

**PROGRESS AND STATUS OF THE INTEGRAL FAST REACTOR (IFR)
FUEL CYCLE DEVELOPMENT**

C. E. TILL
Associate Laboratory Director
Engineering Research

ANL/OTD-ER/CP--79006

DE93 009967

and

Y. I. CHANG
General Manager
IFR Program

Argonne National Laboratory
Argonne, Illinois

Presented to

AMERICAN POWER CONFERENCE
Illinois Institute of Technology

MAR 26 1993
OSTI

Chicago, Illinois

April 13-15, 1993

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

The submitted manuscript has been authored by a contractor of the U. S. Government under contract No. W-31-109-ENG-38. Accordingly, the U. S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

**PROGRESS AND STATUS OF THE INTEGRAL FAST REACTOR (IFR)
FUEL CYCLE DEVELOPMENT**

C. E. TILL
Associate Laboratory Director
Engineering Research

and

Y. I. CHANG
General Manager
IFR Program

Argonne National Laboratory
Argonne, Illinois

establish the safety database required for licensing. Electrorefining experiments are routinely carried out on a 10-kg engineering scale. Anodic dissolution was successfully demonstrated, speeding dissolution, which had previously slowed the rates in the IFR pyroprocesses.

The focus is now on the demonstration of the entire IFR fuel cycle. The next key milestone is to complete the refurbishment of the original EBR-II Fuel Cycle Facility. Rapid progress is being made on the facility modifications as well as the fabrication of process equipment modules. The status of the EBR-II Fuel Cycle Facility modifications project is presented in a companion paper.

ABSTRACT

The Integral Fast Reactor (IFR) fuel cycle holds promise for substantial improvements in economics, diversion-resistance, and waste management. This paper discusses technical features of the IFR fuel cycle, its technical progress, the development status, and the future plans and directions.

METAL FUEL PERFORMANCE

Metal fuel is key to realizing the inherent passive safety potential of the IFR and, importantly, it also allows compact pyroprocessing. Therefore, development of a complete technical database on metal fuel performance is an essential element of the IFR Program. The irradiation test program has included a comprehensive range of design and operating conditions. The U-Pu-Zr composition has varied from no plutonium to 28 wt% plutonium with zirconium variation from 2 to 14 wt%. Three cladding materials have been used, which include the two austenitic claddings, SS316 and D9, and the ferritic cladding, HT9.

INTRODUCTION

The Integral Fast Reactor (IFR) development covers the entire reactor system, not only the reactor itself, but also its fuel cycle, both fuel refining and fabrication, and waste management as well. The goal is to develop a reactor system that answers the questions facing the next generation of nuclear power plants with respect to economics, safety, and waste disposal. As such, the program emphasis has shifted each year as the development has proceeded.

The lead irradiation tests began in February 1985, and reached a burnup level of 18.4 atom%, far exceeding their design target of 10% burnup. Interlinkage of gas bubbles and rapid gas release at 1 to 2% burnup mitigates fuel-clad mechanical interaction because the resulting porous fuel is plastically compliant at temperature. This is the key factor for achieving a high burnup with metallic fuel.

Initially, the focus was placed on demonstration of passive safety characteristics. The loss-of-flow without scram and loss-of-heat-sink without scram tests at EBR-II were a spectacular demonstration of the inherent passive safety potential of the IFR concept. The emphasis then shifted to development of a fuel performance and pyroprocessing technology database, and rapid progress has been made during the past few years.

Radial fuel growth dominates the swelling process; at ~2% burnup the clearance between fuel and cladding has closed and the resulting fuel-cladding contact causes frictional forces to impede further axial growth. Initial apparent axial and radial anisotropy has been related to radial gradients in creep and irradiation growth of noncubic phases in the fuel, and fuel swelling is now thought to be well-understood.

