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INTEGRAL FAST REACTOR PROGRAM
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ABSTRACT

This report summarizes highlights of the technical progress made in the Integral Fast Reactor (IFR) Program in FY 1993. Technical accomplishments are presented in the following areas of the IFR technology development activities: (1) metal fuel performance, (2) pyroprocess development, (3) safety experiments and analyses, (4) core design development, (5) fuel cycle demonstration, and (6) LMR technology R&D.

I. IFR PROGRAM OVERVIEW

The IFR Program continued to make progress during the reporting period in all aspects of fuels performance demonstration, pyroprocess development, safety experiments and analyses, core design development, fuel cycle demonstration, and LMR technology R&D. A steady progress has been made on the Fuel Cycle Facility (FCF) modification project. It reached a major milestone when the main hot cell was filled with argon gas for the first time in almost 20 years. All major process equipment systems required for the IFR fuel cycle demonstration, namely, element chopper, electrorefiner, cathode processor, injection casting furnace, etc., have been procured, assembled, tested for engineering qualification, and are in the final stage of being installed in the hot cell.

The University of Chicago Special Advisory Committee for the IFR continues to be enthusiastic about the potential of the IFR and is highly impressed with the technical progress as indicated by the following excerpts from the executive summary of the Committee report:

"The Committee compliments the Argonne National Laboratory (ANL) staff on the major accomplishments in the fuel program during the past year. Progress in 1993 toward the development of a fuel specification has been particularly substantial.

...The development of waste treatment processes leading to acceptable waste forms will be no less demanding than development of the current process flow sheets and the design of equipment for recycle of U, Pu, and other actinides. The conversion of chemical process wastes to forms acceptable for national repository disposal must meet the stringent specifications and quality control requirements of the regulatory authorities. Argonne has made notable progress in this direction in the past year.

...Prevention of proliferation of nuclear weapons is a national priority, and is the foremost concern of many critics. It is important that this be recognized during the fuel cycle demonstration and that nothing be done which reduces the nonproliferation advantage of the IFR cycle. Because plutonium cannot be separated in a pure form in the IFR fuel cycle; because the plutonium is always accompanied by higher actinides and highly gamma-active fission products; because even the impure plutonium is always in a very radioactive state and within a sealed reprocessing plant; and because the plutonium need never leave the reactor site, the IFR fuel cycle is highly proliferation-resistant, much more so than the solvent extraction (PUREX) fuel cycle normally used with oxide fuels.

...We recommend the design and initiation of construction of a demonstration IFR plant at an appropriate time in the development program. This first plant, with government subsidies as necessary, should be followed at suitable intervals by other demonstration plants, each plant benefiting from what was learned in preceding plants and involving progressively more private-industry involvement. The objective should be to have a standardized, and certified, design available for commercial exploitation on a broad scale as national and international energy supply needs develop in the future. Ideally, this effort should be made as an international cooperative program, and it is timely to initiate steps toward such international collaboration now."

In parallel to the IFR fuel cycle demonstration, Actinide Recycle Program has been developing application of similar pyrochemical processes to extract actinides from the LWR spent fuel. This application involves first reducing the LWR oxide fuel into metal phase, and three or four promising flowsheets were

identified for extraction of actinides from uranium. The pyrochemical processes for the LWR spent fuel are in the early stage of R&D and much engineering development is needed before economic feasibility can be demonstrated.

However, there have been a couple of major technical breakthroughs during the past year. First, laboratory-scale experiments confirmed that Li reduction process is feasible and in fact preferable to the original Ca reduction process. The advantages of the new Li reduction process are:

- Operating temperature can be reduced from 800-850°C range to 500-650°C range, solving the high temperature materials development issue. With the Li reduction process stainless steel can be used for vessels and transfer lines, instead of tungsten or other exotic materials required for the Ca reduction process.
- Li reduction process allows the electrorefining process being demonstrated for the IFR fuel cycle to be directly applicable to the LWR processing as well.
- The LiCl salt waste treatment will be identical to the IFR waste salt treatment processes, and the resultant final waste forms (for disposal in a high-level waste repository) will be the same, simplifying the qualification of these wastes for disposal.

For these reasons, the new Li reduction process followed by electrorefining was selected as the reference flowsheet for the LWR application. The original Ca reduction followed by salt transport process to extract actinides is kept as a backup option.

A second breakthrough also follows from the Li reduction process. since the metal precipitates conglomerate at the bottom of the reduction vessel, a much simplified LWR/IFR symbiotic fuel cycle is possible which incorporates the LWR actinides directly into the IFR fuel cycle without the extraction step. The metal precipitates which contain uranium, entire actnides, and noble metal fission products can be directly made into the IFR fuel by injection casting along with externally supplied plutonium or actinides. This very simple LWR/IFR

sympiotic fuel cycle is described in Fig. I.1. In this scheme, the LWR spent fuel processing is minimal--a simple oxide to metal reduction. Except for volatile fission products and most of rare earth fission products that remain in the salt, the entire spent fuel is directly incorporated into the IFR fuel cycle. This also provides a quick measure to denature excess weapons plutonium or other separated plutonium.

Fig. I.1. LWR/IFR Sympiotic Fuel Cycle Based on Lithium Reduction Process

II. METAL FUEL PERFORMANCE

A. Zirconium-sheathed Fuel Element Designs

1. Introduction

The current fabrication process for metallic fuel includes injection casting of the molten fuel alloy into quartz molds. The quartz is broken from the fuel slug after the fuel has solidified, resulting in a contaminated quartz waste stream. This waste stream should be eliminated, if possible, for EBR-II fuel, and must certainly be eliminated from commercial IFR designs such as PRISM.[1] Development efforts on alternate fabrication techniques are underway. These advanced fabrication alternatives include moldless fabrication with the Creare continuous casting machine[2] and niobium-based reusable molds.[3] This report documents a preliminary design study of a third alternative; namely, the use of zirconium molds that remain as part of the fuel slug and are permanently incorporated into the fuel element. This concept will be referred to as Zr-sheathed fuel.

Previous experience with Zr-sheathed fuel is limited to the experiment X492[4] which is still undergoing irradiation and is currently at 7.7% burnup. Experiment X492 includes 28 binary (U-3Zr) and 12 ternary (P-20.5Pu-3Zr) fuel slugs that were cast into 8-mil Zr sheaths and clad in 316SS. The fuel smeared density was 75%. Interim exams have shown that these Zr-sheathed fuel elements are performing well, and in fact, offer some improvement in fuel performance over standard cast fuel (e.g., reduced fuel axial growth). A hoped-for improvement in fuel/cladding chemical interaction (FCCI) from the Zr sheaths, however, has not been observed; the Zr sheaths in X492 split, apparently early in life, exposing the fuel to the cladding.

Despite the good performance to date of X492, the design used in that experiment would probably not be acceptable as a driver fuel. First, 3 wt% Zr may be too little Zr in the fuel alloy for some reactor/fuel designs. The X492 fuel alloy has a solidus $\sim 90^{\circ}\text{C}$ lower than the solidus for standard Mk-V fuel (U-20Pu-10Zr), reducing the margin to melting of the as-cast fuel by approximately 20% at nominal operating temperatures. Furthermore, the low Zr

fuel that extrudes through the sheath cracks and contacts the cladding may pose an increased FCCI problem, as well as lower the fuel-clad eutectic temperature relative to the standard Mk-V design. Second, the sheath thickness has not been optimized for reliable fabrication. As thin a sheath as is acceptable to assure reliable fabrication is desired so as to minimize the deviation of the fuel alloy from the standard Mk-V composition. A thicker sheath may be necessary for strength as the casting temperature increases; the casting temperature will increase for fuel compositions containing higher Zr contents.

This report presents the results of a scoping study to identify acceptable Zr-sheathed fuel designs based on the experience with X492. Both Mk-V equivalent (EBR-II) designs and PRISM equivalent designs are considered. Two different approaches may be taken to fabricate Zr-sheathed fuel. As in experiment X492, the Zr sheath may be used essentially as a mold into which the fuel is cast. In this case, the Zr sheath must have sufficient strength to contain the injected fuel melt at the casting temperature with minimal to no dimensional changes to the sheath. A second approach would use a thin Zr sheath (possibly only a few mils thick) as an insert into a reusable mold.[5] In this case, the Zr sheath serves simply as a barrier to interaction between the fuel melt and the permanent mold material; thus, the Zr sheath can be thin since it does not need the strength to contain the injected fuel melt on its own. The calculations presented in this report are pertinent to either design strategy. The designs considered encompass Zr contents of 6 to 12 wt%, Pu contents up to 30 wt%, and sheath thicknesses of 0 to ~30 mils.

2. Methodology

For the purpose of this study, a Mk-V or PRISM equivalent Zr-sheathed fuel design is defined as an element design that would produce an equivalent element power to that of a standard Mk-V or PRISM fuel element. This definition is assumed to be the same as requiring that a Mk-V equivalent Zr-sheathed fuel slug have the same quantity of fissile U and Pu as a standard Mk-V slug, and likewise for the PRISM designs. This criterion was considered necessary to minimize impacts of new sheathed fuel elements on the core designs.

The methodology used for calculating Mk-V equivalent Zr-sheathed fuel elements is as follows. Three parameters--fuel smeared density, fuel Zr content, and Zr sheath thickness--were independently varied. Calculations were made for smeared densities of 75 and 80% and for Zr contents of 6, 8, 10 and 12 wt%. The smeared density was taken as the ratio of the cross-sectional fuel area (at sheath inner diameter) to the sum of the cross-sectional areas of the fuel and the slug-cladding gap; the cross-sectional area of the sheath is not included as part of the fuel. For each smeared density and Zr content, calculations were made for varying sheath thicknesses (in 1-mil increments) starting at 0 and going up to either a 30-mil sheath thickness or 30 wt% Pu content in the fuel (whichever came first). For each case the Zr content, smeared density, sheath thickness (and therefore fuel radius), and fissile mass were fixed. Thus, the parameters to be determined were the fissile uranium enrichment and the resulting U and Pu weight fractions in the fuel alloy. The Pu isotopic distribution was assumed fixed at the standard Mk-V ratios. This turned out to be an iterative calculation since the fuel density changes continuously with the alloy composition. The correlation used to describe the variation of the U-Pu-Zr alloy density was the following:

$$\rho = 18.811 - 2.793 W_{\text{Pu}} + 3.782 W_{\text{Pu}}^2 - 28.431 W_{\text{Zr}} + 16.112 W_{\text{Zr}}^2 + 16.112 W_{\text{Zr}}^2 \quad (1)$$

where

- ρ = density in g/cm³ (at 293K),
- W_{Pu} = the weight fraction of Pu in the alloy, and
- W_{Zr} = the weight fraction of Zr in the alloy.

The correlation was developed for this study using the data given in the Metallic Fuels Handbook[6] for U-Pu-Zr alloys having Pu contents from 11.1 to 20.0 wt% and Zr contents from 6.3 to 13.5 wt%, as well as elemental densities. The fit to the data is excellent; the correlation coefficient for the fit is 99.98% and the standard deviation of the correlation with respect to the fit data is ± 0.06 g/cm³.

The calculations for the PRISM equivalent Zr-sheathed designs were essentially the same as for the Mk-V equivalent designs. For the PRISM designs, however, depleted uranium was employed for the U-component in the fuel.

Calculations were made for fissile Pu enrichments of 0.856 (standard PRISM enrichment) and 0.93 (weapons-grade Pu); only a smeared density of 75% was investigated for the PRISM equivalent designs.

3. Results

The results of the calculations described in the previous section are shown in Figs. II.1 through II.4. The calculated solidus and liquidus temperatures for each alloy, according to L. Leibowitz,[7] are given in Figs. II.5 through II.8. In the legends of these figures, the Zr contents listed refer to the Zr weight fraction of the fuel; FBE denotes the PRISM Fissile Break-Even design, and W-Pu denotes the PRISM design that employs weapons-grade Pu. Figures II.1 and II.2 show how the uranium enrichment and Pu weight fraction vary with sheath size for the Mk-V equivalent designs. Figures II.3 and II.4 show how the Pu weight fraction varies with sheath size for the PRISM equivalent designs (the uranium is assumed to be depleted U). Correspondingly, Figs. II.5 through II.8 show how the fuel solidus varies with sheath size for each of the cases considered.

Fig. II.1. Mk-V Equivalent Designs for 75% Smeared Density Fuel

Fig. II.2. Mk-V Equivalent Designs for 80% Smeared Density Fuel

Fig. II.3. PRISM Equivalent Designs for 85.6% Enriched Pu

Fig. II.4. PRISM Equivalent Designs for 93% Enriched Pu

Fig. II.5. Variation of Fuel Solidus for 75% Smeared Density Mk-V Fuel Designs

Fig. II.6. Variation of Fuel Solidus for 80% Smeared Density Mk-V Equivalent Fuel Designs

Fig. II.7. Variation of Fuel Solidus for 85.6% Enriched PRISM Equivalent Fuel Designs

Fig. II.8. Variation of Fuel Solidus for 93% Enriched PRISM Equivalent Fuel Designs

4. Discussion

Each Zr-sheathed fuel design shown in Figs. II.1 and II.2 is equivalent to a standard Mk-V fuel element. They will all produce essentially the same power since they have identical fissile masses. Variations in U-238 and Pu-240 content will affect element powers slightly; element power estimates including U-238 and Pu-240 contributions show that the power from the sheathed fuel designs differ from the standard designs by up to 3%. This difference could be eliminated with further composition refinement. The neutronic characteristics of the different element designs will be slightly different due to the differing masses of U-238 (which is a resonant absorber). However, the neutronic effects of varying U-238 contents were not considered in this study.

Selection of an optimum Zr sheath thickness will be influenced by fuel fabrication as well as irradiation performance considerations. If the sheath is used as the primary mold and is too thin it will not have the strength to contain the injected fuel melt during casting without significant dimensional changes. If the sheath is used as an insert into some other permanent mold and is too thin, dissolution of the sheath could occur resulting in chemical interaction between the melt and the mold. On the other hand, the consequences

of a sheath that is too thick are unclear. The 8-mil sheath used in X492 split early in life and radial swelling and gas release for this sheathed fuel appear consistent with that of unsheathed fuel. If the sheath were so thick that it did not split (or were to split late in life), the gas release and fuel swelling (axial and radial) would be expected to differ significantly from that of unsheathed fuel, resulting in a different overall fuel performance. Experimental studies will be required to find the optimal sheath size or range of acceptable sizes.

Figure II.1 shows how uranium enrichment and Pu content vary for the 75% smeared density, Mk-V equivalent designs. In the range of expected acceptable sheath thicknesses (~10 mils), an increase in the sheath thickness of 2 mils requires an increase in Pu content of ~1% and an increase in U-235 enrichment of ~3% to retain the standard Mk-V element power. Figure II.2 shows the same data for the 80% smeared density, Mk-V equivalent designs. Using an 80% smeared density design decreases the necessary Pu content by ~1% and the U-235 enrichment by ~5% over the corresponding 75% smeared density design.

Figures II.3 and II.4 show the variation of Pu content for the PRISM equivalent Zr-sheathed fuel designs. The impact of the Zr sheath on the fuel composition for the PRISM equivalent designs is much less significant due to the larger dimensions of the PRISM elements. For reasonable sheath thicknesses (~10 to 20 mils), a change in sheath thickness of up to 10 mils requires only an increase in Pu content of ~5% using the standard PRISM fissile Pu enrichment of 85.6%. The impact on Pu content is even less when employing the weapons-grade Pu enrichment of 93%.

Figures II.5 through II.8 show the dependence of fuel solidus temperature on sheath size for the designs considered. For the Mk-V equivalent designs, approximately 3°C of operating margin to solidus temperature is sacrificed for each mil of sheath incorporated into the design; alternatively, ~10°C of solidus temperature is sacrificed for every percent of Zr removed from the fuel alloy. Again, the PRISM equivalent designs are less sensitive to design changes than are the Mk-V equivalent designs.

For Mk-V equivalent designs, it is possible to retain at least the Mk-V as-cast solidus temperature for the 75% smeared density designs only by increasing the Zr content above 10 wt%. For the 80% smeared density designs, the Mk-V solidus temperature can be retained for 10 wt% Zr designs with small sheaths; thus, the Zr-sheath insert into a different permanent mold could be attractive. The 12 wt% Zr fuel can retain the Mk-V solidus temperature for sheaths up to 10 mils; this sheath size could be sufficient for use with a Zr sheath employed as the primary mold. Alternatively, an as-cast fuel solidus temperature lower than the Mk-V reference might be acceptable, particularly if the fuel solidus temperature, not of the as-cast fuel, but rather of a Zr-depleted zone formed during irradiations is the limiting factor.

5. Conclusions and Recommendations

Acceptable Zr-sheathed fuel elements can, in principle, be designed for use with both EBR-II and PRISM reactors. The goal of this effort was to calculate a wide range of Mk-V equivalent and PRISM equivalent Zr-sheathed fuel designs that can be evaluated in more detail with subsequent experimental studies and safety/thermal performance calculations. This study has identified potentially acceptable designs for both reactor types.

While an acceptable Zr-sheathed fuel element can be designed for use in EBR-II, it can be done so only with significant deviations in fuel composition from the standard Mk-V design. Potentially acceptable designs (8- to 15-mil sheath thicknesses) using a 75% smeared density element require a fuel alloy having a Pu content of 22-28 wt% and a U-235 enrichment of 55-75%; going to an 80% smeared density element improves the situation only slightly. With the higher Pu content and/or lower Zr content in the fuel alloy, comes a degradation of the alloy solidus temperature (the Zr sheath and fuel alloy compositional changes will also affect the thermal conductance of the fuel element, although preliminary calculations indicate temperature changes should be small).

A Zr sheath can be incorporated into the PRISM fuel element designs with significantly less impact on the fuel alloy composition. A 10 wt% Zr fuel alloy can accommodate 10- to 20-mil sheaths with Pu compositions of 24 to 28 wt% with no uranium enrichment; if 6 wt% Zr fuel alloy is used only 22 to 26 wt% Pu

is required. Since EBR-II is only a research reactor, it seems more important to demonstrate the Zr-sheathed fuel fabrication method and its fuel performance for the fuel system more prototypic of a commercial IFR (i.e., PRISM). This turns out to be easier to do for PRISM fuel than for EBR-II fuel.

B. Compatibility of IFR Fuel and Austenitic Stainless Steel

1. Introduction

The IFR utilizes a U-Pu-Zr metallic fuel clad in stainless steel,[8] the reference design using a martensitic stainless steel. Once irradiated, this U-Pu-Zr fuel swells and contacts the cladding. As a result, fuel, cladding, and fission product elements interdiffuse and form potentially low melting intermetallic phases that may affect the structural integrity of the cladding. This fuel/cladding chemical interaction (FCCI) has been investigated using systematic diffusion couple techniques[9-13] that focus on interactions between cladding (both martensitic and austenitic), U-Zr fuel, U-Pu-Zr fuel, and Ce (a major fission product). This report will compare the results from interdiffusion experiments using D9* cladding, an austenitic steel, with those from recent melting tests using U-19Pu-10Zr fuel and 15-15 Ti stainless steel,** a steel close in composition to D9.[14] Additionally, it will compare the results from diffusion tests and melting tests with those from recent analyses of actual irradiated IFR specimens clad with D9. In the light of available phase diagrams, this report will discuss the particular phases that can be expected to form in an irradiated D9 clad IFR fuel element in the context of FCCI, with comments on the relative diffusion behavior of the fuel, cladding, and fission product elements. While the D9 is an austenitic stainless steel, and the reference cladding is martensitic, the comparisons offer insights into the difference in

*D9 is austenitic stainless steel with a nominal composition in wt% of:
13.45Cr, 15.61Ni, 1.65Mo, 0.32Ti, 0.01V, 0.01W, 2.05Mn, 0.85Si, 0.034C,
0.0051N, 0.00620, bal. Fe.

**15-15Ti has the following ranges in composition in wt%: 0.08-0.12C, 14.5-15.5Cr, 14.5-15.5Ni, 1.0-1.4Mo, 0.3-0.55Si, 1.5-2.0Mn, 0.04-0.06B, 0.3-0.55Ti, 0.05-0.1(Co+Ta), 0.015Cu, 0.015P, 0.015S, 0.05V, 0.01Ca, 0.01N, bal. Fe.

behavior with and without a significant amount of Ni and other factors. Understanding the interaction allows optimization of performance.

2. Main Observations from Diffusion Couple Studies

Keiser and Dayananda[9,10] investigated the interdiffusion behavior of U-23Zr* fuel bonded with Fe, Ni, various alloys of the Fe, Ni, Cr system, and the cladding materials HT9 (a martensitic steel with a nominal composition in wt% of 12.02Cr, 0.57Ni, 1.03Mo, 0.34V, 0.51W, 0.50Mn, 0.22Si, 0.21C, bal Fe), D9, and 316 (an austenitic steel with a nominal composition in wt% of 16.30Cr, 10.50Ni, 2.11Mo, 1.47Mn, 0.67Si, 0.05C, bal. Fe). They noted that the presence of Ni had a drastic effect on the diffusion structure size and morphology and the relative interdiffusion behavior of the elements in diffusion couples annealed at 700°C. The D9 couple had a relatively large diffusion structure with many phases. The U-23Zr versus D9 couple had the following intermetallics based on point-by-point SEM-EDS analysis: σ (an approximate composition of 50Fe-50Cr), U(Fe,Ni,Cr)_2 , $\text{U}_6\text{(Fe,Ni,Cr)}$, high-Zr precipitates, $\text{(Zr,U)(Fe,Ni,Cr)}_2$, $\text{(Zr,U)}_2\text{(Fe,Ni,Cr)}$, and a (U,Zr) matrix with up to 6 at.% Fe. The intermetallic designations of the phases, based on binary phase diagrams,[11] are based on compositional analysis by SEM-EDS. The designation of a phase corresponds to the ratio of the sum of the cladding elements (Fe+Ni+Cr) to the sum of the fuel elements (U+Zr). The fuel and cladding elements are listed in parentheses in decreasing order of concentration. All phases will be designated in this fashion throughout this paper. Table II.1 enumerates the typical composition ranges for the various phases observed in U-23Zr versus D9 couple.

Interdiffusion experiments were also conducted at 650°C using U-22Pu-23Zr fuel and D9 cladding.[12] The U-22Pu-23Zr versus D9 couple exhibited the development of five distinct phases. These phases included: $\text{(U,Pu)(Fe,Ni,Cr)}_2$, $\text{(Zr,U)(Fe,Cr,Ni)}_2$, (Zr,U)(Fe,Cr,Ni) , (U,Pu,Zr) matrix, and $\text{(Zr,U,Pu)}_2\text{(Fe,Ni)}$. The typical compositions for these phases are presented in Table II.2. One common observation noted for both the U-23Zr versus D9 and U-22Pu-23Zr versus D9 couple is that, of the fuel elements, U forms intermetallics with Fe on the cladding side of the diffusion structure and Zr

*All compositions will be in atomic percent unless otherwise noted.

TABLE II.1. Composition Ranges for the Phases Observed in the U-23Zr vs. D9 Couple (at.%)

Phase	Fe	Ni	Cr	U	Zr
σ	56	6	35	2	0
$U(Fe,Ni,Cr)_2$	33-42	10-17	12-22	28-34	0
$U_6(Fe,Ni)$	14	4	1	80	1
Zr precipitates	2	2	1	14	80
two $(Zr,U)(Fe,Ni,Cr)_2$	27	4	35	5	27
phases	8	42	6	4	36
two $(Zr,U)_2(Fe,Ni,Cr)$	13	22	1	2	59
phases	13-31	4-24	0	23-25	38-39
(U,Zr) matrix	6	2	1	72	17

TABLE II.2. Typical Compositions of the Phases Detected in the U-22Pu-23Zr vs. D9 Couple (at.%)

Phase	Fe	Ni	Cr	U	Pu	Zr
$(U,Pu,Zr)(Fe,Cr,Ni)_2$	49	7	13	24	3	4
(U,Pu,Zr) Matrix	5	1	0	70	18	7
$(Zr,U)(Fe,Cr,Ni)_2$	37	3	25	2	0	32
$(Zr,U)(Fe,Cr,Ni)$	36	3	15	14	0.6	32
$(Zr,U,Pu)_2(Fe,Ni)$	21	18	0	18	3	40

forms intermetallics with Fe on the fuel side of the diffusion structure. This reflects the fact that U diffuses faster towards the cladding than the other fuel constituents, and Fe diffuses faster towards the fuel than the other cladding constituents.

In order to investigate lanthanide fission product interactions with cladding materials, Tortorici and Dayananda performed isothermal furnace anneals using Ce (a major fission product in IFR fuels) and Fe, Ni, selected alloys in the Fe-Ni-Cr system,[13] and selected cladding materials.[14] The Ce versus D9

couple developed the three distinct phases Ce(Fe,Ni,Cr)_3 , Ce(Fe,Ni)_2 , and $\text{Ce}_7(\text{Ni,Fe})_3$. The compositions of these phases are presented in Table II.3. The $\text{Ce}_7(\text{Ni,Fe})_3$ was consistently found in all of the Ni-containing couples on the Ce side of the diffusion structure. In Ce versus austenitic steel couples, Ni diffuses faster than Fe.

