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THE INTEGRAL FAST REACTOR - A PRACTICAL APPROACH TO WASTE MANAGEMENT*

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1. INTRODUCTION

The Integral Fast Reactor (IFR) is a concept for a passively inherently safe fast reactor concept with great potential as a future power source. A basic precept in the development of the IFR concept has been the achievement of a complete nuclear power system that is safe, environmentally sound, and economically competitive with other sources of electrical power. These attractive features are obtained in large part by the use of a metallic fuel alloy, U-Pu-Zr, which in turn permits the use of an innovative recycling method that has been termed "pyroprocessing" (Laidler 1993). This is a non-aqueous method that incorporates electrorefining to separate the fission products present in spent fuel from the actinide elements, which can be recycled for

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further fissioning and power generation. Pyroprocessing involves a simple process flowsheet and the use of simple, compact equipment; it has the further attribute of producing a minimal volume of high-level waste and virtually no low-level wastes. Accordingly, a natural evolution of the pyroprocess technology development has been an evaluation of the potential of this system for dealing with the broader problem of the disposal of high-level nuclear wastes. It presently appears that a combined fuel cycle, extracting transuranics from spent light water reactor (LWR) fuel for recycle and burning them in the harder spectrum of the IFR core, can provide tangible benefits. These benefits accrue from the elimination of the transuranics from geologic disposal, from the reduced waste volumes possible with the pyroprocessing operation, and from the reduced uranium mining, milling, and enrichment services otherwise required to support the startup of the IFR generating capacity that can be fueled with the recovered transuranics.

2. PYROPROCESSING AND THE IFR FUEL CYCLE

Pyroprocessing refers to the complete set of unit operations required to recover actinide elements from spent fuel and recycle them to the reactor for use as fuel materials. In the case of the IFR spent fuel, these operations begin with the dismantling of the irradiated fuel assembly and the removal of individual fuel rods. These fuel rods are sent to a chopper, where they are chopped into short (typically, 6.5 mm) lengths. The chopped fuel rod segments are loaded into perforated steel baskets and placed in an electrorefiner. The electrorefining step performs the task of separating the actinide elements from the fission products present in the spent fuel. The process is the same as the electrorefining process used for many years in the minerals industry: an impure metal is made the anode, and it is deposited at a cathode in a condition of greater purity by electrotransport through a suitable electrolyte. In the IFR electrorefiner, virtually pure uranium is collected at a solid mandrel cathode and a mixture of plutonium, americium, neptunium, curium, uranium, and some rare earth fission products is collected at a liquid cadmium cathode suspended in the electrolyte salt. The cathode deposits are recovered after the desired amount of material has been collected and then sent to a cathode processor, which is basically a high-temperature vacuum furnace. The deposits are consolidated in the cathode processor by melting; in the process, any volatile materials that were included in the cathode deposits are removed by vaporization. This includes the electrolyte salt, in the case of the solid mandrel uranium deposits, and cadmium, in the case of the liquid cadmium cathode deposits. The distillates from the process crucible are transported to the condenser region of the cathode processor, where they are collected for recycle to the electrorefiner. The metal ingots resulting from the cathode processing operation are free of unwanted impurities and become the feed material for the next operational step, injection casting.

The function of the injecting casting system is to obtain the appropriate blend of uranium, plutonium, minor actinides, and zirconium and then cast the fuel alloy into slugs suitable for loading into new fuel rods. The fuel batch is induction-melted under vacuum and homogenized, after which the system is pressurized and the fuel alloy is injected into closed-end molds, which are rapidly cooled. The fuel-bearing molds are then sent to the fuel pin processing step; there, the molds are removed, then the fuel slugs are cut to length, inspected, and inserted into fresh fuel pin cladding, which also contains a small amount of sodium for thermal bonding. The top end caps for the new fuel rods are then welded closed, and the bond sodium is distributed along the length of the fuel slug. Another inspection step follows, and the accepted fuel pins are loaded into bundles and installed in new fuel subassembly hardware for insertion into the reactor.

All of these operations are performed remotely, in a highly shielded hot cell facility, because the decontamination factor for fission products in the fuel product is purposely kept low to provide self-protection for the fuel and thus afford a high degree of diversion resistance to the nuclear materials contained therein. Details of the pyroprocess equipment and the plans for demonstration of this process in the Fuel Cycle Facility at the Argonne-Idaho site have been reported previously (Battles et al. 1992, Lineberry et al. 1993). This demonstration began in September 1993 and will extend for several years, for the purpose of verifying the technical and economic feasibility of the IFR fuel cycle.