The high burnup potential of the new IFR fuel alloys has been demonstrated. EBR-II is now fully converted to the IFR fuel alloys and statistically significant fuel performance data are being generated. A new whole-pin furnace experimental facility was installed, which permits in-depth evaluation of fuel failure mechanisms over a broad range of temperatures and transient conditions to

At high burnup (between 15 and 18% burnup) a gap between fuel and cladding tends to reestablish itself near core midplane due to the high swelling nature of the austenitic, D9 cladding. The 20% cold-worked D9 cladding performed satisfactorily to the 18.4% peak burnup reached. Peak diametral strain due to creep and swelling ranged from an average of ~1% at 10 atom% burnup to 7% at 18 atom% burnup. Bundle-duct interaction then set the irradiation limit.

Since the cladding strain in the ferritic HT9-clad fuel is much less at high burnup, the irradiation tests with HT9 cladding, which now have achieved 17.7% burnup, are expected to continue their irradiation beyond 20% burnup.

Complementary irradiation testing of metallic fuel in FFTF was conducted to demonstrate that the database generated with fuel of the limited EBR-II core height (34.3 cm) is directly applicable to performance of commercial IFR cores approximately three times this height. The IFR-1 test in FFTF with D9 cladding and Pu compositions up to 19% was completed, having reached the goal burnup of 10 atom%. Postirradiation examinations indicate perfectly satisfactory performance. The behavior of the fuel pin bundle was good and stability of the full length fuel column was excellent. Axial growth was at the lower end of the data scatter of equivalent EBR-II fuel pins. There were no signs of fuel movement or densification in any of the pins examined. Profilometry data did not show any step changes at the end of fuel column, suggesting very little fuel-cladding mechanical interaction.

Irradiation tests to date have clearly demonstrated excellent performance characteristics of the metallic fuel in both steady-state and off-normal operating conditions. EBR-II is now fully loaded with the IFR fuel alloys and statistically significant fuel performance data are being generated.

PYROPROCESSING

Pyroprocessing, which utilizes high temperatures and molten salt and metal solvents, can be advantageously utilized for processing metal fuels because metal is both a suitable feed for such processes and the product as well and is thus suitable for fabrication into new fuel elements. Direct production of metal as the product avoids the further chemical conversion steps necessary in reprocessing by the conventional Purex solvent extraction process.

Electrorefining is the key step in the IFR pyroprocessing. Following disassembly of the fuel assemblies, the fuel pins are chopped into short lengths, which are packaged to form a batch for dissolution in the electrorefiner. Bulk uranium, especially from the blanket, is electrorefined by deposition on a solid cathode. The remaining uranium along with plutonium and other minor actinides are electrorefined by deposition into a liquid cadmium cathode. The cathodes are removed from the electrorefiner cell, the cadmium and occluded salt removed by retorting, and the uranium or uranium-plutonium-actinide product is consolidated by melting.

To assure criticality-safe configuration during retorting process, the cathode deposit size of 10 kg has been

selected as the reference. Uranium deposits at the 10-kg scale are now routinely produced in the engineering-scale electrorefiner facility. The deposit rate of 10 kg in about 24-hour period promises economic throughput for commercial-scale operation when multiple electrodes are employed to increase the batch size.

The plutonium electrotransport to liquid cadmium cathode has been demonstrated on laboratory-scale involving few hundreds of grams. This combined with simulated uranium deposits in liquid cadmium cathode indicate that large-scale plutonium electrotransport would be successful in the EBR-II Fuel Cycle Facility as well, and this is the reference process for this facility.

IFR WASTE TREATMENT PROCESSES

Pyroprocessing lends itself to much simplified waste treatment operations. The volume of low-level waste is minimal and troublesome effluents such as tritium, carbon-14, krypton, iodine and ruthenium can be easily contained or immobilized.

The principal high-level wastes from the IFR pyroprocessing are the metals and salts discharged from the electrorefiner. To recover the actinide elements in the salt and separate them from rare earths, the salt is first contacted with a Cd-U solution in a multi-stage, countercurrent extraction train. The uranium in the metal phase exchanges to a greater extent with actinide elements in the salt phase than with rare earths. It is expected that four to six stages in a countercurrent extraction train will suffice to obtain the desired separation of actinides from lanthanides and reduce the actinide content of the salt to the 10^{-3} to 10^{-4} range.