3. Melting Tests

Recently, Sari et al., [15] reported results from isothermal melting tests. These tests investigated the melting temperatures that would be observed if various alloys containing U, Pu, Zr, minor actinides, or rare earths were sandwiched between two 15/15 Ti pins, placed in a dilatometer, heated to temperatures at which large dimensional changes occurred, and then cooled to room temperature. Supposedly, the dilatometric analysis enabled one to detect minor phase transformations and to determine the onset of melting at the interface of an alloy-steel diffusion couple. The heat treatment was repeated several times for each sample to check the accuracy of the test as a function of the heating rate and of the state of the surfaces in contact. The specific alloys that were sandwiched between the 15/15 Ti steel pins included (in wt.%, see Table II.4 for at.%): U-19Pu-10Zr, U-19Pu-10Zr-2Am-3Np-3.6Nd-1.2Ce-0.2Y, Nd-16Ce-4Y-8Pu-20Am, and Nd-10Ce-10Gd-10Y. All alloys were cast by arc melting. The U-Pu-Zr alloys containing minor actinides and rare earths dissolved Am and Np but not the rare earth elements. Rare earth alloys were able to dissolve Am and Pu. Those alloys that consisted of U, Pu, Zr, minor actinides and rare earths were two-phase with a U-Pu-Zr-Np matrix and Am-Pu-Rare Earth (RE) precipitates. These investigators reported seeing noticeable interaction between the various alloys and the 15/15

TABLE II.3. Typical Compositions of the Phases Detected in the Ce vs. D9 Couple (at.%)

Phase	Fe	Ni	Cr	Ce
Ce(Fe,Ni,Cr)_3	50	4	18	28
Ce(Fe,Ni)_2	63	3	0	36
$\text{Ce}_7(\text{Ni,Fe})_3$	1	29	0	70

TABLE II.4. The Compositions of the Alloys Coupled with 15/15 Ti, the Experimental Conditions and the Results for Sari et al., Melting Tests

Couple	Alloy Compositions (at.%)	Cycle	Heating Rate (K/min)	Temperature (°C)	
				Start of Reaction	Melting
CR13/3 Steel	$U_{61}Pu_{16}Zr_{23}$ 15/15 Ti	1	2	817	820
		1a	2	717	740
		2	7	702	747
		3	5	702	
CR13/3H2 Steel	$U_{61}Pu_{16}Zr_{23}$ 15/15Ti	1	2	822	827
		2	7	707	727
		3	2	697	722
CR12/3 Steel	$U_{51}Pu_{15.6}Zr_{22}Am_{1.6}Np_{2.5}Nd_5Ce_{1.7}Y_{0.5}$ 15/15Ti	1	2	697	792
		2	2	697	727
		3	7	697	737
CR12/3H2 Steel	$U_{51}Pu_{15.6}Zr_{22}Am_{1.6}Np_{2.5}Nd_5Ce_{1.7}Y_{0.5}$ 15/15Ti	1	2	725	747
		1a	2	687	727
		2	2	627	697
		3	5	627	712
		4	7	657	717
		5	7	657	727
		6	7	647	712
REMAAL Steel	$Nd_{55.7}Ce_{17.6}Y_9Pu_{5.1}Am_{12.7}$ 15/15Ti	1	7	687	697
		2	2	597	625
REAL Steel	$Nd_{66.2}Ce_{9.7}Gd_{8.7}Y_{15.3}$ 15/15Ti	1	7	717	732
		2	1	607	625
		3	2	625	630
		4	2	625	630
		5	2	597	617

Ti steel using electron probe microanalysis and metallographic examination, and visually they observed collapse of the samples due to the formation of low melting reaction products.

The intimacy of contact of the samples, the sample cleanliness, and the conditions of the sample surfaces are all unknown for these tests, but interdiffusion did occur and phases did form. These tests were conducted to promote diffusion and to determine when particular phases would melt, not to determine the diffusion kinetics of the process. So, in this respect these tests are noteworthy. Table II.4 shows the experimental conditions for the tests, the atomic percent composition of the alloys, and the temperature at which the specimens lost their integrity and collapsed due to the formation of low melting reaction products. The melting temperatures are reported to be accurate to

within $\pm 10^\circ$. Four different types of couples were run for this investigation, as shown in Table II.4, namely the CR13-steel, CR12-steel, REMAAL-Steel, and REAL-steel couples.

The following will describe the types of phases that developed for each of these couples.

a. CR13-steel Couple

The CR13-steel couple reportedly developed three multiphase regions. The compositions of the various phases are listed in Table II.5. By converting the wt% compositions reported by Sari et al., for the various phases to ones in at.%, it is observed: Region 1 contained the three phases $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(U,Pu,Zr)_6(Fe,Cr,Ni)$; Region 2 contained the four phases $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(U,Pu,Zr)_6Fe$, $(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(Zr,U,Pu)_2(Fe,Ni)$; and Region 3 consisted of the three phases $(U,Pu,Zr)_6Fe$, $(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(Zr,U,Pu)_2(Fe,Ni)$. Hence, this couple developed four distinct types of phases viz. $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(U,Pu,Zr)_6Fe$, $(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(Zr,U,Pu)_2(Fe,Ni)$.

b. CR12-steel Couple

The CR12-steel couple developed phases similar in concentration to those observed for the CR13-steel couple. Six distinct phases were detected in this couple, and they included: $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(U,Pu)_6Fe$ -with 2 at.% Np, $(Zr,U)(Fe,Cr,Ni)_2$, $(Zr,U)(Fe,Cr)$, $(Zr,U,Pu)_2(Fe,Ni)$, and $(Ce,Nd,Am,Pu,U)(Ni,Fe,Cr)_3$. The phase compositions are listed in Table II.6.

c. REMAAL-steel Couple

The REMAAL (Nd-Ce-Am-Y-Pu alloy) versus steel couple formed four phases. The phase compositions are listed in Table II.7. Heading from the cladding towards the fuel, a $(Nd,Ce,Pu,Am,Y)Fe_2$ phase developed followed by a $(Nd,Ce,Am,Y)_3Ni$ phase, which contained 42 at.% Nd and 18 at.% Ce, and two phases that were composed of Ce, Nd, Am, and Y. Pu was only detected in the $(Nd,Ce,Pu,Am,Y)Fe_2$ phase, with a concentration of 16 at.%. This amount is higher than the 5 at.% Pu in the original REMAAL alloy.

TABLE II.5. Composition of the Types of Phases Detected in the CR13-steel Couple (at.%)

Constituent	Region 1			Region 2				Region 3		
	UFe ₂	Unknown	ZrFe ₂	UFe ₂	U ₆ Fe	ZrFe ₂	ZrFe	U ₆ Fe	ZrFe ₂	Zr ₂
Cr	11.48	3.99	16.14	8.69	0.00	19.63	0.00	0.00	19.61	0.21
Fe	47.87	28.18	44.62	50.78	14.73	42.62	31.28	14.68	42.15	23.79
Ni	5.55	2.06	4.71	6.76	0.36	4.71	12.89	0.00	2.54	14.44
Zr	7.62	2.65	17.79	4.84	0.23	19.73	28.45	1.15	31.18	38.78
U	22.76	43.42	15.38	26.40	75.01	12.28	23.52	75.47	4.22	20.39
Pu	4.72	19.69	1.36	2.53	9.67	1.03	3.86	8.70	0.31	2.38

TABLE II.6. Composition of the Types of Phases Detected in the CR12-steel Couple (at.%)

Constituent	(U, Pu, Zr) (Fe, Cr, Ni) ₂	(U, Pu, Np) ₆ Fe	(Pu, Nd, Ce, Am) (Ni, Fe)	(Zr, U) (Fe, Cr, Ni) ₂	(Zr, U) (Fe, Cr)	(Zr, U) ₂ (Fe, Ni)
Cr	12.32	0.81	1.00	11.94	7.08	0.46
Fe	51.86	13.54	4.20	49.81	48.89	25.15
Ni	4.61	0.72	45.64	2.14	0.96	11.15
Zr	7.8	0.92	0.71	32.18	27.01	35.74
Ce	0.71	0.00	5.20	0.05	0.00	0.08
Nd	0.21	0.00	12.90	0.00	0.00	0.00
U	19.31	68.96	1.53	3.32	15.43	23.94
Np	0.42	2.92	0.22	0.16	0.12	0.65
Pu	2.63	12.13	18.39	0.39	0.50	2.68
Am	0.12	0.00	10.22	0.00	0.00	0.15

TABLE II.7. Composition of the Types of Phases Detected in the REMAAL-steel Couple (at.%)

Constituent	(Pu, Am, Ce, Nd) (Fe, Ni, Mn, Cr) ₂	(Ne, Ce, Am) ₃ (Ni, Fe)	(Nd, Ce, Am, Y)	(Nd, Ce, Am, Y, Ni)
Cr	1.94	0.49	0.29	0.56
Mn	1.43	0.47	0.55	0.00
Fe	59.97	2.29	0.27	0.26
Ni	0.57	22.46	0.51	3.50
Y	3.66	5.18	5.07	1.49
Ce	4.25	18.09	23.05	35.72
Nd	3.04	41.37	58.96	49.77
Pu	16.25	0.47	1.11	0.36
Am	9.89	9.17	10.20	8.33

d. REAL-steel Couple

The REAL (Nd-Ce-Gd-Y alloy) versus steel couple had a diffusion structure very similar to the one described for the REMAAL-steel couple, except that no (Nd,Ce,Pu,Am,Y)Fe₂ phase developed. The reported phases for this couple include: (Nd,Ce,Y,Gd)₃(Ni,Fe)-consisting of 50 at.% Nd, two lanthanide-rich (Nd,Ce,Y,Gd) phases, (Nd,Ce,Y,Gd)(Ni,Fe)₅, (Nd,Ce,Y,Gd)(Fe,Ni,Mn), and (Nd,Ce,Y,Gd)Fe₂.

In terms of the melting results where heating rates were varied along with the number of annealing cycles, Sari et al., report little difference in melting temperatures for the CR13-steel or CR12-steel couples (see Table II.4). The lowest melting temperature observed for either couple was near 720°C. On the other hand, the REMAAL-steel and REAL-steel couples, which contained no U or Zr, melted at temperatures nearer 625°C.

4. As-irradiated Results

A U-16Pu-23Zr fuel element with D9 cladding irradiated to 11.3 at.% burnup, where burnup is the amount of heavy metals fissioned, has been analyzed using an electron microprobe.[16] Particular attention was given to the development of phase layers at the fuel-cladding interface. The major observations from this study were: (1) Pu is found in high concentrations in a phase layer closest to the unreacted cladding; (2) Fe and Ni penetrate into the fuel to a maximum depth of 175 mm; (3) Ni-Zr phases are observed in the diffusion structure; (4) of the lanthanide-series fission products, Ce and Nd are found in the highest amounts in all regions of the diffusion structure; (5) a 40-mm-thick phase layer develops along the fuel-cladding interface, and it contains Ce, Nd, La, and some Cr; (6) lanthanides are not found in high concentrations in the phase nearest the cladding; (7) Fe seems to form phases with U and Pu towards the fuel and primarily with Pu nearest the unreacted cladding; and (8) negligible Ni is observed in the phase nearest the unreacted cladding.

Another electron microprobe analysis of an irradiated U-16Pu-23Zr fuel element with D9 cladding has revealed the development of two different

lanthanide-containing phases near the fuel-cladding interface.[17] These phases contain the elements U, Pu, Zr, Nd, Ce, La, Pr, and Pd. The compositions for "Phase 1" and "Phase 2" in atomic percent are 1.15U-1.91Pu-0.3Zr-20.22Pd-23.67La-17.6Ce-3.79Pr-31.35Nd and 3.81Pu-28.94La-29.21Ce-3.91Pr-34.12Nd, respectively. This demonstrates that lanthanide-rich phases will develop near the cladding in irradiated fuel elements.

Related to melting temperatures in irradiated fuel elements, the first signs of liquefaction will depend on many factors, e.g., burnup, linear power, etc., and one of the largest contributing factors is the interdiffusion of lanthanides and cladding elements to form low melting phases. For an irradiated fuel element with D9 cladding, the lowest observed melting temperature was 675°C.[18] In this element, which was irradiated to a high burn-up resulting in the generation of a large amount of fission products, the first signs of melting have been observed along the fuel-cladding interface where lanthanide-cladding element phases developed.

5. Discussion

a. Phase Development

If one compares the various types of phases that develop in these reported investigations, it becomes clear that the relative diffusion behavior in the various D9 couples is similar. Firstly, by comparing the U-23Zr versus D9 couple with the U-22Pu-23Zr versus D9 couple, as presented in Tables II.1 and II.2, one can conclude that even with the presence of Pu no drastic changes occur in the types of phases that will form. With the presence of Pu, the major phases that will develop in the diffusion structure are (U,Pu)(Fe,Ni,Cr)₂, (Zr,U)(Fe,Cr,Ni)₂, (Zr,U)(Fe,Cr,Ni), (U,Pu,Zr) matrix, and (Zr,U,Pu)₂(Fe,Ni). This result has been corroborated by Sari et al., who report the same types of phases for the CR13-steel couple, as shown in Table II.5. In the Sari et al. CR12-steel couple (see Table II.6) where not only fuel elements are present but also lanthanides and minor actinides, the same types of phases develop as discussed previously. A (Pu,Nd,Ce,Am)(Ni,Fe) phase is the only phase that seems to develop due to the presence of lanthanides and minor actinides. This phase could possibly be predicted to develop based on the Pu-Ni, Ce-Ni, and

Nd-Ni binary phase diagrams, where the only common intermetallic with Ni on the Pu, Ce, and Nd side of the phase diagram is one with a 50-50 ratio. Furthermore, the development of this Pu-lanthanide-Ni intermetallic in the CR12-steel couple demonstrates that Ni plays an increased role in determining what phases will form in this couple compared to the U-23Zr versus D9 or U-16Pu-23Zr versus D9 couples where Fe played the most important role.

When the elements U and Zr are not present in diffusion couples with Pu, lanthanides, minor actinides, and D9, completely new types of phases develop based on the REMAAL-steel and REAL-steel couples reported by Sari et al, as presented in Tables II.7 and II.8. When Pu is present in a couple, it is only observed in notable concentrations in the $(\text{Pu}, \text{Am}, \text{Ce}, \text{Nd})(\text{Fe}, \text{Ni}, \text{Mn}, \text{Cr})_2$ phase closest to the unreacted cladding. Based on the Pu-Fe and Am-Fe phase diagrams, the two major elements in this phase (viz., Pu and Am) will form PuFe_2 and AmFe_2 , respectively, when combined with Fe. Nd, which exists in larger concentrations than Ce, and Ni dictate the next phase to develop towards the lanthanide-minor actinide alloy. According to the Nd-Ni and Ce-Ni phase diagrams a Ce_7Ni_3 and Nd_7Ni_3 phase are possible intermetallics for these elements with Ni, and only Nd will form the Nd_3Ni type phase. Yet, the Nd_3Ni type phase is the one observed to form in the REMAAL-steel couple, probably due to the fact that Nd exists in higher concentrations in the original alloy. In the Ce versus D9 couple reported by Tortorici and Dayananda (see Table II.3) a Ce_7Ni_3 type of phase does in fact form. So it is clear that without Nd present it is likely for this intermetallic

TABLE II.8. Composition of the Types of Phases Detected in the REAL-steel Couple (at.%)

Constituent	(Nd, Ce, Y, Gd) Fe_2	(Nd, Ce, Y, Gd) (Fe, Mn, Ni)	Nd, Ce, Y, Gd_5 (Ni, Fe)	(Nd, Ce, Y, Gd)	(Nd, Ce, Y, Gd)	$(\text{Nd, Ce, Y, Gd})_3$ (Ni, Fe)
Cr	0.90	0.18	0.00	0.00	0.00	0.00
Mn	0.99	7.80	0.23	0.25	0.26	0.00
Fe	63.00	38.52	5.54	0.25	0.25	3.18
Ni	0.93	3.73	10.33	0.00	0.00	20.93
Y	16.10	8.13	12.80	9.69	4.75	10.63
Ce	2.66	14.12	9.18	12.30	21.49	8.85
Nd	9.39	23.41	55.16	69.29	69.58	49.65
Gd	6.03	4.11	6.76	8.22	3.67	6.76

to form. Comparing the relative diffusion behavior of the Ni and Fe, it seems that even though D9 contains less Ni than Fe, the Ni is diffusing faster than Fe because Ni intermetallics are found closer to the REMAAL alloy side of the diffusion zone. This is consistent with Tortorici and Dayananda who report faster diffusion for Ni and the resulting formation of the $\text{Ce}_7(\text{Ni,Fe})_3$ intermetallic on the Ce side of the diffusion structure. Therefore, Ni dictates the types of intermetallics that will form on the lanthanide alloy side of a lanthanide alloy versus D9 cladding diffusion couple.

In reference to the effect of Pu concentrations on intermetallic formation, the REAL-steel couple, which does not contain Pu, does develop a $(\text{Nd,Ce,Y,Gd})\text{Fe}_2$ phase that was also observed in the REMAAL-steel couple. Yet, overall, more types of phases develop in the non-Pu-containing REAL-steel couple than the REMAAL-steel couple. This suggests that the presence of Pu will reduce the number of observed phases in a diffusion couple that contains lanthanides and minor actinides. In both couples, lanthanide-rich phases containing Nd, Ce, and Y have been observed, thereby shedding light on what lanthanide-rich phases are likely to form when Nd and Ce are present.

Based on electron microprobe analysis results, irradiated IFR fuel elements with D9 cladding exhibit similar interdiffusion behavior and phase development as compared to the diffusion couple experiments and melting tests discussed in this paper. A Pu-rich phase, probably $(\text{Pu,Nd,Cr})\text{Fe}_2$, seems to form nearest the unreacted cladding in the irradiated fuel element, and this phase has negligible Ni. Then, moving towards the fuel, the presence of the fission products Nd and Ce have been identified along with the cladding element Ni, thereby suggesting the possible formation of a $(\text{Pu,Nd,Ce})(\text{Ni,Fe})$ phase. Continuing to move towards the fuel various intermetallics between fuel elements and cladding elements may form. These can include all or some of the following intermetallics: $(\text{U,Pu,Zr})(\text{Fe,Cr,Ni})_2$, $(\text{U,Pu})_6\text{Fe}$, $(\text{Zr,U})(\text{Fe,Cr,Ni})_2$, $(\text{Zr,U})(\text{Fe,Cr})$, and $(\text{Zr,U})_2(\text{Fe,Ni})$. In addition, phases consisting almost entirely of lanthanides have been identified to exist by Cohen and Sanecki. The lanthanide-rich phases observed in Sari et al. REMAAL-steel and REAL-steel couples had similar ratios of Ce and Nd compared to those observed in an irradiated fuel element. This suggests that since lanthanides have mutual solubility and little solubility in the (U, Pu, Zr) matrix they will form

lanthanide-rich phases in a fuel element. Also, in cases where lanthanides migrate through the fuel and form lanthanide-rich phases near the cladding, intermetallics may develop due to interdiffusion of lanthanides and cladding elements. In this type of circumstance, when U and Zr are not present, a NdNi_3 type of intermetallic may form based on the REMAAL-steel couple.

b. Phase Liquefaction

The fuel components U, Pu, and Zr dictate what phases will form at the fuel/cladding interface, even with the presence of minor amounts of actinides and lanthanides, based on the CR12-steel couple. In this couple, the same phases develop as for the CR13-steel couple and the U-22Pu-23Zr versus D9 couple annealed at Argonne (compare Tables II.2, II.5, and II.6), except for the $(\text{Pu}, \text{Nd}, \text{Ce}, \text{Am})(\text{Ni}, \text{Fe})$ phase. Even with formation of this phase, the CR12-steel couple melted at temperatures comparable to what is observed for couples between U-Pu-Zr fuel and austenitic cladding, namely 720-730°C. Based on binary phase diagrams, the CeNi, PuNi, and NdNi intermetallics will melt at 680°C, 800°C and 780°C, respectively. The concentration of Ce is only 5.2 at.% in the $(\text{Pu}, \text{Nd}, \text{Ce})(\text{Ni}, \text{Fe})$ phase while the concentrations of Nd and Pu are 12.9 and 18.4 at.%, respectively. Therefore based on the results of the CR12-steel couple, this phase may have melted near 730°C along with the eutectic combination of $(\text{U}, \text{Pu}, \text{Zr})(\text{Fe}, \text{Cr}, \text{Ni})_2$ and $(\text{U}, \text{Pu}, \text{Np})_6\text{Fe}$ which can melt at 725°C according to the U-Fe phase diagram.

The REMAAL-steel and REAL-steel couples display lower melting temperatures than the CR12-steel and CR13-steel couples, as shown in Table II.4. Melting was observed in these couples at around 620-630°C. The presence of Pu dictated what phase would form nearest the unreacted cladding, viz. $(\text{Pu}, \text{Am}, \text{Ce}, \text{Nd})\text{Fe}_2$. The binary PuFe_2 intermetallic melts at around 1240°C. Whereas the presence of Nd caused the $(\text{Nd}, \text{Ce}, \text{Am})_3\text{Ni}$ phase to form. The binary Nd_3Ni intermetallic melts at around 600°C, and a eutectic develops between Nd and Nd_3Ni that exhibits melting at around 570°C. Therefore, it is possible that the appearance of an Nd_3Ni type of phase contributed to the overall liquefaction of the diffusion structures in these couples.

For actual IFR fuel elements with D9 cladding irradiated to high burn-ups, melting has been observed near 675°C, and based on the diffusion couple results it may be due to eutectic melting that develops between lanthanide-rich phases and the Nd_3Ni type of multicomponent phase. In the cases where only small amounts of minor actinides and lanthanides are available in the fuel, the melting behavior of the irradiated fuel will be close to that of the CR12-steel couple where melting was seen near 725°C. On the other hand, when large lanthanide-rich phases develop adjacent to the cladding, which can be the case for fuel elements irradiated to high burn-ups, a eutectic can develop with the Nd_3Ni type of multicomponent phase. This eutectic may be where melting first occurs locally at temperatures near 675°C.

6. Conclusions

The following are the main conclusions of this report on compatibility of fuel and cladding in IFR fuels with austenitic stainless steel cladding:

1. U-Pu-Zr fuel alloys coupled with D9 cladding develop five major phases, namely $(\text{U,Pu,Zr})(\text{Fe,Cr,Ni})_2$, $(\text{Zr,U})(\text{Fe,Cr,Ni})_2$, (U,Pu,Zr) matrix, $(\text{Zr,U})(\text{Fe,Cr})$, and $(\text{Zr,U})_2(\text{Fe,Ni})$, and the presence of lanthanides and minor actinides adds a new phase, $(\text{Pu,Nd,Ce,Am})(\text{Ni,Fe})$.
2. Lanthanide-actinide alloys without U and Zr coupled with D9 exhibit a $(\text{lanthanide})\text{Fe}_2$ type of phase, a Nd_3Ni type of phase, and other lanthanide-rich phases "or" containing actinides. When Pu is not present in the couple, additional types of phases such as NdNi and Nd_5Ni develop.
3. The presence of Ni, not Fe, in fuel-D9 cladding couples dictates the types of phases that form on the lanthanide-actinide alloy side of the diffusion structure.
4. A $(\text{Pu,Nd,Ce})\text{Ni}$ type of phase forms in couples between D9 cladding and alloys with U, Pu, Zr, and small amounts of

lanthanides and minor actinides. The development of this phase does not lower the expected 725°C melting temperature based on the U-Fe phase diagram.

5. When lanthanide-actinide alloys without U and Zr are bonded to D9 cladding, a (Pu,Nd,Ce)Fe₂ and Nd₃Ni type of phase develops, along with other lanthanide-rich phases containing actinides. Based on the binary Nd-Ni phase diagram, the appearance of the Nd₃Ni type of phase adjacent to a lanthanide-rich phase can initiate eutectic melting at temperatures near 600°C.
6. Irradiated IFR fuels with D9 cladding that do not develop large lanthanide-rich phases at the fuel-cladding interface will exhibit evidence of melting at around 725°C. Whereas, in cases where lanthanide-rich phases do develop near the cladding, a eutectic can develop with the Nd₃Ni type of multicomponent phase that will initiate melting at lower temperatures. Those fuel element locations where this eutectic develops may be the locations where liquid phases first develop in fuel elements that show signs of liquefaction at temperatures near 675°C.

7. Future Work

Further investigation of FCCI is required for IFR fuel clad with martensitic stainless steels that do not contain Ni, since these types of steels are being considered for use in IFR fuel elements. The presence of Ni has been found to drastically affect the types of intermetallics observed at the fuel-cladding interface, and it is imperative to better understand phase formation when Ni is not present in the cladding. This work should involve experiments like those conducted by Sari et al., using U-Pu-Zr alloys with lanthanides and minor actinides. Furthermore, these alloys should deploy other fission product elements, like Pd, that may play a role in determining the intermetallic phases that form at the fuel-cladding interface.

C. Results of Compatibility Testing in Mk-V Fuels

This report summarizes the results and conclusions from the fuel/cladding compatibility studies of Mk-V (U-20Pu-10Zr/HT9) fuels. More in-depth descriptions and discussions of the tested samples and results are available.[18-20]

The study focused on the effect of reactor operating variables on the compatibility of the fuel and cladding. It included analyses of steady-state (as-irradiated) samples as well as post-FBTA sample; FBTA testing extends the information about fuel/cladding interaction to temperatures above those normally seen in the reactor. The reactor operating variables evaluated were burnup, peak inner cladding temperature (PICT), and linear heat rating (LHR) or power.

Most of the samples for compatibility testing came from three elements irradiated as part of the DP-1 experiment in subassemblies X441/X441A. All three elements had U-19Pu-10Zr fuel and HT9 cladding. The experiment was irradiated at PICT of 600°C and LHR of 15 kW/ft at Beginning-of-Life (BOL). These parameters bound the Mk-V operating envelope. To study the burnup effect, two different burnup regimes were evaluated. DP-17, DP-21, and DP-16 achieved burnups of 5.7, 11.4, and 11.5%, respectively. To study the cladding temperature effect, samples taken at different axial elevations from the same element were examined. Because EBR-II has a relatively flat axial power profile, element T-608 (also with U-19Pu-10Zr fuel and HT9 cladding) from subassembly X429B was included to study the effect of linear power on compatibility. Subassembly X429B had a PICT of about 590°C and an LHR of about 13 kW/ft at BOL. Element T-608 achieved a maximum burnup of 13.4 at.%.