3. PYROPROCESSING OF OTHER SPENT FUEL TYPES

The IFR pyroprocess is being developed in accordance with rigorous guidelines: (1) make the process applicable with minimum modifications to both IFR and LWR spent fuels, (2) keep the process simple and economical, (3) minimize waste volumes, and (4) produce ultimate waste forms that have acceptable performance characteristics. At this stage in process development, all of these objectives appear to be attainable. Of particular importance is the applicability of the pyroprocess to a broad variety of fuel types; the existence of large quantities of spent fuel and nuclear materials of various types is posing a disposal challenge to the U.S. government and nuclear utilities, and the IFR pyroprocess can afford the means for dealing with this material in a manner that is rational, environmentally sound, and economical.

3.1 Commercial LWR Spent Fuel

The spent fuel discharged from currently operating light water reactors represents a valuable resource for use in future power generation. By the year 2010, there will be over 40,000 metric tons of LWR spent fuel in storage at reactor sites or in monitored retrievable storage. If the decision is made to proceed with the direct disposal of LWR spent fuel in a geologic repository, and if the current reactors continue to operate for the duration of their licenses, and if the first repository is opened by the target date of 2015, the legislated capacity of the first repository will be exceeded before the projected end of spent fuel emplacement. Thus, the preparation of a second repository, possibly with similar costs and similar siting problems, would be required shortly after the first repository goes into operation. At issue is the advisability of a policy based on the disposal of all LWR spent fuel. If the nuclear generating capacity in the U.S. is to increase in accordance with Department of Energy projections, then the fuel resources available in spent LWR fuel could make a significant contribution to the realization of these projections. This fuel contains about 1% transuranic elements that can be used effectively as fuel material in the IFR, where the fast neutron energy spectrum promotes the efficient fissioning of these elements. The spent fuel also contains about

96% uranium, having a ^{235}U content slightly greater than that in natural uranium; this uranium could be re-enriched and recycled to LWRs, and a portion of it could be used as makeup feed to the IFR, either as a blanket material or as makeup core fuel material. Alternatively, the recovered uranium, being free of fission products, could be converted to U_3O_8 and appropriately disposed. Actinide recycle permits sustained growth in nuclear generating capacity without severe environmental penalties from increased uranium mining and milling, and without economic penalties if the process proves to be as inexpensive as expected. The transuranic content of the current annual U.S. output of LWR spent fuel, for example, is sufficient to provide adeq: ate fuel to support the startup of about 1,500 MW(e) IFR generating capacity yearly.

3.2 DOE Spent Fuel Inventory

The U.S. Department of Energy currently holds a large inventory of unprocessed spent fuel, arising from decades of operation of special test reactors, research reactors, and defense- materials production reactors. The spent fuel inventory includes about 100 distinct fuel types, with enrichment levels ranging from natural uranium to highly enriched uranium. Licensing of this wide variety of spent fuel for direct repository disposal could prove to be prohibitively expensive, should it be necessary to qualify each individual fuel type for repository acceptance.

The IFR-LWR pyroprocess can be applied with great effect to the problem of DOE spent fuel disposition. Because the process has been developed for use with metal or oxide fuel, it can be easily adapted for the processing of virtually all of the DOE spent fuel types: metal, oxide, graphite, cermet, matrix, etc. At this time, pyroprocessing appears to be practical for all fuel types except aluminum-based fuels. These fuels are perhaps better treated by conventional aqueous methods, because the aluminum tends to form stable intermetallic compounds with the actinide elements and makes their removal more difficult. Pyroprocessing is eminently suited for treatment of all other fuel types, requiring only a modification to the head end of the process to adapt it to each broad fuel class.

The use of pyroprocessing for DOE spent fuel management has a number of benefits: (1) the recovery of actinide elements, such as highly enriched uranium, for subsequent re-use in power generation; (2) the elimination of the need to dispose of highly enriched material, thereby avoiding concerns with *in situ* criticality events; (3) a substantial reduction in packaged waste volume for ultimate disposal; and (4) the production of a common waste form regardless of starting fuel type. Pyroprocessing of this wide variety of materials can be done with a common basic process, with common equipment and procedures. This would result in greatly improved economics of waste management. The actinides recovered in the course of pyroprocessing, as in the case of the IFR fuel cycle, are co-deposited so that there is no production of a separate stream of weapons-usable material.