After the actinide elements are separated from rare earths in the extraction train, the entire batch of electrorefiner salt is fed to the stripper vessel where three steps are performed: stripping, cladding hull washing, and alkali metal chlorination.

Although the treated salt will contain only trace amounts of actinides and very small amounts of rare earth fission products, it will contain Cs and Sr and therefore needs to be suitably packaged as high-level waste. The goal is: (1) to immobilize the waste salt in an inert, leach-resistant matrix, and (2) to densify the matrix to yield a mechanically strong, solid monolith. This matrix will then be sealed in a durable metal container. As the first step toward identifying a potential matrix for chloride salts, the composition of various minerals were considered. Naturally occurring minerals which occlude chloride salts are known. Examples of chloride occluding minerals are sodalite, scapolite, and cancrinite. These minerals are zeolites, i.e., crystalline alkali metal and/or alkaline earth aluminosilicates that are

characterized by an open framework with relatively large cavities and channels. Salt molecules are typically held in these cavities and are not readily released.

Efforts to identify zeolites that would occlude IFR waste salt and a process that would yield analogs of these naturally occurring minerals have been successful. Promising results were obtained by contacting dehydrated A50, a synthetic zeolite having sodium as an exchangeable cation, with a molten salt. The experiment consisted of pouring the molten salt at 450°C into a column of A50 supported on a quartz frit and allowing the salt to percolate through the column. The salt that flowed through the column and frit was collected and analyzed.

Two types of reactions, ion exchange and salt occlusion, occurred between the molten salt and the zeolite. Most of the sodium ions on the zeolite were exchanged for ions in the salt, and the zeolite occluded about 40% of its weight in salt.

From the chemical analyses, it could not be determined which cation is in the zeolite structure and which is in the occluded salt. However, it is clear that sodium in the original zeolite has exchanged almost completely with other cations. In addition, the cations in the zeolite-salt were enriched in Li, Ba, Sr, and Cs compared to their concentrations in the original salt.

These results are encouraging. They demonstrate that zeolites can sorb the cations of interest. The zeolite materials used in these experiments are in the form of powders, which would not be suitable for a high-level waste form. Laboratory work to identify a method for solidifying the zeolite-salt compounds is at a very preliminary stage. Experiments have been conducted to test the feasibility of densifying a matrix consisting of zeolite and copper powders. Hot isostatic pressing of various mixtures at 375°C and 69 MPa (10,000 psi) yielded solid monoliths that were relatively strong. Further experiments are planned in which zeolites with sorbed salt will be hot isostatically pressed with copper powder.

The metal waste from electrorefiner are combined with zirconium, cladding hulls, and the stripping alloy, which contains the rare earth fission products and uranium. This mixture is fed to the waste metal retort, where it is mixed with copper, then most of the cadmium is distilled off and recycled. A small fraction is left with the metallic residue, the remainder is equivalent to the fission product cadmium and the CdCl₂ added to the electrorefiner to oxidize rare earth, alkali metal, and alkaline earth fission products in the fuel. It is expected that the cadmium, uranium, and most of the fission products will be dissolved in the copper. The metal waste mixture will be densified

by pressing or melting, then encapsulated in a thick, corrosion-resistant container for disposal.

APPLICATION OF PYROPROCESS TO LWR OXIDE FUEL

In IFR pyroprocessing, minor actinides accompany the plutonium product stream. Therefore, actinide recycling occurs naturally in the IFR, which is another important attribute as a next-generation reactor concept. A logical question is whether the pyroprocessing approach can be extended to extract actinides from LWR spent fuel. And, in fact, it turns out that there is an extensive experience base at Argonne in applying pyrochemical processes to oxide fuel.