Table II.9 summarizes the examination results from the steady-state samples. The reactor operating parameters are interpolated from the LIFE-METAL calculations for the given element; the PICT and LHR are burnup averaged. The wastage value listed represents the maximum amount of penetration by fuel constituents into the cladding at a given axial elevation. The wastage is not circumferentially uniform and only the maximum value, which is the most conservative for design and evaluation purposes, is reported.

TABLE II.9. Steady-state (as-irradiated) Results

Element No.	X/L	Sample BU (at.%)	LHR (kW/ft)	PICT (°C)	Cladding Wastage (μm)
Low BU Sample					
DP-17	0.87	4.6	12.8	561	28
DP-17	0.78	4.9	13.9	551	33
DP-17	0.13	5.1	14.3	439	0
DP-17	0.53	5.5	15.5	514	0
Higher BU Sample					
DP-16	0.22	10.9	14.6	451	0
DP-21	0.50	11.4	15.2	505	4
T-608	0.78	12.4	10.7	530	32
DP-16	0.78	10.3	13.7	536	31
DP-21	0.78	10.2	13.5	546	57
DP-21	0.90	9.2	10.9	560	37

Table II.10 presents results from the elevated temperature testing in the FBTA. Three figures of merit are reported: total cladding wastage, transient cladding wastage, and eutectic formation percentage. Similar to the steady-state wastage presented in Table II.9, the total cladding wastage represents the maximum amount of penetration seen in the cladding after the elevated temperature test. By interpolation of the data on sibling samples, a steady-state wastage value was calculated and subtracted from the total wastage to yield the transient wastage. The transient cladding wastage then approximates the amount of interaction that occurred as a result of the elevated temperature exposure. Eutectic formation (a.k.a., liquefaction) percentage is a measure of the areal fraction of the fuel at the periphery that has undergone a phase transformation during the test due to the interaction with the cladding. Eutectic formation occurs as iron from the cladding creates an alloy with uranium and plutonium from the fuel that is above its solidus at FBTA test temperatures; the depth of the fuel affected is a direct measure of the depth of iron penetration into the fuel. The lowest test temperature at which eutectic formation can be seen is referred to as the threshold temperature for eutectic formation.

TABLE II.10. Elevated Temperature Test Results

Steady-state Irradiation					Ex-reactor Testing				
Element No.	X/L	Sample BU (at.%)	LHR (kW/ft)	PICT (°C)	Test Temp. (°C)	Test Time (h)	Cladding Total (μm)	Wastage Transient (μm)	Eutectic Formation (Areal %)
Effect of Test Temperature on Low BU Samples									
DP-17	0.57	5.5	15.4	520	740	1.0	0	0	0
DP-17	0.76	5.0	14.1	548	750	1.0	37	4	16
DP-17	0.39	5.6	15.7	489	770	1.0	27	27	10
DP-17	0.55	5.5	15.4	517	800	1.0	114	114	34
Effect of Test Time on Low BU Samples									
DP-17	0.42	5.6	15.6	494	800	0.1	18	18	9
DP-17	0.45	5.6	15.6	501	800	0.5	78	78	33
DP-17	0.55	5.6	15.4	517	800	1.0	114	114	34
DP-17	0.50	5.6	15.6	510	800	2.0	142	142	26
DP-17	0.48	5.6	15.6	505	725	7.0	74	74	4

TABLE II.10. (Contd.)

Steady-state Irradiation					Ex-reactor Testing				
Element No.	X/L	Sample BU (at.%)	LHR (kW/ft)	PICT (°C)	Test Temp. (°C)	Test Time (h)	Cladding Total (μm)	Wastage Transient (μm)	Eutectic Formation (Areal %)
Effect of Test Temperature on Higher BU Samples (1-h Tests)									
DP-16	0.69	10.7	14.2	525	625	1.0	34	1	0
DP-16	0.75	10.4	13.8	533	650	1.0	46	10	0
DP-21	0.88	9.3	12.4	559	650	1.0	29	0	0
DP-16	0.72	10.5	14.0	528	675	1.0	59	24	5
DP-16	0.83	9.7	13.0	540	700	1.0	58	21	9
DP-21	0.83	9.8	13.0	554	700	1.0	76	23	13
DP-21	0.89	9.2	12.3	560	740	1.0	87	46	18
DP-21	0.85	9.6	12.8	556	770	1.0	80	31	66
DP-21	0.80	10.0	13.3	550	800	1.0	100	44	100
Effect of Test Temperature on Higher BU Samples (12-h Tests)									
DP-16	0.61	11.1	14.8	515	650	12.0	36	12	0
DP-16	0.64	10.9	14.6	517	660	12.0	81	53	12
DP-16	0.67	10.8	14.4	521	675	12.0	140	109	31

TABLE II.10. (Contd.)

Steady-state Irradiation					Ex-reactor Testing				
Element No.	X/L	Sample BU (at.%)	LHR (kW/ft)	PICT (°C)	Test Temp. (°C)	Test Time (h)	Cladding Total (μm)	Wastage Transient (μm)	Eutectic Formation (Areal %)
Tests on Samples Irradiated at Lower LHR Than DP-21/DP-16									
T-608	0.76	12.6	10.8	527	675	1.0	16	0	0
T-608	0.71	13.0	11.2	521	700	1.0	53	22	0
T-608	0.73	12.8	11.1	524	725	1.0	32	0	2
T-608	0.69	13.1	11.3	518	660	12.0	108	80	0
Effect of Test Time at 650°C on Higher BU Samples									
DP-16	0.75	10.4	13.8	533	650	1.0	46	10	0
DP-16	0.61	11.1	14.8	515	650	12.0	36	12	0
DP-16	0.59	11.1	14.8	510	650	36.0	75	55	0
Effect of Test Time at 675°C on Higher BU Samples									
DP-16	0.72	10.5	14.0	528	675	1.0	59	24	5
DP-16	0.67	10.8	14.4	521	675	12.0	140	109	31
Effect of Test Time at 800°C on Higher BU Samples									
DP-21	0.68	10.8	14.3	534	800	1.0	42	0	27
DP-21	0.72	10.6	14.1	540	800	0.5	79	21	63
DP-21	0.80	10.0	13.3	550	800	1.0	100	44	100
DP-21	0.73	10.5	14.0	541	800	2.0	104	45	100

TABLE II.10. (Contd.)

Steady-state Irradiation					Ex-reactor Testing				
Element No.	X/L	Sample BU (at.%)	LHR (kW/ft)	PICT (°C)	Test Temp. (°C)	Test Time (h)	Cladding Total (μm)	Wastage Transient (μm)	Eutectic Formation (Areal %)
Effect of PICT									
DP-16	0.25	11.0	14.7	456	675	1.0	29	29	0
DP-16	0.72	10.5	14.0	528	675	1.0	59	24	5
DP-21	0.54	11.3	15.0	512	740	1.0	58	37	12
DP-21	0.89	9.2	12.3	560	740	1.0	87	46	18
DP-21	0.52	11.3	15.1	508	800	0.5	37	27	58
DP-21	0.72	10.6	14.1	540	800	0.5	79	21	63

Based on the results in Table II.10 and Refs. 19 through 21, the following conclusions can be drawn:

1. Steady-state Irradiation Behavior

Cladding wastage is modest during the steady-state irradiation and will not impede the performance, including the burnup potential (≥ 10 at.%), of the Mk-V fuel.

In a given element, steady-state cladding wastage is a function of both PICT and LHR. The general trend is as PICT increases, steady-state wastage increases as well. However, for the DP-1 elements examined, at the very top of the fuel column, there was a drop off in wastage that follows the drop in LHR. For the EBR-II elements examined, the worst-case combination of irradiation power and temperature occurs between $X/L = 0.70$ and 0.90 .

Steady-state wastage increases with increasing burnup, as the inventory of lanthanides at the interface increases.

Neodymium and cerium were the lanthanide fission products found in the greatest abundance in the fuel/cladding interaction zone.

Ahead of the visible demarcation of fuel cladding chemical interaction (FCCI) in the cladding, a phase transformation has changed the tempered martensite structure of HT9 to a large grain structure with a similar composition to HT9.

On examination of the denuded (decarburized) band under the scanning electron microscope, lanthanides were found as very fine phases precipitated apparently along grain boundaries of the large grains in the band. Therefore, the denuded band should be included as part of the steady-state cladding wastage.

2. Elevated Temperature Testing

The threshold temperature for eutectic formation in the Mk-V-type elements is between $740-750^{\circ}\text{C}$ for a 1-h test on a 6-at.% BU sample. For 11-at.%

BU samples, the threshold is between 650-675°C for a 1-h test and between 650-660°C for a 12-h test. Eutectic formation was minimal at the threshold temperature.

The rate of transient cladding wastage decreases with increasing time at a given test temperature.

The transient cladding wastage values fall within the data band defined by all of the previous FBTA tests. In other words, the wastage of the Mk-V fuel elements as a result of the elevated temperature exposure is no worse than the other fuel and cladding types previously studied.

Increasing burnup lowers the threshold temperature and increases the amount of transient cladding wastage for a given set of FBTA test conditions.

Lowering of LHR during irradiation can slightly raise the threshold temperature for eutectic formation at the fuel surface, but it can significantly reduce the amount of transient cladding wastage.

PICT has no significant effect on either the threshold temperature or the transient cladding wastage.

D. Casting of Metallic Fuel Containing Minor Actinide Impurities

1. Introduction

The Integral Fast Reactor (IFR) utilizes pyroprocessing of metallic fuel to separate U, Pu, and the minor actinides (Am, Np, and Cm) from the nonactinide constituents. The actinides are reintroduced into the fuel and reirradiated. A standard IFR fuel is based on the alloy U-20Pu-10Zr (in weight percent). The metallic fuel system eases the requirements for reprocessing methods and enables the minor actinide metals to be incorporated into the fuel with simple modifications to the basic fuel casting process. The initial casting experience of IFR fuel containing minor actinides is reported with a description of future experiments. The nominal fuel composition used in this experiment is U-20.3Pu-10.0Zr-2.1Am-1.3Np (wt%).

2. Materials and Methods

The typical injection casting temperature for U-20Pu-10Zr is 1465°C. The superheat is about 150°C which is required to fill the molds which are 4.3 mm x 460 mm. The feedstock used was elemental metallic Np, U, Zr and Pu. The Am feedstock was a metallic alloy of Pu-20Am. Potential concerns in casting metallic fuels with minor actinide elements were the wide variations in vapor pressure between Am, Np, Pu and U, and the very highly negative free energy of oxide formation for Am. Over the range of melting temperatures, Am has nearly three orders of magnitude higher vapor pressure than Pu, and five orders of magnitude higher than Np or U.[22,23] This implies that a significant amount of Am could be lost as a vapor at casting temperatures. The potential for Am oxidation arises because Am_2O_3 is considerably more stable than ZrO_2 , UO_2 , or Pu_2O_3 at all temperatures.[24] It was possible that the Am could become part of the casting dross and not be incorporated into the fuel. The challenge becomes how to make these castings with minimum Am loss under the characteristic range of casting conditions. In order to reduce Am loss through evaporation and reaction with oxygen, the time at elevated temperatures where the Am evaporation and reaction rates are high should be kept to a minimum. The fabrication facility was therefore modified to include a "cup" device allowing the Pu-Am feed material to be added to molten fuel within the crucible late in the melt cycle to minimize the time at temperature. Furthermore, the atmosphere above the melt was extremely pure Ar (99.9997%), containing less than 1.5 ppm oxygen.

The elements (U, Zr, Np and some Pu) were loaded into a Y_2O_3 coated graphite crucible and induction heated under an Ar atmosphere. After reaching a temperature of 1495°C, the melt was stirred causing the temperature to drop to 1465°C. The melt was reheated and stirred again. At this point, the Pu-Am was added to the crucible via the "cup" which sat next to and above the crucible inside the furnace. When the Pu-Am feedstock was added, "sparks" were observed emerging from the melt. Casting proceeded and three full-length pins were successfully cast into ZrO_2 coated quartz molds. The entire crucible charge was consumed during casting and the pins were removed from the molds. Two pins were encapsulated and made ready for in-reactor testing while the remaining pin was sacrificed for metallographic and chemical characterization.

3. Results

a. Bottom of Fuel

The as-cast microstructure at the bottom of the fuel is illustrated by the traverse view in Fig. II.9. The salient features are a darker gray, irregularly shaped phase in a light matrix. Two areas of mold-melt interaction are arrowed, (a) and (b). At higher magnification (Figs. II.10 and II.11) the irregular phase is found to be composed of a collection of two different smaller particles. Microchemical analysis by energy dispersive spectroscopy (EDS) given in Table II.11 indicate that the smaller light colored (5-20 μm) particles in Fig. II.11(b) are composed primarily of Zr while the larger dark particles (30-60 μm) have a widely variable composition, but contain at most 20 wt% Zr. Very fine (0.5-1.5 μm) round particles termed "beads" [Fig. II.11(a)] were observed internal to the Zr-rich particles and found to be composed of U. Examination of the matrix by wavelength dispersive spectroscopy (WDS) revealed that the major constituents did not vary radially across the section. A few high Am peaks were detected, but could not be associated with any particular feature. A similar alloy described[25] revealed a majority of the Am segregated to a second phase along with Nd and Ce.

An area of mold-melt interactions arrow A (Fig. II.9) is enlarged in Fig. II.12(a). Figure II.12(b) gives the WDS elemental traces across the deposit, demonstrating that the deposit is enriched in Pu, Am and slightly in Zr, relative to the matrix. The interaction layer at B is morphologically similar but compositionally different to the typical mold-melt interaction layers seen in as-cast U-20Pu-10Zr fuel.

b. Top of Fuel

The microstructure at the center and top sections of the fuel are analogous. The Zr-rich regions are not collections of irregular particles as seen at the bottom of the fuel, rather the Zr was present mainly in a dendritic morphology. No unusual mold-melt interaction layers were observed and the matrix was uniform in composition as detected by WDS. A typical transverse section is shown in Fig. II.13.

Fig. II.9. Transverse Optical Micrograph of the Bottom of the As-cast U-20.3Pu-10.0Zr.1Am-1.3Np Fuel (ANL Neg. No. MCT 281827)

Fig. II.10. A Magnified View of a Dark Gray Region from Fig. II.9 (ANL Neg. No. MCT 281816)

Fig. II.11. Individual Features in Bottom Section of the Fuel. (a) Secondary electron image of the two particle type (ANL Neg. No. MCT 318325); (b) Higher magnification secondary electron image (ANL Neg. No. MCT 318327)

(a)

(b)

Fig. II.12. Morphology and Microchemistry of a Peculiar Edge Deposit. (a) Optical image of deposit at (a) in Fig. II.9 (ANL Neg. No. MCT 281811); (b) Microchemical trace across the deposit as analyzed by WDS

TABLE II.11. Microchemical Analysis of Individual Phases in the Bottom of Fuel

	U	Pu	Zr	Am*	Np*	Others
3a Small Particles (Average of six)	6	3	86	ND	6	
3a and b Large Arrowed Particles	21-47	14-49	9-19	0-25	0-18	Al, Ca, Cr, Ni, Si
3a Matrix Nominal	64	23	12	0.5	0.5	
3b "Beads" 1	87	1	11	ND	Trace	

*These elements are estimates only.

ND--not detected.

Fig. II.13. Transverse Section from the Center of the Fuel, Etched to Show the Zr-rich Phases White and the Matrix Dark. Some grain boundaries are also visible.

c. Bulk Chemistry

Sections from the top, center and bottom of the fuel were chemically analyzed and the results are given in Table II.12. The U, Pu, Zr and Np levels are axially uniform, within experimental error, while the Am levels were all significantly below that amount charged, the Am was lowest in the bottom section versus the top and center sections. There is a notable increase in the amount of Si present in the bottom section versus the top and center sections. This is most likely due to melt reaction with the quartz (SiO_2) molds. The ZrO_2 wash does not completely protect the molds from the melt. The mold ends are submerged in the melt for the entire casting time and fuel located at the bottom of the slug cools last after injection casting.

4. Discussion

A representative transverse section from a U-20Pu-10Zr fuel slug without minor actinide additions is shown in Fig. II.14.

TABLE II.12. Bulk Chemistry of Casting (wt%)

Element	Top	Center	Bottom	Average
U	67.31	65.95	68.02	66.06
Pu	19.51	21.24	19.85	20.20
Zr	9.12	9.22	9.02	9.12
Am	1.33	1.32	1.03	1.23
Np	1.39	1.19	1.36	1.31
Si	0.1790	0.1790	0.4220	0.2600
Al	0.1440	0.0770	0.0450	0.0887
Ca	0.1890	0.0067	0.0046	0.0668
Mg	0.0170	0.0028	0.0065	0.0088
Cr	0.0042	0.0018	0.0015	0.0025
Mn	0.0012	0.0009	0.0008	0.0010
Fe	0.0018	0.0009	0.0007	0.0011
Ni	0.0040	0.0040	0.0040	0.0040

Fig. II.14. Backscattered Electron Image of Conventional U-20Pu-10Zr As-cast Fuel. The globular Zr-rich features appear dark while the U-Pu matrix is light.

In this backscattered electron image, the dark phase is rich in Zr (~80% by weight) akin to the Zr-rich regions observed in the minor actinide bearing fuel; however, they differ morphologically. In conventional ternary fuel the Zr-rich regions consist of contiguous globular features, but in this casting at the bottom of the fuel these "globular" regions were broken up into numerous small particles. This difference in shape could be the result of impurities in the melt. Conventional U-Pu-Zr fuel contains less than 300 ppm Si while the fuel in this experiment contained up to 14 times more Si. The microchemical analysis revealed impurity segregation in the larger particles (Fig. II.11 and Table II.11), i.e., Al, Ca, Si, etc. This analysis is insensitive to the light elements such as C, O and N, and bulk chemical analysis of these elements was not available. Investigations into the U-Zr phase diagram[26] uncovered significant changes in the phase relations at the U-rich end of the phase diagram due to small amounts of oxygen. It is reasonable to expect that the high Si content in the bottom of the fuel would influence the morphology of the Zr-rich phase.

Further investigations into the phase relationships in fuel containing impurities, such as Si, O, C, N, and Ca, need to be done to determine what influence they have on microstructure and phase relationships.

The sparking observed during the minor actinide addition was found to be small droplets of the melt emerging from the crucible. Examinations of the inner furnace surfaces and exhaust lines after the casting revealed increased alpha activity. Solidification of these droplets on the inside bottom surface of the molds is most likely the source of the peculiar deposit found on the metallographic section from the bottom of the fuel [Figs. II.12(a) and (b)]. The "sparks" were probably induced by: boiling contaminants in the Pu-Am feedstock and the high vapor pressure of Am at the casting temperature 1465°C. The Pu-Am feedstock contained ~3 at.% Ca and Mg (2000 ppm by weight). Both of these elements boil below 1440°C causing the melt pool to become agitated. In addition, the relatively cool Pu-Am feedstock contributed to melt agitation. Secondly, the equilibrium vapor pressure of Am at 1465°C is 3.3 torr which yields an evaporation rate of about 0.1g/cm²-s using the Knudsen-Langmuir equation for pure Am in a vacuum. The high vapor pressure of Am is a material property which cannot be eliminated. However, Am loss during casting could be reduced through the development of specialized casting procedures.

5. Future Experimental Plans

Am losses during casting were attributed to agitation of the molten pool due to the Pu-Am feedstock addition and evaporation of Am. The melt pool agitation could be eliminated by loading the Pu-Am feedstock into the crucible with the other elements. However, this technique could result in excessive Am evaporation prior to casting. The high vapor pressure of Am at the casting temperature cannot be altered, but large Am losses can be reduced. Experimentation employing lower superheats would help, but may not be feasible because a lower-viscosity melt might not completely fill the molds. The melt could be prepared under a positive pressure which would reduce Am evaporation somewhat.

During casting, a partial vacuum (~10 ~25 torr) existed above the melt for approximately 20 min. The present controls on the casting furnace

preclude an exact measurement or control of the pressure during casting. Using data on evaporation rates of tungsten[27] under increased pressures, it was estimated that the evaporation rate of Am could be reduced by ~200 times by casting under a pressure of about 800 torr. Furthermore, based on experience with U-Zr injection cast fuel,[28] the vacuum level in the molds could be as high as 150 torr and the molds would still fill. Therefore, the EFL furnace is to be refurbished with new pressure monitoring and control devices. Installation of a capacitance manometer and solenoid valve is planned, which will control the furnace vacuum in the range of 0.01 torr to 1000 torr to reduce the loss of Am during casting. The present vacuum line (2 in.) and air operated valve are too large for the fine control needed to reduce the loss of americium. The modification involves: 1) installation of a smaller bypass line with a solenoid control valve, and 2) installation of an instrument line with the capacitance manometer connected to the furnace. The second line has a filtered connection which will allow safe calibration of the gauges without the spread of contamination.

Upon completion of the modifications, the casting procedure will be modified to incorporate appropriate pressure control methodologies to minimize Am evaporation. Because the processes involved (heat transfer, mass flux and phase change rates), have complex interactions, experiments will be performed on a nonradioactive surrogate alloy such as U-Mn-Zr prior to casting fuel with Am additions. An U-0.5Mn-7.5Zr alloy has the identical vapor pressure and liquidus (thus requiring the same superheat) as the Am-Np bearing fuel and will be used to investigate process variables without the radiological hazards. Facility modifications are nearly complete and testing will soon begin.

6. Conclusions

1. Three full-length pins containing minor actinide additions were successfully cast.
2. No unusual macrosegregation of major constituents was observed.

3. The Zr-rich phase displayed an unconventional morphology in the bottom section appearing as a dense collection of small particles instead of the usual contiguous globular shape. This is probably the result of significant levels of impurities present.
4. Approximately 40% of the initial Am charge was lost during casting due primarily to volatile impurities (Ca and Mg) in the Am-Pu feedstock and through evaporation. Modifications of the current facility instrumentation and the use of new casting procedures are underway to reduce Am loss.

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III. PYROMETALLURGICAL PROCESS DEVELOPMENT

A. Process Development Studies

1. Electrotransport to Liquid Cadmium Cathode

In last year's report[1] the design and initial operation of the pounder cathode was described. This liquid cadmium cathode is needed in the IFR electrorefining process for the collection of plutonium and other transuranic (TRU) elements. Uranium is collected on a solid cathode but, in the presence of UCl_3 in the cell electrolyte, TRU elements will not collect on the solid cathode. Any transuranic material which deposits on the solid cathode reacts with UCl_3 from the salt and deposits metallic uranium in its place. Transuranic elements deposited in cadmium are at much lower chemical activity so that they will coexist with uranium chloride in the electrolyte. To reduce the size of the cathode it is necessary to collect plutonium and other TRU elements in the liquid cadmium well in excess of their solubility (e.g., 3.62 wt% Pu at 500°C). The precipitating TRU phases in this case are intermetallic compounds such as $PuCd_6$. Uranium will also collect in the liquid cadmium cathode; however, when its solubility is exceeded the precipitate is metallic uranium. The problem in cathode development has been in handling the solids; to settle them and compact them into the cadmium ingot. Growth of solids, deposited at the cadmium-electrolyte interface, into the electrolyte phase is to be avoided. This uncontrolled growth causes problems of electrical shorting and prevents clean separation of the electrolyte and cadmium ingot after solidification.

During the year a number of Pu/U electrorefining runs were carried out with the liquid cadmium cathode pounder design. In 12 of the runs, variables were investigated which are important to scale-up (scale-up from 5-cm crucible diameter to 18-cm). The 18-cm crucible is the size needed for plant-scale collection of noncritical batches of transuranic elements during electrorefining of IFR metal fuel. In order to achieve the goal, the cadmium-TRU ingots produced at the cathode must contain at least 10 wt% TRU.

Fig. III.1. Schematic of the Pounder Cadmium Cathode

Figure III.1 is a schematic of the pounder cadmium cathode which shows the variables investigated. The pounder cathode is basically an insulating ceramic cylinder which moves up and down and rotates about the cathode current lead. The current lead terminates in a cadmium liquid metal pool within a ceramic crucible. The cadmium liquid pool interfaces the cell electrolyte with access to the bulk electrolyte of the cell provided through a notch in the ceramic cylinder. The vertical motion of the pounder produces forces at the cadmium-electrolyte interface which causes cathode products to sink into the cadmium pool.

The variables investigated in this series of runs were: the rotational speed (also the vertical motion frequency since the pounder makes two drops for each revolution), radial gap, axial gap, length of stroke, and current. Table III.1 summarizes the variables investigated and the results obtained. Table III.2 gives operating information and information about the product ingots obtained.

