fission product-bearing cadmium is contacted with a molten aluminum-copper alloy having little solubility for cadmium. The fission products tend to precipitate in the Al-Cu phase as intermetallic compounds, leaving the cadmium phase virtually free of fission products. The cadmium is recovered by retorting and then recycled to the electrorefiner. The Al-Cu alloy, now containing the transition metal, rare earth, and noble metal fission products, can be cast directly into a waste container for repository disposal.

Although the IFR waste treatment and packaging processes are at a relatively early stage of development, they appear to be technically feasible and fully amenable to waste volume minimization initiatives. These processes will be developed and demonstrated at a large scale with simulated fission products, with confirmation of the processes carried out on a somewhat smaller scale as part of the IFR Fuel Cycle Demonstration.

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