In the late 1960's, the EBR-II Fuel Cycle Facility operated for about five years using a simple drossing process, known as melt refining. In this process the electropositive fission products were separated from the fuel by reaction with a zirconium oxide crucible. The volatile elements were released and collected on a fume trap or condensed cryogenically from the cell atmosphere.

A pyrochemical process for recovering the actinides occluded with the dross, or crucible skull, was developed and demonstrated on an engineering scale with simulated fuel. This skull reclamation process employed liquid zinc-magnesium and molten chloride salt as process solvents. Also a blanket process for recovery of plutonium from metallic uranium blankets was demonstrated on a bench scale. This process involved the selective extraction of plutonium from molten uranium into an immiscible magnesium phase.

The techniques developed to process EBR-II skulls and blankets were then extended to processing uranium oxide and mixed oxide fast reactor fuels. Rapid methods for reducing dense oxide fuels were demonstrated, and a liquid metal-fused salt extraction step was developed for isolating the uranium, plutonium, and fission products. This "salt transport process" was demonstrated on a laboratory scale, but funding was terminated before a planned pilot-plant demonstration could be completed.

The earlier pyrochemical process development efforts were discontinued because there was no clear advantage over the traditional Purex process that produces a pure plutonium product stream. Today, however, the goal has changed. In traditional reprocessing based on Purex, the goal was to produce a highly decontaminated, pure Pu product stream. However, when LWR processing is viewed as a waste management strategy, the goal is quite different. Neither a pure Pu product stream nor a high decontamination factor is required. In fact, just the opposite is desirable. The new

process goals, when LWR spent fuel processing is viewed as a waste management strategy, are as follows:

- Direct extraction of all actinides (Pu, Np, Am, Cm, etc.) from the spent fuel as a single product stream.
- An actinide recovery target of 99.9% to 99.99%.
- The process should be incapable of producing pure Pu product.
- The process should be incapable of achieving a high decontamination factor for fission products.
- The process should be simple enough to achieve acceptable economics.

A preliminary assessment has been made to investigate the feasibility of using pyrochemical processes for directly extracting actinides from LWR spent fuel, satisfying the new process goals. It appears that pyrochemical processes are compatible with the new process goals and two promising flowsheet options have been identified: (1) a salt transport process and (2) a magnesium extraction process.

Pyrochemical processes appear to be a natural fit to the LWR actinide extraction application and should provide significant advantages over the traditional Purex-based processes. First, potentially all actinide elements are extracted in a single product stream, along with most rare earth fission products. A pure plutonium product is not possible. The product is highly radioactive and is not much more attractive than the original spent fuel as far as the diversion risk is concerned. The process as such therefore provides some nonproliferation protection.

Second, in these processes uranium remains as metal ingots with some noble fission product contamination. In this form the uranium can be easily stored for later recovery and use in the IFRs. The actinide extraction processes then deal with only 1 or 2% of the total heavy metal. This small mass flow and the few process steps involved lead to compact equipment systems and small facility size, and should portend favorable economics.

As a spin-off from the IFR technology, a small-scale R&D program has been initiated at Argonne to develop the necessary database to judge technical feasibility of the proposed flowsheets by the end of FY 1995.

FUTURE DIRECTIONS

The IFR Program is now at a critical juncture. Technical feasibility has been demonstrated and the technology database has been established to support its practicality. The Program is entering its demonstration phase. EBR-II is now completely fueled with IFR fuel alloys, and rapid progress is being made on the refurbishment of its Fuel Cycle Facility. When completed next year, EBR-II and its Fuel Cycle Facility will then start operating as a pilot-plant scale IFR prototype.

This prototype demonstration, expected to be completed by the end of 1995, will include near-commercial-scale IFR fuel cycle processes, actinide recycle technology, and substantial progress in waste form certification. The IFR prototype fuel cycle demonstration is the crucial step in proving the practicality of the next-generation nuclear fuel cycle.

ACKNOWLEDGMENT

Work supported by the U.S. Department of Energy under Contract W-31-109-Eng-38.

END

**DATE
FILMED**

6 / 22 / 93