The radial gap was a concern because it is not practical to make a close fit between the pounder outer diameter and crucible inner diameter. In Table III.1, Run NE-18 results indicate that increasing the radial gap from 1.5 mm to 3 mm did not lead to uncontrolled growth up the crucible wall. The

TABLE III.1. Pounder Cathode Geometric and Operational Variables Testing

Run No.	Variable Testing	Results
NE-11	No Pounder Motion	Material Growth Outside of Ingot
NE-12	No Pounder Motion	Material Growth Outside of Ingot
NE-13	Position vs. Cell Resistance	20% increase (0.5 ohm $\rightarrow$ 0.6 ohm) from top of stroke to bottom 5% change--Rotational Position of Pounder Gap
NE-14	Current Doubled 1 $\rightarrow$ 2A	Same Favorable Result as at 1 amp
NE-15	Current Tripled 1 $\rightarrow$ 3A (current density 165 ma/cm ²)	Same Favorable Result as at 1 amp
NE-16	Lowered Speed 35 rpm $\rightarrow$ 7.5	Excess Growth Outside of Ingot
NE-17	Higher Speed 35 rpm $\rightarrow$ 68	Same Favorable Result as at 35 rpm
NE-18	Increased Radial Gap 1.5 mm $\rightarrow$ 3 mm	Same Favorable Result as at 1.5 mm
NE-19	Increased Axial Gap 5.8 mm $\rightarrow$ 9.7 mm	Developing Short Due to Buildup on Crucible Rim
NE-20	Standard Operations Cadmium Pool Drawdown	Same Favorable Result
NE-21	Decreased Stroke 9.4 mm $\rightarrow$ 4.3 mm	Developing Short, Buildup on Rim
NE-22	Decreased Stroke 9.4 mm $\rightarrow$ 4.3 mm Increased Speed 35 rpm $\rightarrow$ 68	Developing Short, Buildup on Rim

TABLE III.2. Experimental Results

Test No.	Operating Time, h	A-h	Wt. of Product	Wt. HM in Ingot, g	Wt% Pu in Ingot	% Collection Efficiency ^{ab}
NE-14	5.7	13.1	209.34	32.22	12.39	82.8
NE-15	3.15	9.16	183.10	26.30	11.26	96.7
NE-16	6.03	12.06	206.24	25.34	9.82	70.9
NE-17	8	10.97	190.77	32.05	13.64	98.4
NE-18	5.97	12.65	274.55	39.52	11.73	105.2
NE-19	2.55	5.87	243.13	16.65	5.13	95.5
NE-20	6.03	13.88	358.27	42.95	9.38	104.2
NE-21	2.30	5.29	260.25	16.41	4.89	104.4
NE-22	2.05	4.72	236.12	18.09	6.24	129.0

^aDefined as amount of heavy metal recovered divided by predicted amount of heavy metal based upon 100% current efficiency.

^bValues above 100% are due to sampling error.

axial gap is the spacing between the bottom surface of the pounder and the liquid cadmium pool surface when the pounder is at the bottom of its stroke. For Run NE-19 the axial gap was increased from the nominal 5.8 mm to 9.7 mm. As noted in Table III.1, material buildup on the rim of the crucible occurred; this led to electrical shorting. When the length of the stroke was shortened from 9.4 mm to 4.3 mm, in Run NE-21, shorts due to material buildup on the lip of the crucible also occurred. Buildup of material on the lip of the crucible can occur only when there is an electrical connection between the buildup and the cadmium pool. This can occur when a metallic (electrically conductive) path forms up the crucible wall. In Run NE-22 the short stroke was retained but the speed was doubled; this did not keep the buildup on the crucible rim from occurring. By doubling the speed, the same overall salt displacement rate takes place as for the normal speed (35 rpm) and normal stroke (9.4 mm). When the speed was lowered from 35 rpm to 7.5 rpm (Run NE-16), material again accumulated outside the cadmium ingot. Increasing the current up to a current density of 164 ma/cm^2 , at the cadmium surface, did not have an adverse effect. Runs NE-11 and -12 were conducted with no pounder motion and in each case material growth outside the cadmium ingot was found. In Run NE-13, the effect of pounder position on cell resistance was measured.

Operating information and product information is given in Table III.2. The appearance of the product ingots were similar to the appearance of ingots from Runs NE-1 through NE-10 which are shown in last year's report.[1]

Except for those cases where shorting occurred the goal of loading the cathode to more than 10% plutonium was achieved. This study has defined the variables and their limits which need to be observed for scale-up. These studies have been made with only plutonium present, or with a (Pu/U) ratio in the electrolyte greater than 2.0. Further work will be done with plutonium, greater uranium, and rare earths present and will investigate materials of construction for the crucible and pounder parts.

2. Electrorefiner Drawdown Process Development

During the drawdown of the Fuel Cycle Facility (FCF) electrorefiner it may be necessary to remove large amounts of uranium from the salt. In the early stages of this drawdown it is convenient to use a normal solid cathode to collect uranium. In last year's report,[2] a series of drawdown operations were described in which the uranium concentration in the cell electrolyte was reduced from 6.68 wt% to 0.015 wt% using a number of solid cathodes to collect the product. Recently, Run 101 was completed to demonstrate the growth of a large uranium cathode deposit at reduced concentration of uranium in the electrolyte, as will occur during the drawdown. For the test, Li-Cd alloy was added to the system to reduce the uranium concentration in the salt from the nominal 2 mol % HfCl_3 (7.7 wt% HM) to 3.4 wt% uranium in the salt. The cadmium pool was then made the cell anode and uranium was transported to the solid cathode. About 1.5 wt% rare earths were in the electrolyte also. A deposit of 13.18 kg was obtained. The deposit contained 81.2 wt% U, 13.6 wt% salt, 2.7 wt% Zr and 0.8 wt% Cd. The rare earth metals which were in the cathode were consistent with the amount of rare earths in the salt contained in the product, indicating no rare earth metal deposition, as expected. The uranium dendrites were much coarser than those normally found with 2 mol % uranium chloride in the salt. This test demonstrated that 10 kg of uranium can be removed on the normal solid cathode during the early stages of a drawdown run.

3. Recovery of Zirconium Removal by Electrotransport

Zirconium is 10 wt% of normal IFR fuel which is dissolved in the electrorefiner. With repeated normal electrotransport cycles, uranium is removed from the electrorefiner with the solid cathode, and the bulk of the zirconium is left to accumulate in the electrorefiner (in the cadmium anode pool and on vessel and component surfaces). Tests in the engineering-scale electrorefiner to remove this zirconium in separate electrotransport steps were successful and reported previously.[3] In this report period, experiments were conducted to investigate conditions which results in the co-transport of uranium and zirconium to the same cathode. This has the potential advantage of direct recycle of zirconium, one of the main constituents of the fuel, with the actinides to make new fuel.

For these experiments, uranium-zirconium alloy (9.165 kg U and 1.018 kg Zr in the alloy) was loaded into the four baskets of the anodic dissolver. The first step (Run 95) in the operation was the direct electrotransport of metal from the anodic dissolution baskets to the solid cathode mandrel. Only partial dissolution was attempted, then the Cd pool of the ER was made the anode and the U and Zr which passed into the Cd pool (fallen dendrites from the solid cathode) were transported to the cathode. This two-step cycle was repeated and then the dissolution was completed using the cadmium pool as the cathode. After this, the baskets were inspected and no undissolved fuel was found. Low mixing rates (0-20 rpm for the anode baskets, 0 rpm for the Cd mixer and 0-50 rpm for the salt mixer) were used, because earlier tests had indicated that this was favorable for simultaneous transport. In normal dissolution the anode basket speed is 75 rpm and under this condition, 664 ampere-hours are required to dissolve each kilogram of uranium. Under the low-rotating speed (about 20 rpm) about 20% more total current flow than normal was required to complete the dissolution from the anode baskets.

After Run 95, two more solid cathode deposition experiments (Runs 96 and 97) were carried out to remove uranium and zirconium from the cadmium pool to complete the transport of the materials which were originally in the anode baskets. At the end of Run 97 the uranium and zirconium concentrations in the cadmium pool were 0.025 and 0.084 wt%, respectively.

Results for the uranium and zirconium material balances for the codeposition tests are given in Table III.3. The weights of U and Zr in the cadmium pool were calculated from chemical analysis of filtered cadmium samples and the estimated weight of the cadmium pool. The weights of U and Zr in the solid cathodes were calculated from multiple samples taken by a systematic sampling method developed from experience in sampling many solid cathode products. The estimated weights of uranium and zirconium at startup were 9.17 and 1.98 kg, respectively. After the tests, the estimated weights were 9.40 kg and 1.94 kg, respectively. These results show a good mass balance and confirm the cotransport of U/Zr. Compositions for the cathode products are shown in Table III.4.

TABLE III.3. Uranium and Zirconium Material Balance for Codeposition Tests

Electrorefiner Component	Weight of Uranium, kg	Weight of Zirconium, kg
Cd Pool (startup)	—*	0.96
Anodic Dissolution Baskets (startup)	9.17	1.02
	Total (startup)	1.98
Cathode Product		
Run 95	6.56	0.78
Run 96	2.01	0.30
Run 97	0.73	0.51
Total (product)	9.30	1.59
Cd pool (after testing)	0.10	0.35
Anodic Dissolution Baskets (after testing)	—*	—*

*Negligible; less than 0.01 wt% uranium.

TABLE III.4. Composition of Deposits from Uranium and Zirconium Codeposition Tests

Component,* wt%	Solid Cathode Deposit Test Number		
	95	96	97
Uranium	71.1	63.6	32.0
Salt	13.5	18.6	34.6
Zirconium	8.5	9.3	22.5
Cadmium	0.8	1.3	1.2

*Estimated uncertainty in the chemical analysis is ± 5 to 10%.

B. Waste Treatment Process Development

Work continued on developing processes to recover TRU elements from the spent electrolyte salt and metal discharged from the electrorefiner and to

separate the fission products so the treated salt and metal can be recycled. During this period, experimental process development studies have been carried out on 1) Salt Extraction; 2) Salt Stripping, and 3) Pumped Filtration of Salt and Cadmium.

1. Salt Extraction Process

Several steps in the IFR salt purification process employ the use of high-temperature centrifugal contactors ("pyrocontactors") to carry out salt/metal extractions. One of the goals of this work is to develop a continuous metal-salt extraction process that uses multistage, countercurrent pyrocontactors to recover the TRU elements from the spent electrorefiner salt. To that end, a pyrocontactor test system has been built and is being tested with molten cadmium and salt at 500°C. The test system (Fig. III.2) includes a single-stage pyrocontactor with 4-cm-dia rotor (Fig III.3), tanks to feed salt and metal to the contactor, tanks to receive salt and metal from the contactor, tanks to clean and treat salt and metal, and heated transfer lines, all housed in a glovebox with an argon purification system.

The initial tests included (1) charging, melting, and mixing 74 kg cadmium in the metal treatment tank and 15 kg LiCl-KCl eutectic salt in the salt treatment tank, (2) transferring materials between the treatment tanks, the raffinate tanks, and the feed tanks, and (3) feeding liquid cadmium from the feed tank through the pyrocontactor to the treatment tank. The first two operations were successfully carried out. The maximum gas pressure required for the liquid transfer between tanks was 0.2 MPa (35 psi). During this testing, the equipment furnaces and heaters operated satisfactorily. The agitators for the tank were operated at up to their maximum speeds of 350 rpm.

The flow tests with liquid cadmium through the pyrocontactor were also satisfactorily completed using both a 0.5- and 0.3-cm dia orifice in the feed line. In tests at 500°C with the contactor rotor spinning at 1800-2700 rpm, the mechanical performance of the pyrocontactor was excellent--no rotor vibration or over-temperature of the motor bearing occurred.

Fig. III.2. Schematic of Pyrocontactor Test System

Fig. III.3. Pyrocontactor

Next, two tests were completed to assess the pyrocontactor operation while liquid cadmium and molten salt at 500°C were fed to the spinning contactor from the feed tanks. The effluents from the contactor flowed to the treatment tanks. The first test was terminated when flooding of the contactor was observed. The flooding was caused by plugging of the metal effluent line and salt feed and effluent lines. Metal and salt samples were collected from the feed and effluent lines after the test and were analyzed by inductively coupled plasma/atomic emission spectrometry. Results indicated essentially no phase crossover from the contactor, i.e., metal exited from the metal effluent line, while salt exited from the salt effluent line. The finding of little phase crossover is particularly encouraging because the metal-to-salt feed ratio was high, about 3:1. After this test, the transfer lines were modified to prevent plugging from occurring again. A second test was then performed with liquid cadmium and salt flowing through the spinning pyrocontactor at 500°C. The average feed rates were 1.0 and 0.67 L/min for cadmium and salt, respectively. During this flow test, the contactor operated very smoothly, no vibration was observed, and the contactor rotation speed was easily controlled. This test demonstrated that feed rates could be controlled and indicate that the entire contents of the electrorefiner can be treated in 4-6 h. A four-stage pyrocontactor is now being designed for further testing of salt extraction.

2. Salt Stripping Process

Another step in the salt purification process is salt stripping, in which salt from the extraction step is contacted with a Cd-Li alloy. The lithium reduces essentially all of the uranium and residual TRU elements from the salt, as well as most of the rare earths, but none of the alkali metals and alkaline earths. Tests have been carried out using a salt stripper system which previously had been installed in an argon-filled glovebox adjacent to the engineering-scale electrorefiner.

Before the stripping experiments, a submersible centrifugal pump and transfer line designed for high-temperature service were inserted into the electrorefiner. The pump and transfer lines were connected with freeze joints in which silver solder formed the seal. Some difficulties were encountered

during the initial attempts to pump the salt. However, after some modification, 90 kg of LiCl-KCl eutectic salt (containing U, Nd, Ce, and Y as chlorides) was pumped from the electrorefiner at a slow rate of about 4.5 L/min and an impeller speed of 800 rpm. The freeze seals held up extremely well, and the seals were easily made and disassembled. The pump will be modified further to increase its capability for more extended operation. After the salt was transferred, the pump and transfer line were removed, and 150 kg of cadmium was then charged from the electrorefiner to the stripper vessel, equivalent to a depth of about 18 cm.

During the stripping experiments, an inverted cup was used to charge 85 wt% Cd-15 wt% Li alloy into the cadmium layer at the bottom of the stripper vessel (Fig. III.4). The charging device consisted of an inverted

Fig. III.4. Schematic of Salt Stripping Apparatus

cup on a rod. The cup contained the Cd-Li alloy which has been case into it. The Cd-Li alloy is a liquid at the 500°C temperature of the stripper contents and is lighter than cadmium. When the cup was immersed into the cadmium, the Cd-Li alloy melted, and the lithium reductant was released slowly into the cadmium layer and then came into contact with the salt at the salt-cadmium interface. After insertion of the cup with the alloy, the mixer was turned on, and samples of cadmium and salt were taken at various times. In certain tests, cadmium chloride was added to the stripper vessel. The transfer of uranium and rare earths between the salt and cadmium is dictated by the amount of reductant (Li) or oxidant (CdCl_2) added and by the equilibrium between uranium and the rare earths.

Figure III.5 illustrates the typical behavior of uranium and the rare earths (Nd, Ce, and Y) during these tests (Nos. S4 through S9). Uranium initially transferred from the salt as the Cd-Li alloy reductant was added. Further additions of alloy caused Nd and Ce to also transfer from the salt, and finally yttrium began to transfer after several more additions of the reductant alloy. The experimental data agree quite well with the values predicted by the XTRACT computer code, which was based on earlier laboratory-scale experimental determinations of separation factors.[4]

During the tests, the voltage output of an Ag-AgCl reference electrode in the salt was monitored. Figure III.6 is a plot of the reference electrode output vs. the actual analytical determinations of uranium content, and also the XTRACT code predicted values of uranium concentration in the salt. There is considerable scatter with the data; however, the Ag-AgCl reference electrode will give an estimate of the actual uranium concentration in the salt and will also aid in indicating the approach to steady state conditions.

3. Pumped Filtration of Salt and Cadmium

Preparations were made to conduct filtration tests which will consist of pumping salt or cadmium through a sintered metal filter housed in an assembly mounted atop the pump assembly. Modifications have been made to the centrifugal sump pump assembly in order to increase its capability to run

Fig. III.5. Measured Concentrations of Uranium and Rare Earths in Electrolyte Salt During Stripping Tests S4 Through S9. (Curves show predicted behavior from the XTRACT code.)

Fig. III.6. Uranium Concentration in Salt vs. Reference Electrode Output

for longer durations for these filtration tests. The modifications to the pump included conversion from gear to V-belt drive, drilling holes in the pump shaft housing to assure lubrication of the journal bearing in the pump, replacement of two sets of upper pump shaft ball bearings with bearings of greater internal tolerances and modification of the upper bearing housing. A schematic of the pump impeller assembly is shown in Fig. III.7.

A series of pump tests will be carried out to evaluate the effectiveness of the pump modifications and then filtration tests will commence.

Fig. III.7. Schematic of Pump Impeller Assembly

C. Waste Management

High-level wastes from the IFR process are placed in either a mineral or a metal waste form, depending on their chemical character. "Noble" metal wastes (those elements that are at least as difficult to oxidize as zirconium) are placed in a metal waste form. The metal waste form thus includes cladding hulls, alloy zirconium, and noble metal fission products. Easily oxidized metals are placed in the mineral waste in their oxidized form, along with the nonmetal fission products. The mineral form accommodates cations derived from coolant sodium and alkali, alkaline earth and rare earth fission products as well as nonmetal fission product anions. Neither waste form will contain more than about 10 ppm of transuranium elements.

1. Mineral Waste Form Development

Two candidate aluminosilicate materials are being developed as a mineral waste form for the molten salt waste: glass bonded zeolite and sodalite. Both are formed from a mixture of fission product-loaded zeolite A and "anhydrous" sodium zeolite A.

In the IFR flowsheet, the process salt is passed through a column containing Li-K zeolite A that has been pre-equilibrated with liquid LiCl-KCl. Each nominal unit cell of this pre-equilibrated zeolite contains 12 equivalents of charge-balance cations, M^{+n} , and up to 12 equivalents of salt that is contained within the crystal structure of the zeolite; the chemical formula may be written $M_{12/n}(SiAlO_4)_{12} \cdot 12/n MCl_n$, where n is the mean charge on the cations. The column also contains some free liquid salt that fills the interstices between the zeolite particles, both before and after the IFR process salt has been passed through it.

Fission products--the halides, chalcogenides, alkali metals, alkaline earths, and rare earths--are removed from the IFR process salt by exchange onto the column. The fission products replace Li and K ions, which enter the IFR process salt. The process salt is returned to the electrolyzer. The fission product-loaded column, together with any liquid salt that remains on it after mechanical salt removal, is blended at about

450°C with excess "anhydrous" (actually containing several weight percent water) sodium zeolite A to occlude the remaining free liquid salt. The blended zeolite is fabricated into a sodalite waste form or a glass-bonded zeolite waste form.

2. Mineral Waste Form Precursors

A detailed understanding of the ion exchange properties of zeolite A is necessary for designing and optimizing column operations. Several types of experiments have been completed. The objective of one experiment was to determine if preloading zeolite A pellets with KCl-LiCl affected its ion exchange properties. Two samples of dehydrated zeolite 3A pellets (3-mm dia) were used. One (P34) was heated with excess LiCl-KCl at 450°C for 24 h, then filtered with pressurized argon to remove most of the excess salt, the other (P33) was used as is. Both samples were then heated for 24 h at 430°C in a simulated IFR salt containing rare earth elements at a salt:zeolite weight ratio of 4.5:1. Analyses of both samples showed that the multivalent ions are preferentially exchanged into the 3-mm zeolite pellets based on the relative concentrations in the salt. The rare earths show the greatest exchange, alkalis the least.

In another experiment, ion exchange was measured between 1.5-mm pellets of zeolite A occluded with a simulated IFR salt and dehydrated zeolite 4A powder in a 1:3 weight ratio. The mixture was heated to 500°C for 24 h under an argon atmosphere. After heating, a sample of the powder and several pellets were analyzed. Exchange of ions between the pellets and powder is slow. There was very little exchange of Ba and Sr relative to K or Cs. The salt-occluded zeolite appears to retain the divalent ions more strongly than the monovalent ions. Li, K, and Cs are all exchanged in approximately the same proportions, indicating that their rates of exchange are similar over the 24 hour experiment time. Ionic charge apparently plays a big role in the rate of exchange of ions in the back blending step. There may be a nonuniform distribution of fission products on a microscale, particularly for divalent and trivalent ions.

Kinetic experiments that measure the rate of ion exchange between LiCl-KCl-preloaded zeolite A pellets and simulated IFR salt containing cesium, barium, strontium, cerium, lanthanum, neodymium and yttrium were also started. The first time period investigated was 1 min to 24 h (1440 min).

The ion exchange reaction between any two ions can be represented by the following:

where z and s represent the zeolite and salt phases. At the end of the contact period, the salt and zeolite were separated. Each phase was sampled and analyzed for the cations of interest. The concentration data were calculated in terms of an exchange factor, defined by

$$\text{EF} = \{\text{M}^{n+}(z)/\text{M}^{n+}(s)\}^{n+} \cdot \{\text{Na}(s)/\text{Na}(z)\} \quad (2)$$

where the exchange of cations is measured relative to the sodium ion. Sodium is chosen because it is the least strongly retained by the zeolite; the exchange factors thus all have values greater than one. Similar expressions can be written for anions.

The exchange rate data are given in Fig. III.8. Only data for cesium, strontium and cerium are shown for clarity. The barium data are similar to the strontium data and the rare earths (lanthanum, neodymium and yttrium) are similar to the cerium data. The experimental data are shown as points; these were fitted using Eq. (3):

$$C(t) = A(1 - \exp(-kt)) \quad (3)$$

where A and k are fitting parameters and can be thought of as the steady state concentration and the rate constant, respectively. Equation (3) has the form of a homogeneous first order rate expression.

A qualitative consideration of these data indicates that the rate of ion exchange decreased with increasing valence state of the cation. For

Fig. III.8. Exchange Factors for Cs, Sr, and Ce

example, at 2 h contact time, the concentrations of cesium, barium and strontium were 85% or more of their concentrations at 24 h, whereas the concentrations of the rare earths were only about 40-50%. Work is now ongoing to confirm these initial values and to obtain additional data at longer times.

3. Glass-bonded Zeolite

To prepare glass-bonded zeolite, the blended zeolite is converted to a solid monolith by hot pressing the zeolite with glass frit. More than 20 glass-bonded zeolite test specimens have now been fabricated, using four ratios of zeolite and glass frit. The performance of these composites was measured by MCC-1 leach tests. All specimens were tested for 28 days in deionized water at 90°C.

A detailed description of the preparation of salt-occluded zeolite (referred to as zeolite throughout this text) has been published elsewhere.[5] Four grams of zeolite and glass frit were mixed at room temperature, loaded into a 2.5-cm graphite die, cold pressed to 41 MPa and then placed in a uniaxial hot press and evacuated. The chamber was back filled with nitrogen

to atmospheric pressure, then a pressure of 28 MP was applied. Standard pressing conditions were 28 MPa and 720°C. The heating rate was 20° per minute, and typical hold times were 10–20 min. Scoping experiments in which the hot pressing conditions (temperature, pressure and hold time) are varied from the standard conditions are ongoing.

Leach testing of the resulting samples was done following the MCC-1 leach test procedure, but with one exception: all polishing and cutting operations were done dry rather than with a lubricant. Each specimen was cut into quarters, and different quarter was used for each leach test. The leach test results are calculated in terms of the normalized release rate (NRR), which is defined as

$$\text{NRR} = C_i V / f_i A d \quad (4)$$

where C_i is the concentration of the element i in the leachate, V is the volume of the leachant, f_i is the fraction of the element i in the solids initially, A is the geometric surface area, and d is the duration of the test. Each specimen was weighed to 0.1 mg before and after the leach test to determine mass loss. The solids were dissolved in various acid solutions. The concentrations of the cations in the dissolved solids and leachates were measured by inductively coupled plasma spectroscopy-atomic emission spectroscopy (ICP-AES) or inductively coupled plasma-mass spectroscopy (ICP-MS); the chloride and iodide concentrations were measured by ion chromatography.

Most of the new specimens contained equal amounts of glass and blended zeolite. All specimens pressed under standard conditions and with standard materials had NRRs of $<1 \text{ g/m}^2\text{d}$ for all cations. Some specimens that were fabricated under nonstandard pressing conditions or with nonstandard materials had lower leach resistance. This will be discussed in more detail below and in later reports.

Nineteen glass-bonded zeolite test specimens have now been fabricated. These specimens were fabricated from mixtures containing 50, 60, 67, and 75 wt% blended zeolite with the remainder glass frit. All specimens

containing 67 wt% blended zeolite or less met the development of NRR criterion: $<1 \text{ g/m}^2\text{d}$ for all elements. Of the two specimens containing 75 wt% zeolite, the NRRs were $<1 \text{ g/m}^2\text{d}$ for all elements except potassium. Figure III.9 is a plot of the average values of the NRRs for each element versus the blended zeolite fraction. The release rates of the matrix elements (Al, B, and Si) and the divalent cations (Ba and Sr), are only slightly affected by increasing the amount of zeolite from 50 to 75 wt%. The NRRs of the univalent cations, Cs, K, Li and Na, increase more markedly with increasing amounts of zeolite. Interestingly, the NRR for cesium is smaller than the NRRs for the other univalent cations in all cases. The average mass loss in the specimens with 50 wt% blended zeolite was $0.33 \pm 0.05 \text{ wt\%}$ while for the others it was $0.38 \pm 0.03 \text{ wt\%}$. These data suggest that a composite containing equal amounts of glass and zeolite has better overall leach resistance than those with more than 50 wt% zeolite.

Five specimens were leached for 56 days. Two of the specimens contained 50 wt% zeolite, one contained 60 wt% and the other two contained 75 wt%. The mass losses in the 56-day tests were about 15% greater than those in the 28-day tests. The average NRRs for the 28- and 56-day tests are given in Table III.5. The 56-day NRRs were about 40-70% of the 28-day NRRs. These results suggest that the initial release predominates and steady-state conditions are being approached.

Fig. III.9. Normalized Release Rates (NRR) for Four Zeolite-glass Compositions

TABLE III.5. Normalized Release Rates ($\text{g/m}^2\text{d}$) vs. Time and Leachant

Leachant	28 Water ^a	56 Water	28 Water	28 Brine	56 Brine	28 Water
Element						
Al	0.30	0.13	0.28	0.00	0.05	0.28
Ba	0.21	0.08	0.22	0.43	0.30	0.18
B	0.31	0.19	0.35	0.49	0.34	0.33
Ca	0.19	0.14	0.17	0.70	0.25	0.39
Cs	0.60	0.32	0.61	0.57	0.26	0.51
K	0.73	0.38	0.63	b	b	0.48
Li	0.60	0.35	0.54	0.42	0.23	0.49
Na	0.67	0.37	0.71	b	b	0.58
Si	0.25	0.11	0.23	0.15	0.17	0.23
Sr	0.23	0.11	0.24	0.51	0.22	0.23

^aWater=Deionized water.^bConstituent of brine.