4. PYROPROCESSING OF LWR OXIDE SPENT FUEL

Researchers at the Argonne National Laboratory have been working to develop a method for processing LWR spent fuel that is fully compatible with the IFR system. Summaries of progress to that end have recently been reported (Pierce et al. 1993, McPheeters et al. 1993). Basically, the process for recovering actinides from spent LWR fuel involves the reduction of the LWR oxide fuel to metallic form, followed by the separation of fission products and the separation of the bulk of uranium from the transuranic elements.

Two processes are under development for performing this reduction and actinide extraction. In the *Salt Transport Process*, oxide fuel is reduced by reaction with calcium at 800°C in a reaction vessel containing $CaCl_2$-CaF_2 salt and a Mg-Cu-Ca alloy. The transuranic elements present in the spent fuel dissolve in the liquid alloy, and uranium precipitates as solid metal. The TRU elements and rare earth fission products are extracted from the liquid Mg-Cu alloy by contact with $MgCl_2$, from which in turn the TRU elements and rare earths are extracted into a molten Zn-Mg alloy. Nearly all of the noble metal fission products and over 99% of the uranium remain in the Mg-Cu alloy. Uranium in an amount about equal to the quantity of plutonium transports to the Zn-Mg alloy with the TRU elements. The Zn-Mg alloy containing the

TRU product is then retorted to remove the zinc and magnesium, and the remaining actinides and rare earths are sent to the IFR electrorefiner for recovery of the actinide elements. The uranium precipitate, including the noble metal fission products, is recovered from the Mg-Cu alloy and sent to a large electrorefiner, where the uranium is separated from the fission products by electrotransport to a solid cathode. The pure uranium product can then be recycled to the LWR or the IFR.

The second process, the *Lithium Process*, uses lithium as the reductant. Uranium and the transuranic elements are reduced to their metallic state, together with the noble metal fission products, while the other fission products remain in the reductant salt waste. The metals are sent to the electrorefiner for separation of the uranium by deposition on a solid cathode. After a large quantity of uranium has been recovered in this manner, the TRU concentration in the electrorefiner salt will build up to a sufficient extent that the salt can be transferred to the IFR electrorefiner for production of a TRU fuel product that is usable in the IFR. The noble metal fission products remain in the electrorefiner metal phase as in the IFR electrorefining process. The Lithium Process offers the advantages of lower operating temperatures and a salt waste stream that is of the same composition as that produced in the IFR process.

5. WASTE MANAGEMENT

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. This is the first time that a reactor technology development program has had as a major program element the development of high-level waste management operations before the wastes were actually produced.

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 (more electrochemically noble 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 500°C. 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 in progress (Chow et al. 1993).

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 500°C. 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 the rare earth concentration in the salt at comparatively low levels to avoid excessive contamination of the heavy metal product. Experiments with the salt stripper have been completed and reported recently (Carls et al. 1993).

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: up to 25 weight % fission products is possible. After sorption of the fission products has been accomplished, the waste-loaded zeolite pellets are subjected to a blowdown with hot argon gas to remove adherent surface salt. Then a small quantity of anhydrous zeolite powder is added to immobilize any residual surface salt, and the waste-loaded zeolite is hot pressed to form a solid monolithic mineral waste form. The release of fission products from the zeolite-based mineral 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.

An alternative version of the metal waste form, also under active development at this time, incorporates the cladding hulls as the matrix material. Depending on starting fuel type, this material can be either stainless steel or zirconium alloy. With this approach, a substantial fraction of the original fuel assembly hardware can also be included in the waste form while achieving minimum packaged waste volume for 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.

6. CONCLUSIONS

Development of the method for pyroprocessing of spent fuel from the Integral Fast Reactor (or Advanced Liquid Metal Reactor) is progressing well, and has reached the technology demonstration phase, in which recycle will be demonstrated with irradiated fuel from the EBR-II reactor. Methods for recovering actinides from spent LWR fuel are at an earlier stage of development but appear to be technically feasible at this time, and a large-scale demonstration of this process has begun. The utilization of fully compatible processes for recycling valuable spent fuel materials promises to provide substantial economic incentives for future applications of the pyroprocessing technology.

7. ACKNOWLEDGMENT

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