Two specimens that were leached in deionized water were also leached in brine for 28 days, although a different quarter was used. The brine is a concentrated chloride salt solution, containing 29.4, 25.3, and 35.4 mg/mL of Mg, K and Na, respectively. If ion exchange occurs with any of these ions, then the NRRs of the exchanging cations in the waste form should increase measurably in the leachates.

In all of the brine tests, the mass loss was <0.1 wt % and the NRRs for all the elements were <1 $\text{g/m}^2\text{d}$. The average NRRs for specimens leached in brine and deionized water are given in Table III.5. In brine, the NRRs are smaller for Al and Si, about the same for Cs and Li, and significantly higher for the divalent ions and boron. Because the zeolite phase is the only source of Cs and Li, and the glass phase the only source of B and Ca, one may conclude that ion exchange has occurred between the brine and the glass but not between the brine and the zeolite. Al and Si are common to both the zeolite and the glass phase and it is not clear why their concentrations are depressed in the brine, especially since the B release is enhanced.

One 56-day brine leach test has been completed with a specimen containing 67 wt% zeolite. The specimen's mass was the same before and after the leach test, within ± 0.1 mg. The two 28-day and one 56-day brine leach tests can be compared (even though different specimens were used) because their 28-day deionized water NRRs are comparable. (The larger differences in the NRRs for K and Na are partially attributable to the different zeolite fractions used in the three specimens.) A tentative conclusion is that the NRRs in brine decrease with time for all elements except possibly Al, Si and Ca. These small increases may not be significant. Further tests are planned.

Several specimens were fabricated under different pressing conditions and then leach tested. The NRRs for specimens pressed at 42 or 56 MPa were slightly higher than for the specimens pressed at 28 MPa. Other experimental variables that appear to adversely affect leach resistance were incomplete zeolite blending, incomplete densification, and excessive fabrication temperature.

The following criteria have been used to assess the leach resistance properties of glass-bonded zeolite:

- NRR of each element should be $<1 \text{ g/m}^2\text{d}$ in 28 day, 90°C leach tests.
- NRRs decrease with time and reach steady-state values.
- The zeolite should not function as an ion exchanger.

The results obtained to date indicate that these criteria are being met and that glass-bonded zeolite is a promising mineral waste form for salt-derived wastes.

4. Sodalite

Several samples of loaded zeolite A were blended with salt-free dehydrated zeolite A to reduce the chloride ion content of the zeolite to the capacity of sodalite, which is approximately 2 Cl^- ions per sodalite unit cell.

These zeolite mixtures were heated above 425°C for at least 24 h. X-ray diffraction (XRD) of the resulting powders indicated sodalite was the major crystalline phase. Nepheline was usually present as a minor phase.

The samples were consolidated by uniaxial hot pressing. Applied pressures were 20 and 100 MPa and final temperatures ranged from 1050 to 1300°C. The resulting samples were hard and polishable, but brittle. Their densities fell between 2.2 and 2.4 g/cm³. As in the starting powders, sodalite and nepheline were generally the major and minor components after pressing. Carnegieite formation was observed at 1300°C. Most samples showed only small changes in the relative intensities of nepheline and sodalite peaks in powders and hot pressed pellets.

Chemical analysis of several sets of samples are given in Table III.6. They show similar compositions in the initial sodalite powders and resulting pellets. The differences in cesium concentrations for samples c11172, c06083, c06303, and c09023 are consistent with a loss of this ion during hot pressing. For sample c11172, and possibly for c06303, this loss appears to be due to the high pressing temperature. In sample c06083, the loss of cesium is attributed to the nearly complete conversion of sodalite to nepheline.

TABLE III.6. Compositions of Hot-pressed Samples and Precursor Powders (wt%)

Sample	Pressing Temp., °C	Cs	Ba	Na	Al	Si	K	Sr	Li
c11172		0.19	0.01	13.6	16.9	16.7	1.67	<.001	0.30
c1172hp	1300	0.11	0.009	15.5	17.7	16.9	1.66	0.004	0.29
c11242		0.21	0.01	13.9	16.9	17.3	2.74	<.001	0.008
c11242hp	1050	0.19	0.02	14.4	16.8	16.0	3.40	0.01	0.59
c03243		1.70	0.14	13.5	16.1	16.0	1.48	0.03	0.87
c03243hp	1050	1.76	0.12	13.0	16.5	16.5	1.76	0.03	0.90
c06083		0.89	0.30	7.34	16.5	16.1	9.10	0.40	1.07
c06083hp	1050	0.65	0.30	7.64	16.8	16.5	8.65	0.41	1.11
c09023		2.19	0.06	9.99	16.8	16.7	2.71	2.12	1.66
c09023hp	1050	1.72	0.06	9.83	17.0	16.9	2.57	2.20	1.66
c09133		1.69	0.94	11.3	16.9	18.0	2.19	0.34	1.25
c09133hp	1050	1.73	0.92	11.2	16.9	18.2	2.24	0.33	1.27
c06303		2.08	0.79	11.3	7.1	17.3	1.89	1.07	1.30
c06393hp	1075	1.53	0.77	11.2	17.2	17.2	2.36	1.07	1.27
SR-1 (Rock)		*	*	17.7	16.6	16.3	*	*	*

Analysis by ICP-AES. Cs by flame emission.

*Values are near or below detection limits for ICP-AES.

Normalized release rates (NRRs), defined as before, were determined for several of the hot pressed sodalite samples. A modified MCC-1 test method was used. The pellets were leached at 90°C for a total of 28 days with 10 mL of deionized water per gram of sample. After 7, 14, 22, and 28 days, the leachant was removed for analysis and replaced with fresh leachant, and the pellets were dried and weighed. The NRR results are summarized in Fig. III.10.

Surface areas were determined by direct measurement, except for samples where only small irregular pieces were available for which the total bulk surface areas were estimated as $5 \times 10^{-4} \text{ m}^2$. Because of the fragmentary nature of many samples, the surface-to-volume ratio of 10 cm specified by MCC-1 test procedures could not be maintained. Consequently, the results presented are only indicative of the ultimate performance of sodalite.

Fig. III.10. Average Normalized Release Rates (NRR) for Sodalite Pellets

The values shown in Fig. III.10 are averages of the normalized release rates for five different pellets. The NRR values tended to vary over a wide range, especially in the first test period. Cesium release rates, for example, ranged from 1.2 to 80 g/m²d for the first 7-day period. For the other alkali and alkaline earth elements, the variation was much smaller but still significant. Li, K, and Ba also have NRR values above 1 g/m²d in week 1. For the 2nd, 3rd, and 4th weeks, the average NRR values were generally below 1 g/m²d for all elements other than Cs and K. It is clear that there was a large reduction in NRR values after the initial 7-day period for all elements except for Si and Al, the matrix elements. Al and Si release rates were relatively constant over the all test periods with NRR of approximately 0.4.

The pellets tested contained sodalite as the major phase, with a minor nepheline component and NaCl and/or KCl features present in some cases. The large initial NRR values may be attributed to the presence of unoccluded salt that is removed rapidly by the deionized water. Unoccluded salt is often observed when large nepheline phases are present in the synthetic sodalite. A sample of pelletized nepheline which was tested for one 7-day period showed the poorest overall performance of all samples tested. The Cs release rate for this sample was 132 g/m²d, and values for Ba, K, Sr, and Li were 25, 9, 3.3, and 6.7 g/m²d, respectively, the highest measured values in nearly all cases. Si and Al release rates were low, 0.3 and 0.04 g/m²d. Nepheline phases may therefore partially explain the poor initial retention of alkalis and alkaline earth elements in many samples, perhaps because of associated surface salt.

In order to gauge the ultimate effectiveness of sodalite as a waste form, nearly phase pure sodalite free of nonoccluded salt is required. Synthesis and evaluation work continues.

5. Metal Waste Form Development

The objective of the metal waste form development effort is a method for consolidating the metal waste constituents into a monolithic waste form acceptable for disposal in a geologic waste repository. The primary waste streams to be considered include the HT9 stainless-steel cladding hulls, alloying zirconium, and the insoluble noble metal fission products.

The zirconium stainless-steel-based waste form is designed to be a macroscopically homogeneous alloy, made by melting and alloying the noble metal fission products with the zirconium and stainless steel from support hardware. Noble metal fission products represent approximately 5 wt% of the waste form alloy. If metal filters are used, they can be melted with the other waste stream constituents. Melting is carried out under an oxidizing flux to assist with removal of actinide elements from the salt that clings to the metals, and from metal that may remain attached to hull walls.

a. Laboratory-scale Steel Waste Form Experiments

Small alloy samples (15 to 20 g) were generated to simulate waste form compositions and to characterize alloy microstructures and corrosion resistance. Table III.7 summarizes the alloy compositions generated. The

TABLE III.7. Composition of Steel-based Waste Form Samples (wt%)

Sample	Temp, °C	HT-9	Zr	Rare Earths ^a	Noble Metals ^b
HT9-1	1850	100.0	.	.	.
HT9-2	1900	76.0	24.0	.	.
HT9-3	1900	86.5	13.5	.	.
HT9-4	1525	87.5	12.5	.	.
HT9-5	1525	80.5	12.5	7.0	.
HT9-6	1800	75.0	12.5	7.5	5.0
HT9-7	1850	100	.	.	.
HT9-8	1850	95	5	.	.
HT9-9	1850	95	5	.	.
HT9-10	1850	90	10	.	.
HT9-11	1850	90	10	.	.
HT9-12	1850	85	15	.	.
HT9-13	1650	85	15	.	.
HT9-14	1850	80	20	.	.

^a3% Nd, 2% Ce, 1% La, 1% Pr, and 0.5% Y.

^b2% Ru, 1.5% Pd, 1% Te, and 0.5% Ag.

initial samples, HT9-1 through HT9-6, were generated in a high temperature furnace under approximately 0.3 atm of flowing helium. They provided an initial survey of the HT9-Zr and HT9-Zr-RE-NM alloy microstructures over a range of compositions. The intermetallic compound Fe_3Zr was observed in HT9-2 and the rare earth elements were observed to segregate into discrete phases on the order of 5 to 50 μm in size while some of the noble metal fission products (notably Ru) were observed by EDX to be distributed in the alloy matrix.

The furnace used for HT9-1 through HT9-6 was available as a temporary arrangement while a dedicated high temperature system was being established. The system is a refurbished high temperature vacuum furnace with a sealed, controlled atmosphere specimen chamber. The furnace has been fully repaired, tested, and calibrated, and all experimental review procedures have been completed. A schematic diagram of the experimental assembly is shown in Fig. III.11. The assembly consists of a Mo-30% W sample chamber which is suspended inside of the vacuum furnace using a water-cooled O-ring fixture. The furnace chamber (not the sample chamber) is operated under vacuum (10^{-6} to 10^{-5} Torr). The molten samples are contained in stable ceramic crucibles, and the Mo-W sample chamber is lined with a drawn refractory alloy (C-103: Nb-10% Hf-1% Ti) that serves as a secondary container for the experiment. The sample chamber is sealed above the furnace by a second O-ring fixture that is integrated with a packing gland that allows the insertion of a type-C thermocouple and gas inlet and outlet lines. The tantalum gas inlet tube intrudes into the hot zone of the sample chamber to supply flowing, high-purity argon near atmospheric pressure.

Alloy samples HT9-7 through HT9-14 were generated (using the system in Fig. III.11) to be used for microstructural characterization and corrosion testing. The alloys were melted at 1850°C under argon at 1 atm and held for 20 min before cooling. Horizontal cross-sections of the prepared alloy ingots were metallographically polished and examined with an optical microscope. Figure III.12 is from sample HT9-8 (5 wt% Zr), and it shows a dendritic Fe-Zr intermetallic phase distributed in the matrix. A similar structure was previously reported[6] for sample HT9-2 where the dendrites were identified by EDX as Fe_3Zr containing a small amount of chromium.

Fig. III.11. Schematic of the Experimental Metal Waste Form Sample Chamber and Assembly

Fig. III.12. Optical Metallograph of Chemically Etched HT9-5 wt% Zr Alloy (218X)

Following this initial set of HT9-Zr alloys, the next generation of alloy specimens will be HT9-15 wt% Zr with varying amounts of noble metal additions. The metals silver, palladium, and ruthenium will be used to simulate the behavior of noble metal fission products in the waste form alloy. Ruthenium is of special interest because of its expected similarity in alloying behavior to the fission product technetium, which is of concern because of the long half-life of Tc-99 (2×10^5 y).

b. Large-scale Experiment

This effort has involved the definition of an experimental system for a larger-scale demonstration of the steel-based waste form processing and the selection of primary candidates for the molten salt flux and extraction compounds.

(1). Melting Furnace Specifications

A high-temperature tilt-pout induction melting and casting furnace has been specified to melt and alloy 2.5 kg of simulated waste form materials under a molten salt flux. The experiments will demonstrate the large-scale extraction of depleted uranium from the waste form alloy using batch-type operations. The temperature requirements on the system are high. HT9 stainless steel ($T_m \approx 1550^\circ\text{C}$) and zirconium ($T_m = 1855^\circ\text{C}$) must be melted and alloyed together. Although zirconium will dissolve into molten steel according to the Fe-Zr phase diagram (Fig. III.13), the kinetics of the dissolution are not known. Therefore, the furnace should be capable of melting zirconium metal. In addition to the temperature and furnace requirements, the furnace must also interface with an inert atmosphere glovebox because the salt charge and some of the metal charge materials are reactive in air. The configuration of the glovebox and furnace is shown schematically in Fig. III.14.

(2). Molten Salt Flux Selection

Chloride salts have promising actinide extraction properties on the basis of the relative free energies of formation of actinide chlorides as compared to the nobler metal chlorides that could form with the waste form constituents. However, the salt selection is limited because of the high temperatures required to melt the waste form alloy and the high vapor pressures of chloride salts at these temperatures. The minimum liquidus temperature of the molten metal will be very dependent upon the zirconium content in the alloy, but it could be as low as 1300°C with approximately 15 wt% Zr for the IFR waste form alloy and as low as 950°C with approximately 85 wt% Zr for the LWR waste form alloy based upon eutectics in the Fe-Zr binary system (Fig. III.13). The salt extraction will be carried out in the presence of molten metal.

Table III.8 compares the free energy of formation of the actinide chlorides and some noble metal waste form constituent chlorides with several candidate (the salts are listed in order of decreasing stability). Figure III.15 shows the relative vapor pressures vs. temperature of the candidate chloride salts and their relationship to the estimated operating parameters of

G10104

Fig. III.13. Iron-Zirconium Phase Diagram

Fig. III.14. Schematic Diagram of Casting Glovebox with Furnace Location Indicated (hatched areas)

TABLE III.8. Free Energy Formation of Chlorides at 725°C (Kcal/g-atom of chlorine)

Actinides		Noble Metals		Candidate Salts	
				BaCl ₂	-83.0
				CaCl ₂	-76.6
AmCl ₂	-62.6				
AmCl ₃	-60.9				
PuCl ₃	-58.6				
				MgCl ₂	-58.0
NpCl ₄	-54.1				
UCl ₃	-51.5				
		ZrCl ₄	-45.1		
		AlCl ₃	-42.5		
		McCl ₂	-42.3	MnCl ₂	-42.3
		CrCl ₂	-32.5	CrCl ₂	-32.5
		TaCl ₅	-28.0		
				CuCl	-26.7
		AgCl	-19.3		
		NiCl ₂	-18.7		
		WCl ₆	-14.3		
		MoCl ₃	-12.8		
		PdCl ₂	-6.0

Fig. III.15. Vapor Pressure vs. Temperature of Fluxing and Oxidizing Chlorides

the system. The vapor pressures are shown for the entire liquid phase field of the pure compounds. Chloride vapor pressure data does not have a high degree of certainty, but two sources were used[7,8] to provide a best approximation. The data are used as a guide with the understanding that all conclusions must be validated experimentally in the future.

The ideal extraction salt would have low vapor pressure in the fluxing temperature range and a relative free energy of formation such that the actinides would be oxidized and the waste form constituents would remain in the melt. As in most real systems, the physical situation does not match the ideal requirements, but an alternative is possible. Calcium chloride and barium chloride are very stable, low vapor pressure salts which are candidates for a fluxing salt solvent. Calcium chloride has the lower vapor pressure of the two compounds (Fig. III.15), making it more suitable for the process. The other listed candidate salts are potential extraction agents which would be added to the solvent salt. One of the issues to be resolved by process and equipment design is the containment of the extracted chlorides, the most volatile being ZrCl_4 .

Based upon this theoretical comparison, the most promising combination identified so far is CaCl_2 as a fluxing solvent with a controlled amount of CuCl in the salt for chemical extraction of the actinides. The quantity of CuCl must be carefully controlled because the zirconium in the melt will begin to be extracted once the actinides have been oxidized and the melting temperature of the alloy is sensitive to the zirconium concentration. A preliminary process diagram for the steel waste form is shown in Fig. III.16. It indicates CaCl_2 as the salt flux and CuCl as the oxidant, and it shows the salt flux being recycled by adding Ca metal to the TRU-laden salt with the resulting TRU metal returning to the electrorefiner.

Fig. III.16. Preliminary Process Diagram for Steel Waste Form Generation

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IV. SAFETY EXPERIMENTS AND ANALYSES

A. Eutectic Formation and Wastage in Metallic Fuel

This section summarizes the technical facts concerning eutectic formation and discusses their significance for both reactor safety and for fuel pin design criteria. It is shown that during steady-state irradiation solid-state interdiffusion of the U-Pu-Zr fuel, together with the associated fission products, and the cladding constituents leads to a reacted zone which is regarded as wastage of the cladding. The amount of wastage is a strong function of local power, temperature and burnup. The affected surface fuel, while behaving no differently under normal operating conditions may reach its solidus temperature during elevated temperature off-normal events. The solid-liquid mixture that forms at the fuel clad interface is referred to as eutectic formation. In fact, in virtually all transient events eutectic formation affects only already wasted cladding.

1. Fuel Pin Phenomenology Database

During steady-state irradiation, solid-state interdiffusion of the U-Pu-Zr fuel (including fission products) and cladding constituents occurs across the fuel/cladding interface after tight fuel/cladding contact is made. The reacted cladding band, which contains lanthanide fission products and fuel and cladding constituents, is regarded as a normal wastage of the cladding. Concurrent with this process is the migration of cladding constituents (mainly Fe, and in the case of austenitic steel cladding, Ni) into the surface region of the fuel. As can be expected, the extent of this interdiffusion depends strongly on the history of local fuel linear power, cladding temperature, and time, i.e., burnup. The affected surface fuel, while behaving no differently under normal operating conditions, may reach its solidus temperature during elevated-temperature off-normal reactor events. The solid-liquid mixture that thus forms at the fuel-clad interface is hereinafter referred to as eutectic formation.

To determine the fuel and cladding behavior under transient conditions, a large number of ex-reactor tests, using the Fuel Behavior Test Apparatus (FBTA) and Whole Pin Furnace (WPF) at ANL, have been conducted with

EBR-II-irradiated fuel pins. In brief excursions (with durations on the order of seconds to minutes, e.g., design basis transients), no eutectic formation was detected even in "worst-case" fuel pins (i.e., those pins which had experienced high power, high cladding temperature, and high burnup during steady state irradiation)* at transient temperatures up to $\approx 775^{\circ}\text{C}$ (1427°F). Alternately, in tests with longer durations (on the order of hours to days, simulating certain classes of beyond design basis events) eutectic formation occurred in the worst-case pins at test temperatures around $660\text{--}675^{\circ}\text{C}$ ($1220\text{--}1247^{\circ}\text{F}$). For fuel pins irradiated under less aggressive conditions, the threshold temperature for eutectic formation was approximately 50°C higher. (It should be noted that these test pins were irradiated in EBR-II, where the aggressive conditions of high temperature and high power often occur simultaneously near the top of the fuel due to the reactor's relatively flat axial power profile. This is unlikely to be the case in an ALMR with a steeper power profile.)

In an elevated-temperature excursion, the fuel and cladding materials undergo additional interdiffusion. If eutectic formation occurs, the affected fuel surface layer, in the form of a solid/liquid mixture, may consume part of the original solid-state interdiffusion band in the cladding. If the event is prolonged, the reaction may then progress further, and into the previously unaffected cladding.

Regardless of how the wastage progresses, the relevant quantity is the remaining thickness of unaffected cladding available to carry the loading. In the ex-reactor tests, this quantity was determined for EBR-II-irradiated fuel samples which spanned a wide range of fuel compositions, cladding materials, irradiation power, temperature, and burnup. A conservative correlation for cladding wastage rate versus temperature has been derived from these data, and is being used for both the EBR-II and the ALMR core design. While the preirradiation conditions apparently affect the eutectic threshold temperature,

*The peak power, cladding temperature and burnup of these pins were 15 kW/ft, 600°C and 11 at.%, respectively, during steady-state irradiation. In comparison the projected operating conditions for ALMR fuel are 10.1 kW/ft, 565°C and 15 at.%, and these conditions are not present at the same location as they were in the EBR-II test pins.

the rate of cladding wastage during off-normal events appears to be essentially independent of the preirradiation conditions.

The following two tables, Tables IV.1 and IV.2, describe key Fuel Behavior Test Apparatus and Whole Pin Furnace experiments which summarize the current database on the phenomenology of eutectic formation and wastage in metallic fuels.

2. ANL Fuel Pin Design Criteria and Basis

The knowledge of phenomenology obtained from the testing database discussed above form a basis on which to specify design criteria which if met, will assure that the Mk-V and Mk-VA driver fuel for EBR-II will perform in a safe and reliable manner. The same database can be extended to provide for a statistically significant database to support ALMR designs. The two cladding materials that have been selected for the Mk-V and Mk-VA fuel pins are tempered martensitic alloy HT9 and austenitic alloy Type 316 stainless steel, respectively. The cladding damage mechanisms that govern pin performance depend on a few key physical phenomena. These phenomena are 1) mechanical deformation of the cladding, 2) steady-state cladding wastage, and 3) eutectic formation.

The ability of the cladding to function as a barrier to the release of radioactive materials depends on the inherent strength and thickness of the cladding material and on the applied loadings. The two physical properties which represent inherent strength that are used directly for design criteria to assess cladding failure by calculation are the cladding plastic strain correlations and the stress rupture correlations. The cumulative damage function (CDF) method is used to apply the stress-rupture correlations to variable load and temperature conditions. It should be noted that above 650°C (1200°F) the inherent strength of HT9 becomes very weak, with creep rupture times of tens of hours under applied loads as shown in Fig. IV.1.

The loading on the cladding is principally the result of a hydrostatic internal gas pressure from released fission gas in the interconnected porosity in the fuel column and the plenum. The gas pressure loading on the cladding of Mk-V/VA fuel pins has been significantly reduced relative to previous

TABLE IV.1. Summary of Key Fuel Behavior Test Apparatus Results*

Steady-state Irradiation				Ex-reactor Testing			
Test No.	Peak BU (at.%)	Peak Power (kW/ft)	Peak Cladding Temp. (C)	Temp. (C)	Duration (h)	Cladding Wastage (mm)	Eutectic Formation (Fuel Areal %)
Effect of Test Temperature							
91-19	11	15	600	650	1	0	0
91-31	11	15	600	675	1	24	5
91-17	11	15	600	700	1	23	13
91-16	11	15	600	770	1	31	66
Effect of Test Duration							
91-19	11	15	600	650	1	0	0
91-32	11	15	600	650	12	12	0
91-35	11	15	600	650	36	55	0
91-31	11	15	600	675	1	24	5
91-34	11	15	600	675	12	109	31
Effect of Irradiation Linear Power							
91-31	11	15	600	675	1	24	5
92-11	13	13	600	675	1	0	0
Effect of Irradiation Cladding Temperature							
91-31	11	15	600	675	1	24	5
92-02	11	15	475	675	1	29	0
Effect of Fuel Burnup							
91-03	6	15	600	738	1	0	0
91-15	11	15	600	740	1	46	18
Effect of Cladding Material							
91-31	11	15	600	675	1	24	5
92-07(D9)	11	15	600	675	1	0	0
91-17	11	15	600	700	1	23	13
92-10(D9)	11	15	600	700	1	7	4

*All samples with U-19wt.% Pu-10wt.%Zr fuel and HT9 cladding, unless otherwise noted.

TABLE IV.2. Summary of Whole-pin Furnace Tests with Irradiated Fuel Elements

Test ID	Fuel/Cladding Materials	Element No./SA	Cladding OD/ID (mm)	Plenum/Fuel Vol. Ratio ^a	Linear Power (kW/m)	PICT (°C)	Peak Fuel Burnup (at.%)	Test Temp. (°C)	Test Duration (min)
<u>Cladding Integrity Margin Tests</u>									
FM-1	U-10Zr/HT9	T418/X425	5.84/5.08	1.0	39.4	594	3.0	820	67 ^b
FM-2	U-19Pu-10Zr/HT9	T461/X425	5.84/5.08	1.0	39.4	594	3.0	820	112 ^b
FM-3	U-19Pu-10Zr/HT9	T680/X430A	7.37/6.55	1.4	42.6	555	2.2	820	146 ^b
FM-4	U-19Pu-10Zr/HT9	DP-22/X441A	5.84/5.08	1.5	49.2	600	11.3	770	68 ^c
<u>Element Survivability Tests</u>									
FM-5	U-19Pu-10Zr/HT9	DP-15/X441A	5.84/5.08	1.5	49.2	600	11.4	780	3 ^d
FM-6	U-19Pu-10Zr/HT9	DP-39/X441A	5.84/5.08	1.0	49.2	600	11.3	650-670	2160 ^e
FM-7	U-19Pu-10Zr/316SS	T-539/X429B	5.84/5.08	1.0	39.4	600	13.3	650-670	2160 ^d

^aAs-fabricated fuel length = 343 mm; as-fabricated fuel diameter = 4.4 mm for all elements except FM-3 with a 6.57-mm fuel diameter.

^bCladding breaching time during temperature hold at 820°C; breach occurred near the top of fuel.

^cCladding breaching time during temperature hold at 770°C; breach occurred in plenum.

^dDuration of the enveloping transient proof test; no noticeable damage incurred in the fuel element.

^eTotal test duration at a limiting cladding temperature for EBR-II; fuel element incurred a peak incremental cladding strain of 0.84%, but no eutectic reaction and no cladding breach.

^fFuel pins incurred a pin-hole breach at the end of the test. No eutectic reaction.

Fig. IV.1. Failure Time as a Function of Cladding Temperature

EBR-II driver fuel and into the range of the ALMR by using a large plenum and restricting the burnup. ALMR fuel designs have an increased plenum size and a low pressure at goal burnup.

An important factor determining cladding load bearing thickness is cladding wastage. Cladding wastage is defined here as that part of the original cladding wall thickness which is assumed to no longer contribute to carrying the applied loading. Mechanisms identified in the phenomenology tests as contributing to cladding wastage in the Mark-V/VA fuel and cladding systems are: on the outer surface 1) scratches and 2) sodium-induced erosion; on the inner surface 1) scratches, 2) solid-state diffusion of lanthanide fission products and fuel constituents into the cladding, and 3) eutectic formation at the fuel-cladding interface. A graphical representation of the various zones that may form in the cladding and the remaining load-bearing ligament is shown in Fig. IV.2.

Fig. IV.2. Possible Cladding Wastage Mechanisms

There are three basic criteria applied for assessment of transient performance of the Mk-V/VA fuel. These criteria are CDF-based limits, strain limits and a limit on the amount of eutectic formation. These first two are conventional, widely-accepted means of evaluating cladding damage for the materials being used in IFR cladding. Inherent in their use is acceptance of the idea that the key safety function is cladding integrity. The last is derived from current EBR-II limits and is motivated not by cladding load bearing ligament issues but rather to preclude fuel motion through limits on the amount of eutectic that can be formed. *Steady-state* operation is limited to temperatures below the threshold for eutectic formation.

Even though the data show that the eutectic threshold temperature is higher for fresh fuel, in performing the safety evaluation for Mk-V driver fuel in EBR-II, a eutectic threshold temperature of 1200°F (650°C) has been applied irrespective of burnup, cladding type or exact quantity of plutonium or other

actinides in the fuel. The reason for adopting this position is in order to bound the effects of fission product (especially lanthanides) and actinide carryover which will be experienced in currently unknown amounts when the fuel is recycled through the pyroprocessing and the recycled fuel returned to EBR-II,--i.e., to set conservative criteria prior to the availability of data which are to come from the Mk-V fuel irradiations themselves. (The lanthanides appear to play a role in cladding wastage and therefore this assumption is adopted to ensure that the process is treated conservatively).

While this conservative eutectic threshold temperature has been adopted for the reasons given above, it is clear that for both physical phenomena which cause wastage (solid-state or eutectic interaction) reduced time at temperature will reduce the degree of cladding wastage. Thus, consistent with FFTF and CRBR practice, the ANL design criteria for Mk-V pins employ time at temperature based cladding deformation criteria--such as damage fraction criteria--rather than time invariant temperatures as the basis for fuel pin design.

3. Future Work

The steady-state cladding wastage correlation which is in current use for the HT9-clad Mk-V fuel analyses is shown in Fig. IV.3 along with the database upon which it is based. A conservative rather than best fit correlation is used in view of the paucity of data and in view of approximations which have been necessitated to produce the data points from the irradiation measurements. This area (the steady-state wastage phenomena) and its dependencies on pin operating parameters, including temperature history and local linear heat rating--both vs. burnup; initial lanthanide concentration in the fuel (to accommodate carryover from pyroprocessing); choice of cladding material (ferritic vs austenitic); and smear density, is currently the area of greatest technical uncertainty in the Mk-V/VA pin design.

The IFR fuels testing program has as one of its objectives to develop the understanding and robust database to fully characterize the dependencies of wastage phenomena upon pin design choices and operating conditions--including the effects of lanthanide carryover in pyroprocessing. Investigations are underway

Fig. IV.3. Steady-state Cladding Wastage Correlation for HT9 Clad Fuels

to characterize the dependence of the steady-state wastage and eutectic threshold temperature upon irradiation temperature of cladding, local power and hence temperature gradient in the fuel and burnup.

While it has been shown that a 1200°F (650°C) eutectic formation threshold temperature does not present a problem for transient fuel pin behavior, it does indeed represent a constraint upon steady-state operation. Although both EBR-II and ALMR have a peak fuel/cladding interface temperature (with uncertainties) of less than 1200°F, other designs may wish to utilize higher outlet temperatures than these reactors. In these circumstances a eutectic threshold temperature of 1200°F may be a real constraint. This constraint can be alleviated in two ways. Firstly, the understanding of the details of eutectic formation and steady-state wastage may enable a burnup-dependent value for the eutectic threshold to be adopted which will allow higher coolant outlet

temperatures in view of the fact that the high temperatures occur early in life when the threshold is higher and the fission gas pressure is low. The fact that ALMR fuel is not likely to have axial positions with coincident high power, cladding temperature and burnup will likely allow relaxation of current restrictions. Secondly, the use of alternative cladding material or design (e.g., barriers) could be considered. For example, D9 apparently exhibits a higher eutectic threshold temperature but at an apparent price of higher steady-state wastage and greater irradiation-induced swelling; the effects of these differences upon fuel pin performance require investigation.

B. Inherent Nonenergetic Termination of the Initiating Phase of Severe Accident Initiators

The developments in the IFR program have resulted in a reactor system which does not sustain any core damage when subjected to a variety of accident initiators, including unprotected, i.e., unscrammed, accidents. The developments include the use of a metallic fuel, the design of low burnup reactivity swing cores to minimize the reactivity vested in the control rods, and the design of the reactor to take full advantage of the available passive reactivity feedback mechanisms. Previous analyses and experiments have demonstrated that minimum margins to coolant boiling of 200°C or greater during an accident transient can be obtained, even for unprotected accidents which are classified as "double-fault" where there is a failure of a major engineered system along with a failure to scram the reactor.

Since it was clear that the accident initiators which might have resulted in coolant boiling, fuel melting, and fuel pin failures in earlier (non-IFR) LMR designs would not produce any serious consequences in an IFR, the severity of accident initiators was increased in order to investigate the possibility of causing such phenomena in reactor cores using the IFR concept. The resulting accident consequences were calculated using the SAS4A Accident Analysis Code,[11] which has been modified to allow analysis of such accidents in reactor cores which use metallic fuel. For the unprotected loss-of-flow (LOF), if it were assumed that there was no coastdown of the flow, but an almost immediate stoppage, it was possible to achieve severe accident conditions such as coolant boiling and fuel melting. This unprotected "rapid" LOF could correspond to a

simultaneous seizure of all coolant pumps, which might be a consequence of a very large seismic event. For the unprotected transient overpower (TOP), which is normally assumed to be caused by the inadvertent withdrawal of a control rod from the reactor, it was assumed instead that a series of control system failures causing all of the control rods to withdraw from the reactor occurred with a failure to scram. The resulting multiple rod withdrawal accident also leads to severe accident conditions in the core. Both of these accidents are very unlikely, with an extremely low probability of occurrence, even when compared to the probability of 1×10^{-6} per year associated with the "double-fault" events.

1. Unprotected "Rapid" Loss-of-flow Accident

The unprotected "rapid" LOF is initiated by a loss of electric power to all coolant pumps, along with seizure of the pumps to prevent the normal flow coastdown. There is also a failure to scram the reactor. The flow through the core decreases rapidly, reaching 50% of nominal at about 160 ms after the start of the accident, and below 10% by 1.5 s. The subsequent behavior is dependent on various core characteristics, including the balance of the reactivity feedback.

For reactor cores with a low reactivity burnup swing and a correspondingly high sodium void worth, the power remains near nominal until the onset of coolant boiling. Sufficient negative reactivity feedback is generated by axial fuel expansion, fuel Doppler, and radial core expansion to counter the positive reactivity feedback caused by the decrease in coolant density in the core as the core temperatures rise. Initial coolant boiling occurs generally at 2-3 s after the start of the transient. The additional positive reactivity feedback associated with boiling slowly increases the core power. This in turn causes boiling to spread more rapidly to other core assemblies, adding more positive reactivity and increasing the rate of power rise. The increasing power and core temperatures lead to fuel melting along the centerline of the fuel pins of the highest power assemblies. There is little reactivity effect associated with fuel melting until the molten fuel region reaches the top of the fuel, with the fuel pin cladding still intact. At that time, the molten fuel expands axially within the pin, driven by the difference in pressure between the fission gas plenum and the fission gas retained in the fuel. The pressure of the fission

gas retained in the fuel is significantly higher than the pressure in the fission gas plenum due to the much higher temperature in the fueled region of the core and the reduction in space available for the retained fission gas due to the decrease in fuel density. The in-pin fuel motion introduces substantial negative reactivity feedback to limit the rise in power. The initial in-pin fuel motion usually occurs within 0.5 to 1.0 s after the start of boiling, with the peak power limit dependent on various modelling parameters associated with in-pin fuel motion. For reactor cores such as PRISM Mod B/93 and the higher void worth cores used in the sodium void worth/reactivity burnup swing tradeoff study, peak power is in the range of 3-15 times nominal.

In-pin fuel motion introduces negative reactivity feedback to counteract the positive feedback from coolant boiling. If boiling continues to spread so that the power does not drop rapidly, in-pin fuel motion occurs in additional assemblies until sufficient negative reactivity is introduced to reduce the power and stop the rise in fuel temperatures in the core. The first fuel pin failures occur within a few tenths of a second after in-pin fuel motion begins. The failure location is at approximately two-thirds of the core height. As the cladding breaches, the molten fuel in the pin moves toward the failure location, which introduces slight positive reactivity initially. Once the fuel starts to disperse in the channel, the substantial negative reactivity associated with the fuel loss from the core takes the net reactivity subcritical, terminating the initiating phase of the accident. The fuel temperatures peak near 1600°C, just prior to fuel pin failure, and then decrease as the fuel disperses. The temperatures are not high enough to cause fuel vaporization, and there is no mechanism for the intermixing of molten fuel and liquid sodium in the quantity required to vaporize a substantial amount of the remaining liquid coolant. This implies that the energetics potential for this accident is extremely small.

The phenomena encountered for cores with low or negative sodium void worth are similar, except that in-pin fuel motion does not occur. This is due to the relatively unchanging power in the core as the coolant boils, since coolant boiling has little reactivity effect, and fuel pin failures occur at or below nominal power. The end result is quite similar, with peak fuel temperatures 200-400°C lower, and no energetics potential.

2. Unprotected Multiple Control Rod Withdrawal

The unprotected multiple control rod withdrawal is initiated by a series of control and protection system failures which leads to all of the control rods being withdrawn from the core and a failure to scram. The coolant pumps continue to operate, providing normal flow rate through the core. The steam generator also continues to function. For the purposes of the analyses, it is assumed that the rod withdrawal causes reactivity to be added to the core at a rate of $0.05\$/s$, conservatively higher than the expected reactivity addition rate associated with the maximum rod withdrawal speed. The core power and temperatures begin to rise as the control rods are withdrawn from the core. The increasing core temperatures generate reactivity feedback to partially compensate for the reactivity addition associated with removing the control rods. The power rise is relatively slow, achieving twice nominal power at about 10 s after the start of the accident. Initial in-pin fuel motion occurs just above 2.5 times nominal power. For this accident, the in-pin fuel motion contributes only minor negative reactivity feedback, since the temperature and pressure in the fission gas plenum region is comparable to the temperature and pressure within the fueled region of the core. As such, the net effect of the in-pin fuel motion is only to slow the rate of power rise. The initial fuel pin failures occur at the top of the fuel column, with power at approximately three times nominal. The reactivity effect of the fuel leaving the pin is negative and large, with the fuel dispersal causing the power to drop to nominal power or below, depending on the reactor.

For reactor core designs with low burnup reactivity swing, but correspondingly high sodium void worth, the total amount of reactivity vested in the control rods is small. With design uncertainties included, total rod worths at nominal operating conditions are in the vicinity of 1.5–2.0\$. Fuel pin failure in the pins of as few as three assemblies results in shutdown of the reactor, and benign termination of the initiating phase. As the sodium void worth is decreased, the burnup reactivity swing increases rapidly, such that the total control rod worth of low-void-worth core designs can be 10–15\$. The behavior of the low sodium void worth cores is similar to that for the low burnup swing cores, except that pin failures in more assemblies are required to shut down the reactor. The initial pin failures will occur as in the high sodium

void worth core, but there is significantly more reactivity to be added from the withdrawing control rods. This causes another power rise, terminated by more pin failures. The process repeats until sufficient negative reactivity has been obtained from fuel dispersal in the coolant channels to overcome the total positive reactivity addition from removing the control rods. However, the end result is still acceptable, since the performance of metallic fuel in this accident favors early fuel pin failure and fuel dispersal in the channel, without generating excessive fuel temperatures. As with the unprotected "rapid" LOF, no energetics should occur with this accident, with benign termination of the initiating phase.

3. Summary

The analyses of the reactor response to both unprotected "rapid" LOF and multiple rod TOP accidents indicates a benign termination of the initiating phase, with little likelihood of energetics. These results are due to the favorable performance of metallic fuel, with a relatively low melting point, the ability to expand axially within intact cladding prior to cladding failure, and the dispersive character of the fuel motion after fuel pin failure. The molten fuel is also compatible with the sodium coolant, in that the fuel melting point is lower than or comparable to the coolant boiling point, and the fuel does not chemically react with the coolant. These results are due also to an overall favorable reactivity feedback balance, even with core designs with higher positive sodium void worth, which is a result of using metallic fuel and of designing the reactor system to take full advantage of the available favorable passive feedback mechanisms.

C. Whole-pin Furnace Tests Performed to Support EBR-II Mk-V Fuel Safety Case

Conversion of the EBR-II core to one containing HT9-clad ternary U-20Pu-10Zr alloy fuel, designated the Mk-V fuel, is the next step in the development of metal fuels for the IFR fuel system. This Mk-V fuel will be manufactured at the Fuel Cycle Facility currently being outfitted at Argonne-West. A number of Whole-pin Furnace (WPF) tests have been conducted to specifically support the safety case for the Mk-V conversion. Results from the tests have contributed to the database of knowledge regarding the behavior of this fuel during normal

operation and off-normal transients. The principal objectives of the tests were to (1) demonstrate fuel performance capability during the most-severe EBR-II unlikely loss-of-flow (LOF) event (UN-1), (2) determine the safety margin of the fuel pin, (3) investigate cladding breaching behavior, and (4) provide data for validation of the FPIN2 and LIFE-METAL codes.[1-2] (These codes are playing a key role in calculating the neutronic, thermal, and mechanical response of Mk-V-type fuel pins during both normal operation and off-normal transients.)

1. Whole-pin Furnace System Description and Experimental Procedure

The WPF system[3] is in operation in the Alpha-Gamma Hot Cell Facility (AGHCF) at Argonne National Laboratory to simulate the response of fuel pins to reactor transients by heating intact, irradiated fuel pins in a computer-controlled infrared radiant furnace. Its major components are an in-cell furnace rig and test capsule, a furnace control and data collection system, and a fission-gas measurement system. Figure IV.4, a schematic representation of the in-cell components, shows the stainless steel test capsule containing the irradiated fuel pin. The capsule is normally evacuated and sealed during the

Fig. IV.4. Schematic Representation of Whole-pin Furnace System

test. Two pressure transducers at the top of the capsule provide the means for cladding breach detection by measuring the pressure rise due to fission-gas release into the sealed system. The temperature along the length of the fuel pin is measured by six sheathed, chromel-alumel thermocouples located at different elevations in the annulus between the fuel pin and the capsule. A ceramic-insulated, bare-wire Pt/Pt-10%Rh control thermocouple, which is welded on the outside of the capsule, serves to maintain desired test temperatures.

The furnace is a radiant heating chamber powered by four longitudinal tungsten-filament lamps. Elliptically shaped aluminum reflectors behind each lamp focus the radiant energy onto the centerline of the furnace, where the capsule and test fuel pin are located. Control of the furnace is based on the output of the control thermocouple via a feedback algorithm in a microcomputer. Typical accuracy in an extended heating cycle (up to 36 h) is within $\pm 2^{\circ}\text{C}$ of the target temperature. Properly locating the fuel pin axially within the heating zone of the furnace allows a good simulation of the fuel pin's axial temperature profile in EBR-II.

The conditions of the test pins are characterized at the Hot Fuel Examination Facility (HFEF) and AGHCF before and after the tests. The pretest examination typically includes gamma scanning and/or neutron radiography to determine the length of the fuel column and measurement of diameter to determine cladding deformation. The posttest examination also includes metallography to determine fuel/cladding metallurgical interaction. The fission gas released from a breached fuel pin is collected to evaluate release characteristics.

2. Test Descriptions and Results

The reference Mk-V fuel pin design uses a U-20Pu-10Zr metallic fuel slug with HT9 cladding. A design variation (Mk-VA) with Type 316 stainless-steel cladding is also being included in the safety case. The interim goal burnup specified for the Mk-V fuel is 10.0 at.% (peak). A comparison of the design and operating parameters of the Mk-V/VA fuel with those of the pertinent WPF test pins is given in Table IV.3.

Five tests were conducted in the WPF to support the Mk-V conversion safety case. The fuel pins that were tested differed from the reference Mk-V

TABLE IV.3. Comparison of As-fabricated Fuel Pin Parameters and Burnup

Parameter	Mk-V/VA	FM-2	FM-4/FM-5	FM-6/FM-7
Element No./Subassembly	-----	T461/X425	DP22/X441A DP15/X441A	DP39/X441A T539/X429B
Fuel Type	U-20Pu-10Zr	U-19Pu-10Zr	U-19Pu-10Zr	U-19Pu-10Zr
Cladding Type	HT9/316SS	HT9	HT9	HT9/316SS
Smear Density, %	75.0	75.0	75.0	75.0
Cladding OD, mm	5.84/5.84	5.84	5.84	5.84
Cladding ID, mm	4.93/5.08	5.08	5.08	5.08
Plenum/Fuel Volume Ratio	1.5	1.0	1.5	1.0
Linear Power, kW/m	33-49	39.4	49.2	49.2/39.4
PICT,* °C	580	594	600	600
Peak Fuel Burnup, at. %	10	3.0	11.3/11.4	11.3/13.3

*Peak Internal Cladding Temperature.

fuel in several of the parameters listed in Table IV.3; however, these differences, i.e., cladding thickness, plenum-to-fuel volume ratio, and burnup, led to tests that would yield conservative estimates of the safety margin for the Mk-V fuel.

a. Proof test: FM-5

The FM-5 test was a proof test performed to demonstrate that a high-burnup, Mk-V-type fuel pin can survive a simulated worst-case unlikely LOF event in EBR-II. The LOF event chosen for temperature simulation was the UN-1 event, which results in the highest cladding temperatures for the longest duration among the various design-basis transients. The thermal conditions of the test were designed to envelope the cladding temperature history in a UN-1 LOF event with a peak instantaneous cladding temperature of 776°C, which then decreases rapidly to 660°C in ~ 15 s. Figure IV.5 shows the cladding temperature histories at the top of the fuel column during a UN-1 event and during the FM-5 test.

Fig. IV.5. Time/Temperature History of the FM-5 Proof test Showing Simulation of the UN-1 Loss-of-flow Event in EBR-II

The fuel pin endured the simulated UN-1 temperature transient without cladding breach and with negligible incremental cladding deformation. A detailed thermal analysis based on measured thermocouple temperatures established that the top portion ($0.86 \leq X/L \leq 1.0$) of the fuel pin was actually heated above 660°C for ~ 40 s during the test, more than twice as long as during a UN-1 event. Posttest metallographic examination of the fuel pin regions that experienced the highest temperature showed no evidence of fuel melting or fuel/cladding eutectic interaction, as shown in Fig. IV.6. The measured cladding wastage was comparable to that of steady-state sibling fuels, indicating no incremental cladding penetration during the test.

b. Run-to-cladding-breach Tests: FM-2 and FM-4

The FM-2 and FM-4 tests were conducted to evaluate the safety margin of low- and high-burnup Mk-V fuel pins at temperatures on the order of that in the UN-1 event. The FM-2 test with a 3-at.-%-burnup element was conducted at a constant plenum temperature of 820°C with the cladding at the top of the fuel approximately 10°C less. The fuel pin breached in 112 min. The FM-4 test, with a 11.4-at.-%-burnup fuel pin, was conducted at a plenum temperature of 770°C , approximately that of the peak temperature in the UN-1 event, with the cladding

Fig. IV.6. Longitudinal Section Near Fuel Top ($0.94 \leq X/L \leq 0.99$) of FM-5 Fuel Pin, Showing Low-density Fuel Near Top, Fuel Restructuring, and Alloy Constituent Redistribution, but No Evidence of Eutectic Formation

at the top of the fuel approximately 20°C less. The FM-4 pin breached in 68 min. Compared to the total duration of the UN-1 LOF event (≤ 2 min), see Fig. IV.5, both the FM-2 and FM-4 tests demonstrated very significant safety margins for cladding breaching for Mk-V-type fuel pins.

The FM-2 fuel-pin cladding breached near the top of the fuel in the form of a small axial crack ~ 2.0 mm long, with a 0.9-mm opening at the widest point. A transverse section near the mid-height of the crack is shown in Fig. IV.7. Fuel/cladding metallurgical interaction had reduced the cladding to a thin membrane near the crack tip and was prominent in other transverse sections at different elevations as well. Fission-gas pressure apparently caused the

Fig. IV.7. Cladding Breach Near Fuel Top in Low-burnup, Mk-V-type Fuel Pin in FM-2 Test

final rupture of the thin membrane. The rupture was benign, as indicated by the crack dimensions and relatively small diametral strains (2.3 to 4.3%) near the crack tip. Some molten eutectic was ejected from the pin into the capsule where it rapidly solidified from the furnace shutdown. The fuel/cladding interaction led to eutectic formation (i.e., phase transformation from solid to solid-plus-liquid) and eventual fuel foaming. The areal fraction of fuel/cladding eutectic-type formation was $\sim 11\%$ at $X/L = 0.54$ and increased to 100% at $X/L = 0.68$ and above.

In contrast to the benign cladding breach in the fuel-column region of the low-burnup FM-2 pin, the high-burnup FM-4 pin breached in the plenum region ~ 150 mm above the fuel top, with significant cladding deformation (up to $\sim 15\%$ strain close to the breach). The breach, shown in Fig. IV.8, with a length of 12.5 mm and a width of 3.5 mm was a much larger rupture than the FM-2 crack. Examination of the breached-cladding region showed no evidence of fuel and, therefore, no fuel/cladding metallurgical interaction at this location. The breach was a classical creep rupture of HT9 cladding caused by fission-gas

Fig. IV.8. Cladding Breach in Plenum Region of High-burnup, Mk-V-type Fuel Pin in FM-4 Test

pressure loading alone. Far less fuel/cladding metallurgical interaction occurred in the high-burnup FM-4 element than in the FM-2 pin, apparently due to the lower temperature and shorter duration. Maximum fuel/cladding eutectic formation was $\sim 20\%$ areal fraction at $X/L = 0.96$, where the temperature was 750°C during the test. The maximum cladding penetration at this elevation was $\sim 51\text{ }\mu\text{m}$, or 13.3% of the original cladding thickness.

c. Extended-duration Tests: FM-6 and FM-7

In contrast to earlier high-temperature, short-duration transient simulations, the FM-6 and FM-7 tests were conducted to evaluate the

extended-duration (36 h) behavior of Mk-V/VA-type fuel at intermediate temperatures (650°C at the fuel top and 670°C in the pin plenum). The thermal conditions of the tests were chosen to provide information (survivability, fuel-pin cladding strain, and eutectic formation) on a 650°C limiting cladding temperature of the Mk-V/VA fuels in EBR-II.

A HT9-clad, ternary-alloy fuel pin having 11.3 at.% burnup was tested in FM-6. Because of concern that the high-burnup fuel pin may breach prematurely during extended heating, the test was conducted in two stages: a 12- and 24-h test with interim nondestructive examination (contact profilometry and gamma scanning) of the fuel pin after the first 12-h heating period. The Mk-V-type fuel pin endured the 36-h test at 650-670°C without cladding breaching. Measured incremental strains during the test were 0.5% near the top of the fuel (650°C) and 0.9% in the plenum (665°C). Posttest metallographic examination of the fuel pin showed that there was no fuel/cladding eutectic formation although there was evidence of FCCI in the cladding.

Whole-Pin Furnace test FM-7 was conducted to support the safety case of the EBR-II Mk-VA fuel (U-20Pu-10Zr/316SS). The test evaluated the extended-duration (36 h) behavior of a 13.3 at.% burnup, U-19Pu-10Zr fuel pin with Type 316 stainless-steel cladding. The same heating conditions were used in the FM-7 test as in FM-6 (650°C at the fuel top and 670°C in the plenum); however, since the incremental cladding strains were expected to be relatively small, the fuel pin was heated without interruption for the entire 36 h.

Following the test when the fuel pin was removed from the test capsule, a small accumulation of bond sodium was observed on the exterior of the cladding ~ 75 mm below the top of the fuel column. A pin hole apparently formed late in the test since only a small amount of sodium exited the cladding, and there was no indication that fission gas was released during the test. Although a pin hole could be seen on the outside of the cladding once the sodium was removed, extensive metallography of the site provided no additional evidence of a breach in the cladding. There was no evidence of fuel/cladding eutectic formation; however, cladding wastage was greater in the FM-7 element than was observed in the FM-6 fuel pin.

D. SASSIM/EBR-II: A SASSYS-driven Interactive Simulator of EBR-II

Over the past several years, the number of research projects and plant upgrade efforts focused on the EBR-II facility has grown steadily. These include

- modern control developments,
- whole-system diagnostic software development and validation,
- control room hardware upgrades,
- plant protective system upgrades,
- sensor validation software development and testing,
- expert system development,
- formulation and testing of new procedures,
- human factors studies of man/machine interfaces, and
- development and safety analysis of transient materials tests.

Since using EBR-II on demand as a test bed for all these projects places a significant operational burden on the plant, interest has grown in developing an EBR-II interactive simulator to serve as an alternative test bed. An EBR-II simulator would also be useful to

- supplement current operator training, particularly in the area of response to severe off-normal transients, and
- provide an improved means of conducting safety analysis calculations, allowing interactive input preparation and output visualization.

Such an interactive simulator of EBR-II has been developed using two established analysis tools: SASSYS and GRAFUN.

The SASSYS[4] computer program has been developed over many years for use in safety analyses of liquid-metal-cooled reactor systems; it has now been adapted for use as a high-fidelity simulation engine. Several features of SASSYS make it an attractive simulation engine for an EBR-II simulator. First, the flexibility of the computer program allows the user to incorporate whatever degree of complexity is desired in the simulation, from very simple loops and core models which run quickly to very detailed modeling at the expense of longer running times. Second, the high fidelity of the models available in SASSYS make it possible to simulate a wide range of transients. Third, a significant amount of work[5] has already gone into validating the SASSYS EBR-II model against data

from EBR-II. Fourth, since the same SASSYS algorithms are used for the simulation engine as are used for EBR-II safety analyses, the modeling of EBR-II is consistent for all applications, and the simulator can provide a meaningful demonstration of a particular safety analysis. Finally, the fact that SASSYS has a control system model makes it straightforward to allow the user to interactively adjust limiting safety system settings and configure the plant protective system in much the same way as would be done in the actual plant. The only significant change made to SASSYS was to allow for exchange of data with another computer program during its execution in an interactive, rather than batch, mode.

The GRAFUN display management computer program was originally developed to allow diagrammatic display in real-time of data from the EBR-II Data Acquisition System (DAS). GRAFUN accomplishes some of its tasks by calling routines in the commercially available DataViews software package.[6-8] The GRAFUN computer program has been modified to (1) display the simulated EBR-II conditions as calculated by SASSYS and (2) provide the interface which allows two-way communication between the user and the simulation engine.

The user's interface to the simulator is a window under the X Window System on a color graphics terminal connected to a computer workstation running the UNIX operating system. An example of this window is shown in Fig. IV.9. The GRAFUN computer program manages this window and the two-way flow of data between the window and SASSYS. The data values to be transferred are stored in a segment of the computer's memory which is made available simultaneously to both SASSYS and GRAFUN. The mapping between the GRAFUN displays and the SASSYS variables is generalized so that if the component nodalization used by SASSYS is changed, GRAFUN automatically adjusts to the new nodalization.

The user may dynamically select from 20 different window views which display current-time data while operating the simulator. These are the same views which were created to view real-time EBR-II DAS data. Among these are piping and instrumentation diagrams of the primary (cf. main part of Fig. IV.9), secondary, and steam systems which display coolant flow rates and thermodynamic states (e.g., temperature and pressure); a core map of assembly outlet temperatures; and various neutron flux displays (e.g., digital by assembly,

Fig. IV.9. SASSIM/EBR-II Screen View Showing Primary Sodium System Quantities Computed by SASSYS

three-dimensional plot, and contour plot). In addition, information is provided simultaneously on core power, primary and secondary flow rates, core inlet and outlet temperatures, and the ratio of core power to primary sodium flow rate by three types of displays: current values on dials, current values on a kiviatt chart, and value histories on strip charts. There is also a view which provides an overall picture of conditions throughout the plant and includes heat balances and representative temperatures in each loop (i.e., primary sodium, secondary sodium, and steam), sodium loop flow rates, and reactor power. The user controls which one of these views appears on the screen at any time by using point-and-click mouse actions to select from the two columns of menu items shown on the upper left side of the window in Fig. IV.9. The user may interactively alter various control parameters (e.g., safety trip settings, reactivity insertion, pump speed) in SASSYS during the simulation by using point-and-click mouse actions to adjust scrollbars and select from menus in the window; several of these scrollbars and menus are shown in the main part and lower left corner of Fig. IV.10.

Fig. IV.10. SASSIM/EBR-II Screen View Showing SASSYS Case Control Quantities Adjustable by the User

The result of linking SASSYS and GRAFUN is a detailed simulation of the functioning of the EBR-II power plant which allows the user to monitor a wide range of plant parameters and to control the same parameters as would normally be controlled by operators in the actual plant. This simulator runs in real time with the latest SASSYS version (Release 3.0) on current generation computer workstations made by International Business Machines Corporation and Sun Microsystems, Incorporated. Actual calculation times vary with the complexity of the core model and the specific workstation (and its other load) upon which the calculation is run. Future work on the simulator will include expanding the amount of information displayed in the simulator views, adding views which more closely replicate current control room displays, and modifying views to reflect upgrades in the EBR-II control room. This software is adaptable for use with other reactor designs.

E. Transient and Long-term Creep Failure Data for HT9 Cladding

1. Introduction

In-service durability of the IFR cladding alloy HT9 is the concern of this research program. An important thrust of this work is to show how basic mechanical property data, the kind that is obtained in our laboratory and widely published in handbooks, can be used to resolve critical, but more arcane, issues associated with this reactor technology. For example, the results of static creep tests (constant stress and temperature) and tensile tests (constant rate and temperature) on HT9 are used to predict cladding failure behavior under complicated transient conditions involving off-normal thermal and mechanical loadings. Our goal is to develop the means to make reliable failure predictions and thereby avoid in-service cladding breach.

2. Background

Generally, creep, tensile and simple monotonic transient data are used to formulate rupture laws that can easily be incorporated into computer codes that follow the loading histories of complex thermal-mechanical transients. Failure calculations are then made by using the code to keep track of the service-induced damage accumulated in the cladding. The process is made complicated in the case of HT9 cladding by the inherent instability of the alloy's microscopic structure during high-temperature exposure as reflected in an instability in its mechanical properties during service. As a consequence, failure data reveal that two separate rupture laws with very different kinetic equations are obeyed over different time ranges, without much indication as to when to apply which law. This can lead to a serious uncertainty in cladding life prediction capability.

In the earlier work (reported in FY 1992 Annual Report for WBS 300), we addressed the above issue by analyzing published creep and tensile data, correlating the data with a microstructure-based flow model, and testing the model with complementary tensile data obtained in our laboratory. Two major accomplishments were obtained in that work.[9] First, a satisfactory correlation was derived for the intrinsic deformation behavior of HT9. This is the

deformation behavior during loading conditions (temperature, time, stress) under which the microstructure either does not change or, if it does, it does not affect plastic flow. Second, a semi-quantitative understanding was achieved of the relative strengthening of the material during long-time creep flow. The resulting model allows the quantitative prediction of the temperature-dependent shift to lower strain-rate under a given load, a reflection of the strengthening that occurs during long time deformation. We postulated that this strengthening is brought about by secondary carbide precipitation that causes hardening in an otherwise soft part of the HT9 microstructure. The validity of this hypothesis was established qualitatively by scanning electron microscopy. The microstructurally-caused difference between intrinsic short-time flow behavior and long-time creep flow behavior is analogous to the discrepancy between long-time and short-time rupture behavior to which we alluded above. Using the well-established empirical connection between high-temperature deformation and rupture life combined with the above model for creep strengthening, we were able to calculate the change in key rupture-life kinetic parameters. The results of these calculations agree well with measured values of these changes in rupture law parameters (i.e., the stress and temperature dependencies).

3. Current Research

a. Thrust of Current Research

The effort for this year (FY 1993) was directed at how we can use our new understanding of the relationship between microstructure and flow in HT9 to better predict its rupture behavior under relevant complex transient conditions. To accomplish this goal, two major research activities were carried out this year: One has involved efforts to refine the models discussed previously.[10-11] This work includes performing additional tensile tests focused on the appropriate flow regime. The second major activity has involved an endeavor to develop a base of microstructural data to increase our understanding of the physical processes that occur during high temperature deformation and fracture. This information will be of use as an aid to future, more mechanistic, modeling efforts.

b. Research Activity

The strengthening effect discussed earlier involves precipitation that develops over time in a complicated way. Because of this, a mechanistic calculation of rupture life that involves modeling this microstructural change will be very difficult. It is certainly worth doing, but we presently lack the supporting data necessary for this task. However, the separate long-time and short-time rupture correlations mentioned earlier, though quite distinct, are each valid within their own respective regimes. Therefore, in this year's effort, we focused on developing a capability to determine which of these two rupture correlations to use under a given set of loading conditions.

In earlier work, we used the connection observed between high temperature creep flow and rupture life. As a corollary to that relationship we have postulated that the creep strengthening effect observed in HT9 at low rates represents a state of the material associated with the long-time rupture law. Conversely, the intrinsic flow behavior observed at high rates reflects a state of the material associated with the short-time rupture law. Under conditions where the material exhibits the strengthened state, it will also exhibit the long-time rupture law and vice versa. In connection with this, upon close examination of an appropriately normalized plot of published creep data (Fig. IV.11) we noticed "plateau" stress levels unique at each creep temperature. At applied stress levels below the plateau, HT9 deforms in accordance with the precipitation-strengthening law. Above this stress, the alloy deforms according to the intrinsic flow law. For our purposes then, we propose that the plateau stress σ_p acts as a temperature-dependent switch between the two states of the material.

We determined the temperature dependence of the plateau stress from an extrapolation of the published creep data (Fig. IV.11) and found it to obey a simple Arrhenius relation.[10] This allows us to easily calculate σ_p at any temperature, and to follow its value through any thermal transient. We made preliminary calculations of σ_p for the conditions of a variety of static and transient high-temperature mechanical tests on HT9. A comparison of this value with the actual flow stress during the test allows us to decide which of the two rupture correlations should be obeyed. We found our selection by this method to

Fig. IV.11. Normalized Flow Stress vs. Normalized Strain Rate in HT9 for Creep and Tensile Data Taken Over a Wide Range of Temperature and Rate. The plateau stress level is shown for creep data taken at 595°, 540°, and 480°C.

agree reasonably well with the choice of correlation observed to fit the rupture time data.[10]

The above agreement illustrates the importance of the plateau stress model and the intrinsic flow model to which it is referred. For this reason, we conducted several high-temperature moderate-to-low rate tests to more precisely determine the parameters of that model. The data obtained from those tests (516°, 621°, 700°C) are shown in Fig. IV.11.

The strengthening that occurs at stresses below σ_p is produced by the appearance during flow at high temperatures of a very fine distribution of carbide precipitates. At these temperatures, however, coarsening of these precipitates will occur, which diminishes the strengthening effect. Inasmuch as plastic flow is produced by the movement of dislocation lines, it is necessary that new fine precipitates form on the dislocations in a dynamic manner in order to maintain the strengthening effect. This must be true in order for our model

to be correct. It also must be true that the precipitate spacing be proportional to the spacing between dislocation intersections. Figure IV.12 is a transmission electron micrograph taken of an HT9 sample being deformed at 600°C in-situ in Argonne's High Voltage Electron Microscope. The figure clearly shows new precipitates forming at the intersections of dislocation lines, while they move under load to produce plastic flow. The precipitates inhibit their movement giving extra strength to the material. This observation provides mechanistic support for our model.

We have made considerable progress this year toward our goal of predicting the in-service rupture behavior of HT9 cladding. More work is needed both in refining our description of the plateau stress region through further mechanical tests, and in analyzing the abundant amount of microstructural data obtained this year.

Fig. IV.12. Transmission Micrograph of an HT9 Sample Deforming at 600°C In-situ in ANL's HVEM. The image shows new precipitates forming on dislocation lines as they move. (ANL Neg. No. 16581, Frame 17)

F. In-vessel Debris Retention

It was stated in the previous Annual Report (ANL-IFR-202, 75ff) that numerous analyses and tests have shown how all classes of slowly initiated transients in metal-fueled, pool-type fast reactors would be self-terminating, without scram and without damage to the core or its environs. Thus, meltdown could occur only if (1) flow was lost abruptly such that feedback could not reduce power sufficiently rapidly, or (2) decay heat removal was seriously degraded. A preliminary review was completed of the likely course of events following each of these extremely unlikely initiating events. The aim of the review was to assess the needs for experiments and analyses to determine the potential for debris retention in the reactor vessel.

It has been shown that, in a sudden loss-of-flow, the total transient energy release would be sufficient to melt only a fraction of the driver fuel with a negligible amount of destructive work. Thus, it is expected that the reactor vessel and internal structures, except for the cladding, would remain intact. This conclusion raises the possibility that the molten fuel debris could be quenched, then cooled by natural circulation of sodium within the reactor vessel. In this scenario the cladding would fail and the fission gas pressure would cause the molten fuel to flow upward and downward through the coolant channels into the above- and below-core structures. For designs in which the pin bundle structures continue below as well as above active core, the penetration into the structures might be very limited. However, the freezing and plugging problem for metal fuel is significantly different from that for oxide fuel. The metal fuel is more conductive, it freezes over a temperature range, and it might interact with the sodium ahead of the flow differently. Thus, freezing and plugging experiments have been recommended.

Fuel melting in the degraded decay heat removal accident would not occur for several tens of hours. At that time, fuel melting and cladding failure would occur incoherently leading to a gradual buildup of melt frozen in above- and below-core structures. Subsequent fuel melting and relocation would occur very slowly because of the low decay heating level.

The fuel frozen in the ex-core structures for both initiating accidents would reheat, melt, and become mobile because of decay heat. The course of remelting is complicated by the existence of eutectics between the U-Pu-Zr fuel and stainless steel. There is a significant heat of alloying between iron and uranium and between iron and zirconium, suggesting that the eutectic melting of steel might not require much heat of fusion. Clearly, the melting temperature and the heat of fusion depend upon the composition. It was concluded that confident prediction of fuel motion subsequent to core failure requires a limited experimental study of eutectic behavior under postaccident conditions.

Fuel-steel melts are expected to fragment on quenching in sodium in relatively open areas. Experimental studies have shown that quenching results in particulate beds having a high void fraction. Large particles are generated unless the melt enters the sodium at high velocity. Fragmentation behavior of metal fuel at low injection velocities is quite different from that of oxide fuel. Analyses indicated that particulate beds of metal fuel, formed slowly, are likely to be coolable by natural convection of sodium through the bed whereas beds of oxide fuel of significant depths are coolable only by boiling of sodium. Although the initial experiments were very encouraging, an uncertainty remains about fragmentation at high velocity or in a limited volume at high ratios of melt to sodium.

The thermal behavior of melt pools supported on steel structures was investigated analytically. The eutectic interaction results in lowered effective melting temperatures for the steel support. This causes significant thinning of thick structures, however, it is likely that residual thicknesses would provide sufficient structural strength. The analyses were based on a uranium melt in contact with iron. Compatibility experiments between more representative melts such as a mixture of uranium, zirconium, and fission product elements with stainless steel were recommended.

A criticality analysis of a melt pool was performed which indicated that a melt pool would be subcritical so long as the melt was able spread to a diameter significantly greater than the original core diameter. Both the criticality and thermal analyses of melts on steel structures require more confident predictions of spreading capability. Experiments may be required.

More analyses are required to provide an initial assessment of the potential for in-vessel retention.

G. Single Failure Analysis of FCF Ventilation System

The Fuel Cycle Facility (FCF) ventilation systems were analyzed to determine whether any single failure of an active component could result in reverse flow which could be harmful to personnel or contaminate equipment. In order to complete this study, a Failure Modes and Effects Analysis (FMEA) for all the major, active components in each of the FCF ventilation systems was performed. The analysis included the building supply system, building exhaust system (BES), air cell exhaust system (ACES), safety exhaust system (SES), stack exhaust system, and the supporting electrical system. The hydraulics of the ventilation system was modeled using the TORAC computer code to determine the effects of component failures at both the system and facility level. The TORAC model was then used when needed as part of the FMEA.

In evaluating the results of the FMEA and the supporting TORAC analyses, flow reversal as addressed in the FSAR was interpreted to include flow from a contaminated area or suspect contaminated area to either a clean area or to the environment except for release to the environment through the designed HEPA filters and stack. Minor flow reversals between areas of equal contamination were considered acceptable. For example, a reverse flow from one clean office area to another or a reverse flow of clean supply air from a clean area to another clean area through the supply ducting would be considered acceptable. Reverse flow of supply air back to the environment was not considered acceptable.

The scope of analysis covered the effects of a single failure of all major active components in the ventilation systems including fans, motors, pressure transmitters and dampers which are actuated as part of any control system. This scope meets the definition of single active failures as defined in the FSAR. Failures of passive components are excluded from this analysis. The items excluded are filters, manual dampers, grills, seal pots, ductwork, and structural supports.

The method of analysis was based on MIL-STD-1629A, Procedure for Performing a Failure Modes and Criticality Analysis and IEEE Std. 353-1975, IEEE Guide for General Principles of Reliability Analysis of Nuclear Power Generating Systems. A bottom-up approach starting at the component level was selected.

A component list was developed for each of the FCF ventilation systems. The support systems such as electric supply and air were considered separately as a lower level of indenture. The failure of a portion of these support systems which affected only a single component of one of the ventilation systems could then be subsumed into the component failure for purposes of the FMEA analysis.

When possible, deductive logic was used to qualitatively propagate the effects of any single failure to the next higher level. Thus a component failure was analyzed to determine if the failure had any adverse effect on its respective system. If there was no such effect, the analysis stopped at this level. If there was an adverse system effect, the resulting failures were propagated to the facility level. Again, if there was no adverse effect on the facility, the analysis was terminated. If the effect of a given failure was not clearly in a fail-safe direction or if the effect could not be determined by a qualitative analysis, a quantitative analysis using the TORAC model of the ventilation system was used to determine both the potential for flow reversal and the magnitude of change of all the affected air flows.

The TORAC cases used in this study are described below and the network schematic for the normal operation case is shown in Fig. IV.13. Cases B-00 and A-00 are double failure cases included for information only since the damper failure in the flowpath of the failed fan would normally close on fan failure and, therefore, constitutes a second failure.

Building Exhaust System (BES)/Analysis Cases

- B-0. One BES fan down/damper closed, building supply fan off. Single fan automatic outlet damper failure.
- B-00. One BES fan down/damper open, building supply fan off. Single fan failure w/all damper(s) 100% open.

Fig. IV.13. TORAC Analysis Model for FCF Ventilation Systems (Version 8/93)

- B-1. BES fans on and inlet vane dampers 10% open; Building supply fan off. BES inlet plenum DP transmitter or controller failure (common to both fans) causing zero output demand.

Air Cell Exhaust System (ACES)/Analysis Cases:

- A-0. One ACES fan down/damper closed; building supply fan off. Single fan failure
- A-00. One ACES fan down/damper 100% open; building supply fan off. Single fan failure and its damper stays open.
- A-1. Both ACES inlet vane dampers closed to 10% open; building supply fan off. DP transmitter failure or controller failure

Stack Exhaust System/Analysis Cases:

- S-0. Standby fan damper(s) all fail at 100% open--causing reverse flow/recirculation through free-spinning fan. Inlet or outlet damper failure
- S-1. Active fan Inlet vane damper closes to 50% (minimum stop). Standby fan remains secured and inlet and outlet dampers closed; Building supply fan off. Failure of Active fan transmitter signal or inlet vane controller, sends minimum demand to Active (and Standby) fan.

Building Supply System Analysis Case:

- BS-1. Building supply fan off--all others systems normal (all dampers remain in nominal fixed positions.) Building supply fan failure.

FCF Electric Power Failure/Analysis Cases (EBR-II power remains):

- E-0. Single bus fault inside FCF Motor Control Center; other busses normal. Loss of power to one ACES fan, one BES fan and supply fan; all other systems normal.

Once the qualitative and quantitative analyses were completed, the results were evaluated against a severity ranking. The facility effect severity rankings were as follows:

Major:	Reversal from a known contaminated zone to a clean zone or from zones of higher contamination to zones of lower contamination.
Minor:	Reversal between zones with the same level of contamination or from "Suspect Contamination" areas to clean areas.

- Operational: Reversal is not expected to occur under normal circumstances; however, if building pressure transients occur following the single failure, contaminated air may be drawn into areas of lesser or no contamination.
- System: Reversal, if it occurs, involves only process air rather than breathing air and impacts solely on system contamination and not worker safety.
- No Concern: Reversal is judged not to be possible based on examination of flow paths and failure modes or reversal involves only clean air zones.

No major concerns were found. Of the remaining minor concerns, only the differential pressure (DP) transmitters have a sufficiently high probability that a DP transmitter failure may be expected within the next five years. There are two potential failures. One of these is a DP failure within the building exhaust system (BES) which could result in a flow reversal through suspect contaminated ductwork to the basement. The other is an air cell exhaust system (ACES) DP transmitter failure which could result in flow reversal from the wash station back to the passageway and therefore to the high bay areas. The annual probability of either occurrence of this type is 2.5% per year. A possible solution is to increase the minimum setting of the fan flow control dampers. This solution will be investigated during system flow balance testing.

The ventilation flow balancing tests and system acceptance tests will involve a total of 28 flow configurations including most of abnormal operations such as the loss of individual fans in the building exhaust or air cell exhaust systems and simulation of controller malfunctions in these two systems and also in the stack exhaust system. The data from these tests are being analyzed currently and once the data are available, all of the TORAC cases will be rerun using the measured boundary conditions. This will provide additional validation of the TORAC model.

Using the design data, the analysis of the FCF ventilation systems showed that the ventilation system will meet the FCF FSAR requirement of no reverse flow of contaminated or potentially contaminated air provided that a sufficient minimum setting of the fan inlet vane control dampers is assured for both the BES and the ACES fans. The conclusions of the initial analysis were validated based

on data from 28 tests conducted in the FCF. These tests examined flow balancing for normal mode operation and building ventilation status during infrequent operations. Nominal data values from these tests were used as input to TORAC to more accurately balance/initialize the FCF model. Eleven of the most severe single failure analysis cases were then analyzed with the updated model. The updated analysis predicted no reverse interior flows and/or pressure drops and no reverse infiltrations. The conclusion of the single failure analysis is that no flows will occur from contaminated to clean areas or from suspect to clean areas and that there will be no reverse infiltrations from inside to atmosphere.

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V. CORE DESIGN DEVELOPMENT

A. Development and Validation of a Physics Database for IFR Fuel Cycle Analysis

Implementation of the Integral Fast Reactor (IFR) Physics Analysis Database (PADB) and the associated neutronics methods is a necessary part of the IFR fuel cycle demonstration in EBR-II and its adjoining Fuel Cycle Facility (FCF). This neutronics package (of methods, models, and database) serves not only the EBR-II operations and physics analysis tasks but also the FCF operations and Mass Tracking System (MTG) material control and accountancy requirements. The former tasks include EBR-II planning, pre- and post-analysis of all reactor subruns, subassembly exchange worths, configuration estimated critical rod position projections, core-follow analysis, run-report generation, Technical Specifications compliance, special nuclear material requirements and EBR-II material balance area reporting. The latter tasks include (data) interfacing EBR-II with the Fuel Manufacturing Facility (FMF) which produces fresh fueled subassemblies for the reactor and the FCF which will process the irradiated fuel and produce "fresh" subassemblies for reinsertion into the reactor.

The IFR Physics Analysis Database contains all the data required to model the portion of the IFR fuel cycle demonstration that is external to FCF. It is essentially all the data required to simulate the flow of material into EBR-II, the transmutation of these materials through the operating cycles of the reactor, and the flow of material from EBR-II to FCF. The simulation of the EBR-II fuel cycle is performed with the codes REBUS-3,[1] RCT[2] and ORIGEN-RA.[3,4] Presented herein are brief overviews of (a) the application of these three depletion codes to model and analyze the EBR-II fuel cycle, (b) the software developed to utilize the PADB to automate the input preparation and execution of each of these three depletion codes, and (c) results of verification and validation tests of the PADB tables and the neutronics methods and models used to determine the physical characteristics of irradiated subassemblies for demonstration of the IFR fuel cycle in EBR-II and FCF.

1. Simulation of the IFR Fuel Cycle

The first step in the depletion analysis is to model the entire reactor with REBUS-3. The neutronics solution uses DIF3D nodal diffusion theory[5] with ENDF/B-V.2 nuclear data. Although the model is full-core, three-dimensional, and includes BOC and EOC control rod and safety rod positions, it is rather coarse in some respects. Specifically, (i) each subassembly in the reactor grid is homogenized within the hexagonal node of the model and (ii) the depletion chains include only the principal actinides between ^{234}U and ^{246}Cm and "lumped" fission products. Therefore, the REBUS-3 model contains no information of individual fuel elements (and thus the necessity for the RCT calculations) and contains no information for the hundreds of individual fission products and activation products which are created in the fuel and structure (and thus the necessity for the ORIGEN-RA calculations). The second step in the depletion analysis is the reconstruction of the spatial flux distribution within the node (i.e., subassembly) using the RCT code. This intranodal flux distribution is then used to "deplete" each fuel element within the subassembly. The above two depletion steps are adequate to satisfy the standard operational requirements for EBR-II. However, requirements for the FCF necessitate the calculation of additional discharge and post-irradiation quantities, such as fission product inventories, material activation, decay heat, and neutron source. At this stage, a third step in the analysis which includes a much more detailed depletion chain is required. This is accomplished with the ORIGEN-RA code, a zero-dimensional depletion code which solves a set of coupled depletion equations using one-group fluxes and cross sections (for nuclides included in ENDF/B-V.2) based on the multigroup REBUS-3/RCT results. The detailed depletion chains are based on an ORIGEN-RA library of materials which includes 253 light activation nuclides, 101 heavy metal nuclides and 821 fission product nuclides. The combined REBUS-3/RCT/ORIGEN-RA analyses provide all the depletion results necessary for not only the EBR-II operations and physics analysis tasks but also the FCF operations and MTG material control and accountancy requirements.

2. Application of the PADB to IFR Fuel Cycle Analysis

Considerable effort has been made to define a set of database tables (and to generate the data for these tables) which would be suitable for

performing the above fuel cycle analyses for EBR-II. The fruit of this effort is the IFR Physics Analysis Database. Realizing that the database could be of practical use only if it was of very high quality, considerable effort has also been made to create software to generate, maintain, verify and edit the database tables and files. To tap the power of the PADB, a set of software has also been written to utilize the database. Specifically, the codes PRPREB, PRPRCT, PRPORG and ORISOZ have been written to automate the preparation of input (i.e., to largely eliminate the necessity of input by the code user in preparing input "decks") for each step of the fuel cycle analysis of EBR-II. The flow of data in the PADB and the steps in the fuel cycle analysis are driven by operations in FMF, EBR-II and FCF. For example, when a subassembly is manufactured in FMF for EBR-II, data for this subassembly are entered into the PADB. When fuel management is performed in EBR-II, the same processes are modeled analytically using REBUS-3/RCT; and when fuel is to be transferred from the tank of EBR-II to HFEF or FCF, the necessary database files which define the physical attributes of the irradiated fuel are generated using ORIGEN-RA in conjunction with PRPORG and ORISOZ.

3. Verification and Validation of the Fuel Cycle Analysis

Verification studies have been made of both the database files and the database software. Verification of the former is achieved primarily through utility software which update and maintain the database files. This software includes extensive checking of the data for consistency and completeness. Verification of the database software is achieved primarily through benchmark testing and independent coding. Both of these verification efforts remain an ongoing activity throughout the period of application of the database and software. The validation studies include tests of the applicability of these methods and models in the prediction of the characteristics of subassemblies irradiated in EBR-II. These tests are achieved by comparing results obtained with the current methods and models with: (i) results obtained using higher-order methods and more rigorous models, (ii) results based on independent methods and models, and (iii) experimentally measured quantities.

The REBUS-3/DIF3D code system has for many years been used routinely at ANL to perform LMR fuel cycle calculations. A concerted effort has recently

been made to qualify this code system, in conjunction with appropriate cross section data and reactor modeling procedures, specifically for analysis of EBR-II. Conclusions drawn from the qualification tests include:

1. Neutron fluxes and reaction rates in the core are predicted to within ~3% of reference results computed with continuous-energy Monte Carlo.
2. Depletion-dependent fluxes, reaction rates, and nuclide inventories are in acceptably good agreement with the results of procedures used historically to analyze EBR-II.
3. As further described below, local predictions of burnup and fuel isotopics based on the use of REBUS-3/DIF3D (in conjunction with RCT) agree with measured values for small segments of irradiated fuel pins to within the measurement uncertainties.
4. Critical control rod positions for a number of recent EBR-II runs are accurately predicted using eigenvalue differences determined with REBUS-3/DIF3D.

Several tests of the RCT reconstruction method were reported in Ref. 2, where reconstructed power densities for a heterogeneous model of EBR-II were shown to differ by less than 3% from reference results (computed with a very fine mesh) for all axial positions of all core subassemblies, with the largest errors observed near the core axial boundaries. A more comprehensive assessment[6] of the RCT and REBUS-3/DIF3D capabilities involved the analysis of an extensive sequence of EBR-II runs (runs 130A through 154F) and the comparison of calculated depletion-dependent characteristics against experimentally measured local values of burnup and U and Pu isotopic ratios. Discrepancies between computed and measured results were found to be smaller than the combined uncertainties inherent in the comparison, e.g., uncertainties in measurements of irradiated sample characteristics, in the EBR-II power level, and in the preirradiation compositions. These uncertainties, which are unaffected by potential refinements in the physics analyses, were estimated to be ~6% (1 sigma) for sample burnup, ~0.2% for U-235, U-238 and Pu-239 mass fractions, and ~1.0% for U-234, U-236, and Pu-240 mass fractions.

Studies aimed at validation of the ORIGEN-RA code, data libraries, and models for EBR-II/FCF physics analyses have so far focused on predictions of those discharge and postirradiation characteristics which are of central importance to fissile material accountability, criticality safety, heat loads in process equipment and containers, and intensity of penetrating radiation emissions. These characteristics include the principal fissile and fertile nuclide densities and reaction rates (validated by demonstrating consistency with REBUS-3/RCT results), densities of fission products by element and chemical group (validated against independent calculations, and by demonstrating insensitivity to major calculational approximations and uncertainties, e.g., neutron capture data for fission products), and decay heat (validated against published experimental and computational results, and against the U.S. decay heat standard ANSI/ANS-5.1-1979). Errors in the ORIGEN-RA predictions of these different parameters were demonstrated to be acceptably small for EBR-II/FCF applications.

The above verification and validation studies indicate the IFR Physics Analysis Database and the software and procedures used to analyze EBR-II core physics are sufficiently accurate to satisfy operational and material accountability requirements in FCF. In particular, masses of the primary fissile and fertile nuclides in irradiated fuel elements can be computed with an accuracy of ~2% or better, based on assessments of the dominant calculational uncertainties and on comparisons of predictions obtained with the REBUS-3/RCT codes against burnup measurements,[6] EBR-II operational data,[7] and more rigorous calculational models.[8] Relative errors in the predicted amounts of trace heavy isotopes and individual fission/activation product nuclides are potentially much larger. Nevertheless, integral characteristics of the fission product inventories of relevance to FCF operations were found to be rather insensitive to the approximations and uncertainties in the ORIGEN-RA analyses used to compute detailed inventories and postirradiation characteristics. Moreover, some of these integral characteristics have been successfully benchmarked against measurements and independent calculations, as well as the 1979 U. S. Standard for fission product decay heat. Based on these comparisons, and on the uncertainties in EBR-II fission rates, it appears that the uncertainties in the activity and decay heat of discharged subassemblies are each on the order of 10% for the postirradiation times of interest. The uncertainty in the gamma component of the decay heat is larger (~25%).

It has thus been demonstrated that the IFR Physics Analysis Database and associated procedures are adequate to predict physical quantities required for initial phases of FCF operations. Additional validation tests will likely be needed to qualify computed characteristics of previously recycled fuels, whose initial makeup will include carryover constituents from recycling operations. Operating experience in FCF will be instrumental in identifying additional validation needs and in providing the measurements against which the relevant physics predictions can be tested.

B. Parallel Distributed Computing

Large-scale scientific and engineering calculations can now be carried out by three different general approaches: traditional computing, where all number-crunching is performed on powerful centralized computers or supercomputers; MP computing, where the number-crunching is performed on a massively parallel system with hundreds or thousands of tightly coupled identical processors; and distributed computing, where a geographically dispersed, possibly heterogeneous collection of separate computers is used. Distributed computing is relatively new, but has perhaps the greatest potential for radically improving the state of scientific computing. In recent years there has been an explosive growth in the power and availability of desktop, Unix-based, engineering workstations, providing a "virtual supercomputer" to anyone having the proper software to coordinate and control a distributed parallel calculation. Practically speaking, such a system must include support for traditional system services, such as a queuing system for batch work, reliable file system backups, and parallel processing capabilities for large jobs. Large-scale scientific and engineering calculations for IFR analysis are now being carried out effectively in RA Division's distributed computing environment at costs far lower than for traditional mainframes. All ANL computer codes for IFR and other reactor analysis have been adapted successfully to a distributed system based on 14 Sun and two IBM workstations and an X-terminal for each analyst. Distributed parallel processing has been demonstrated to be effective for long-running Monte Carlo calculations.

For over 30 years, Argonne National Laboratory has developed and applied large-scale software systems for the engineering analysis of nuclear reactor

plants. In the past two years, all of the reactor analysis codes applied to the IFR (e.g., VIM, DIF3D, REBUS3, MCC-2, SASSYS, COMMIX, etc.) and supporting software have been adapted to a distributed computing environment. The large capacity of this system now enables more detailed calculations than ever before. For example, EBR-II neutron transport calculations using hundreds of neutron energy groups, IFR thermal-hydraulic calculations of decay heat removal with tens of thousands of mesh cells, and EBR-II Monte Carlo neutronics calculations where spatial reaction rates are required represent an order-of-magnitude expansion of capability over the previous mainframe system. In addition, the Distributed Queueing System[9] (DQS) has been installed to manage the scheduling and load balancing for batch jobs, and the P4[10] parallel processing software is available to support parallel applications. All batch computing work is submitted to DQS, which locates the proper machine resources, schedules the work, and initiates execution of batch jobs. DQS provides load-balancing in a distributed system via a "group" mechanism: Batch queues reside on individual workstations and handle requests on a FIFO basis. Several batch queues may be grouped together, and jobs submitted to that group can be scheduled on the next available, least-loaded machine in the group by DQS. This arrangement has functioned very well, handling about 3000 batch jobs per month with little or no impact on the interactive response time.

To support parallel distributed processing, the P4 software package provides libraries of Fortran- or C-callable subroutines which handle the startup of processes and communication between processes on (possibly) different machines. P4 is used to implement a "message-passing" approach to loosely-coupled, parallel processing on a workstation network. In addition, codes developed in such a manner are very portable, and can generally be run with few or no changes on other machines supported by P4. For parallel processing to be effective on a distributed system, there must be close cooperation between the message-passing system (P4) and the job scheduler (DQS). DQS provides direct support for parallel jobs, in that its scheduling and resource allocation schemes specifically allow multiple workstations to be assigned to a specific job. Other batch work is subsequently scheduled on different machines, to avoid interfering with the parallel job.

1. Parallel Distributed Computing with VIM

Monte Carlo simulations provide a highly accurate, but computationally intensive, means of analyzing the physics behavior of nuclear reactors. The VIM[11] Monte Carlo code from Argonne is a well-known, mature, and proven code, with no essential approximations to the geometric modelling or collision physics. VIM can be used to provide "truth calculations," or computational benchmarks, for calibration and fine-tuning of various faster-running, but less accurate, computational methods. The code is used for either neutron or gamma-ray transport, and can handle both multigroup and continuous-energy cross-section datasets. VIM has been used extensively for reactor core analysis (especially for the ZPR and ZPPR fast critical assemblies) and reactor shielding analysis (fixed-source problems), and will handle fully-detailed 3-D general geometry and lattice geometries. It has been a benchmark for transport methods development which were found important in the analysis of EBR-II reflector and blanket neutronics.[12]

VIM was developed at, and has been supported by, Argonne since the late 1960s. It consists of about 30,000 lines of machine-portable Fortran-77. VIM has been used on a very large number of different computers, including SUN-SPARC2, IBM-RS6000, CRAY, IBM-3084, VAX, CDC, etc., VIM Monte Carlo is very computationally intensive, with a single VIM calculation running continuously for days or weeks on a single workstation. The DQS batch system handles these jobs effectively, in addition to thousands of shorter jobs submitted each month. Current VIM development is focused on parallel processing in a distributed computing environment, using the P4 parallel processing software package also developed at Argonne. The concept of a "virtual supercomputer" is supported by DQS, so that several machines may be allocated to a single VIM job.

Monte Carlo particle transport methods are ideally suited to parallel computers due to the inherent parallelism in the fundamental algorithm. A distributed computing system provides a form of parallel computer, with slow, Ethernet-based communications between nodes. Latencies for sending messages between machines are large, in the 1-10 ms range, and data streaming rates are somewhat less than 16MB/s. Message-passing for distributed parallel processing is thus roughly similar to disk I/O, and is many orders of magnitude slower than

(local) memory access. While the relatively slow message-passing available on a distributed workstation network will inhibit many types of parallel calculation, parallel Monte Carlo can still be very effective since interprocessor communication is minimal, involving primarily the combining of results at the completion of all histories. If sufficient memory is available to each processor, the Monte Carlo code and data can be replicated, and each processor can independently follow a portion of the particles. Existing Monte Carlo codes require changes in only a few high-level locations, permitting the reuse of nearly all coding without change. The key requirement for widespread applicability of particle transport Monte Carlo on distributed computers is memory size. At least 16 MB per processor is required to hold a complete copy of the Monte Carlo code and data for most applications, and many large problems (for which parallelism is crucial) require 32 MB or more per processor.

Parallel VIM calculations are intended to be scalable to any number and combination of SUN and IBM workstations in the network. A "mother/daughter" approach is used, with particle histories (in the current generation) partitioned among the daughter processes. Each daughter tracks its share of the neutrons independently of the other daughters. Synchronization is only necessary between neutron generations or when reaction rate tallies need to be passed to the master process for combination with the tallies from the other daughters. Special consideration is given to the manner in which work is partitioned among processors: Since VIM is run primarily on a workstation network, there is a large potential for wide disparities in the processing speed of each node. First, the individual workstations used in a parallel calculation may have different processor types and speeds. Second, the interactive and system loads on each machine will vary in an unpredictable manner, even during a calculation. Third, it is even possible for a processor to get so busy that it becomes effectively unavailable. In effect, the "virtual supercomputer" configuration and performance could change continuously during the course of a single calculation. To handle these difficulties, the particle histories within a given batch (generation) are not statically partitioned among processors. The histories are processed in "chunks" of a few hundred or thousand histories at a time, sent only to those daughter processes which ask for work. Whenever a daughter process is ready to accept new work, it signals the mother. The mother process then sends a "chunk" of work to the ready daughter. In theory, one machine could wind up handling all

of the histories. In practice, however, the chunks of work for long calculations have been found to be distributed among processors in rough proportion to their relative CPU speeds.

To insure reproducibility, special techniques must be used for both the random walk and the source sampling used in eigenvalue calculations[13]. For each neutron in a generation an initial random number seed is generated based on a specified "stride" through the random sequence. During the random walk, the random number generator is used to sequentially compute random numbers for a neutron based on its own initial seed. Each neutron is thus independent of all others, and hence the scores due to a neutron are independent of the order in which the neutron histories are analyzed. This is sufficient to insure reproducibility within a generation. Complete reproducibility requires a repeatable initial ordering of neutrons for each generation. During a generation of neutron histories, induced fission sites are stored in a "fission bank" for later use in sampling the source for the next generation. Each slave process maintains its own fission bank, and at the conclusion of a generation the individual banks are concatenated into a master bank on the master process. A unique reordering of the progeny in the fission bank, according to their parent neutron number, is accomplished using the algorithm of Ref. 13.

Parallel VIM calculations have achieved reductions in computing time in direct proportion to the number of distributed workstations utilized. While parallel speedups vary somewhat depending on problem type, typical performance results using multiple SUN workstations on the RA Network are shown in Fig. V.1 for a VIM analysis of the TREAT reactor. Even on a moderately busy network, speedups have been nearly linear with the number of workstations, as long as not all of the available machines were used. That is, during the course of a typical calculation, there are always several background network activities (e.g., file backup, mail processing, NFS file transfers, etc.) which may execute at high priority and monopolize a particular machine's CPU cycles. As long as DQS is asked to allocate some, but not all, of the machines to a calculation, it will not allocate the machines which are heavily loaded with system tasks to the VIM calculation. Using the "virtual supercomputer" provided by a workstation network, a typical Monte Carlo calculation which previously required several days or weeks of computing time on a single workstation can now be completed overnight using 4-8 machines.

Fig. V.1. Typical Speedup as a Function of a Number of Workstations

2. Conclusions and Future Needs

Distributed computing provides capabilities for performing more IFR calculations, at much lower cost, than ever before. Our experience in applying a distributed computing approach to IFR analysis has made us aware of a number of challenges and opportunities for continued development. To be effective on a variety of parallel and distributed computer systems, future large-scale scientific software must be able to adapt itself to changes in the computing environment. Key features are fault tolerance, dynamic load balancing, and multiple levels of parallelism in the coding and algorithms. Monte Carlo codes are generally early adopters of advanced features, since the codes are adaptable to any known computer architecture. It is reasonable to expect that speeds of 100-1000 times that of current computers will be possible in the near future, allowing for unprecedented applications for Monte Carlo simulation. Success in adapting Monte Carlo methods will provide experience and expertise in robust, fault-tolerant scientific computation, paving the way for applications in other areas. It is our plan to investigate these areas first for Monte Carlo, and then to extend the lessons learned to other large codes.

C. Sensitivity of Waste Toxicity to Uncertainty in Basic Nuclear Data

Techniques were developed to analyze uncertainties in the radiotoxicity of nuclear waste; these methods also evaluate the sensitivity of the waste radiotoxicity to changes in the basic nuclear data (e.g., nuclear interaction cross sections and nuclear decay constants). The sensitivity analysis techniques have been applied to the IFR fuel cycle to evaluate the uncertainty in the waste radiotoxicity, and to identify key nuclear data impacts.

1. Radiotoxicity Sensitivity Evaluation

Long-term radiotoxicity reduction appears to be one of the most important issues related to radioactive waste transmutation.

Because of many studies related to radioactive waste transmutation, the problem of getting better data for minor actinides and fission products has been the focus of renewed interest.

Uncertainty impact evaluation can be used to find the most urgent data improvements to be fulfilled. Time-dependent sensitivity techniques, which have been used in the past for standard reactor application, have been adapted here to solve this problem. The radiotoxicity R is expressed by mean of radiotoxicity factor f_i for each nuclide i

$$R(t_f) = \sum_i n_i(t_f) f_i$$

where $n_i(t_f)$ is the nuclide i density at the final time t_f . The time interval to be considered starts from t_0 , the initial time of irradiation in a reactor. In the time interval (t_0, t_f) one has to consider the irradiation time steps, the cooling times before possible reprocessing, the fuel cycle processing time and the decay period in long-term storage.

Sensitivity coefficients, which express the value change of an integral parameter (in our case the global radiotoxicity) related to any variation of a specific basic data (e.g., cross sections or decay constants of

a nuclide) can be calculated by solving the adjoint form of the standard nuclide transmutation equation with special final time boundary conditions.

Once the sensitivity coefficients S_x are available, given a covariance matrix D_{xy} on the basic data x , the uncertainty I_R on the global radiotoxicity is easily calculated by

$$I_R = \left(\sum_{xy} S_x D_{xy} S_y \right)^{1/2}$$

2. Applications

The main application we have considered are typical EFR (European Fast Reactor) and IFR (Integral Fast Reactor) reactor systems and associated fuel cycles.[14]

The decay chain used in solving the transmutation equation is quite complex and goes from Pb-210 to Cm-248.

The radiotoxicity is expressed in terms of cancer doses. The related radiotoxicity factors assess the hazard (consequences of exposure) for the actinide elements; hazard analyses assume complete exposure to the radioactive inventory and do not evaluate mitigating effects (e.g., shielding, barriers to release, etc.,). In this section, the fatal cancer dose measure developed by Cohen[15] is utilized to quantify the hazard; this measure is based on dose exposure data from the ICRP publications and cancer risk data from the BEIR reports.

To put the cancer hazard in perspective, the hazard of the spent fuel is normalized to the cancer hazard of the uranium ore required to produce the fuel. The typical fast reactor assemblies investigated here contain roughly 20 kg of transuranics. To obtain 20 kg of transuranics, 2 MT of LWR spent fuel would need to be processed; and this corresponds to roughly 10 MT of uranium ore.

The uncertainty analysis has been performed using typical basic data uncertainties on cross sections and decay constants. Uncertainties on radiotoxicity factors have not been included.

In the case of EFR, one cycle consists of 1500 days of full power irradiation with three different values of constant flux for each 500 day depletion time step.

Three different radioactive waste management strategies have been considered:

<u>Strategy</u>	<u>Disposed to Repository</u>
A	100% MA, 0.3% Pu
B	100% MA, 100% Pu
C	1% Ma, 0.3% Pu

Strategy A assumes all the Minor Actinides (MA) are lost in reprocessing and disposed in the waste; a 0.3% Pu loss in reprocessing is also assumed. Strategy B utilizes a once-through cycle, where all transuranics (TRU) are lost to the waste. Strategy C considers only fractional TRU loss in reprocessing.

The sensitivity analysis performed for different storing times (10^3 , 10^5 and 10^7 years) in the repository has shown values that appear to be quite low. Exceptions were large sensitivities to Pu-242 and Am-247 capture cross sections for strategy A, and significant values for Pu-239 and Np-237 decay constants for large storing times (10^5 , 10^7 years).

In Table V.1 values of the global radiotoxicity are shown along with related uncertainties.

Because all the transuranics are included in the waste stream for strategy B, large radiotoxicities are found. Uncertainties are largest in the case of strategy A because of the importance of transplutonium data which carry larger uncertainties. In any case, the uncertainties appear to be quite acceptable with the present status of knowledge on basic data.

TABLE V.1. EFR One Standard Subassembly Irradiated During One Cycle
[Global radiotoxicity (expressed in relative cancer hazard)
of radioactive waste disposed in repository.]

Strategy	Radiotoxicity	Storing Time in Repository		
		10 ³ Years	10 ⁵ Years	10 ⁷ Years
A	Global	$3.25 \times 10^1 \pm 29\%$	$2.50 \times 10^{-1} \pm 32\%$	$1.77 \times 10^{-3} \pm 39\%$
B	Global	$3.18 \times 10^2 \pm 8\%$	$1.92 \times 10^0 \pm 10\%$	$2.07 \times 10^{-2} \pm 9\%$
C	Global	$1.17 \times 10^0 \pm 10\%$	$1.63 \times 10^{-2} \pm 10\%$	$7.40 \times 10^{-5} \pm 11\%$

In the typical IFR fuel cycle model, a fuel driver subassembly resides in the core for four one-year cycles at 80% capacity factor corresponding to a 292 full power days cycle. At the end of the irradiation 0.1% of the TRU isotopes are lost in the pyroprocessing and disposed to a repository. Isotopic depletion was computed using a 900 Mwt core and an equilibrium recycle model with 5% of the rare-earths (excluding Y, Eu and Sm) carried in the TRU stream.

In Table V.2, values of global radiotoxicities are shown along with their related uncertainties. As expected, radiotoxicity is well below corresponding values of EFR because only 0.1% of TRU go to waste during the reprocessing. Uncertainties are also lower because of less sensitivity for IFR to higher actinide basic data (e.g., Pu-242 and Am-241). These lower sensitivities are attributed to two effects.

1. Both minor actinides and plutonium have a 0.1% processing removal; thus, ratio of MA/Pu is much smaller than strategies A and C described for EFR.
2. The hard neutron energy spectrum associated with the metal IFR fuel leads to less production of higher actinides than is the case for EFR.

TABLE V.2. IFR One Subassembly Irradiated During Four Cycles
[Global radiotoxicity (expressed in relative cancer hazard)
of radioactive waste disposed in repository.]

Radiotoxicity	Storing Time in Repository		
	10 ³ Years	10 ⁵ Years	10 ⁷ Years
Global	$1.82 \times 10^{-1} \pm 4\%$	$3.68 \times 10^{-3} \pm 10\%$	$1.52 \times 10^{-5} \pm 6\%$

3. Conclusions

A sensitivity and uncertainty analysis on radiotoxicity evaluation has been conducted for such applications as EFR and IFR reactor systems.[14] The results indicate that the total hazard due to TRU vis-a-vis that of the original ore are low. In addition sensitivities to basic data, except for the Pu-242 and Am-241 capture cross section and some selected half-life constants, are also quite low.

The present knowledge of basic data lead to quite acceptable (of the order of 30% in the worst case) values for uncertainties related to global radiotoxicity at different storing times in a repository. The methodology employed can be useful to establish optimum strategies for radiotoxicity reduction. Finally, a similar analysis can be done taking into account the long-lived fission products, but for the scope of this work we have limited our application to actinides.

D. MTG Status Update

The Mass Tracking (MTG) System collects, stores, retrieves and processes data during operations in FCF for a number of purposes: support of operations, compliance with operating limits, process modeling and material control and accountancy. The MTG System consists of a number of programs and a variety of data files. Communications between operators and the MTG System is supported through Operator Control Stations (OCSs) and through Terminal Session accessible over the Laboratory network.

Most operational data are collected at OCSs and are stored in the MTG System in one of three forms:

- (1) Oracle relational database tables containing weight measurements, records of accountable items, records of transactions involving those items, and other miscellaneous data. Process material, containers and even criticality zones are treated as accountable items. The design of the relational tables is such that the database represents a complete history (in time and location) of the prior flow of process material through the facility.
- (2) Binary files defining the composition of all process material and holdup.

- (3) A variety of ASCII files containing such information as: parameters used in applying operating-limit rules, modeling parameters, history files for the major process-equipment batches, and other data that do not logically fit in either of the other two forms of data storage.

The time frame for logging data depends on the nature of the particular data. For example, weight measurements are recorded automatically from an OCS when the operator touches the "weigh and record" button. On the other hand, data defining generic, container-to-container or zone-to-zone transfer are recorded before or after at specific steps of those processes.

Data are also collected and stored during other, process-specific operations. Currently software which supports casting operations is coded and in production use. Software supporting pin processing, welding and settling is being tested. In all of these cases the MTG System also computes and stores composition changes when they occur, and prints reports which summarize the operations.

The MTG System supports the enforcement of facility operating limits in two areas: criticality safety and the technical specifications regarding the kinds and amounts of materials which may be exposed to the argon-cell atmosphere (sometimes called the "material-at-risk" rule). Before any kind of transfer, MTG System software is invoked which checks for compliance with many (but not all) of the rules specified in the Criticality Hazards Control Statement. These include rules governing the kinds of containers and materials which are permitted in specific zones, rules governing masses of material permitted, and rules governing the numbers of containers permitted. Whenever container (and certain equipment) are opened or closed, other software checks for compliance with the "material-at-risk" rules.

Samples of material are taken at several places in the overall process for analysis by the analytical chemistry laboratory. These samples themselves, as well as the analysis results, are logged into the MTG System. Additional software is in place to revise the book compositions of the particular accountable items which the samples represent, and then to propagate the revised compositions down stream to any other, affected items. Although most of the

interactions between the MTG System and facility personnel are through the OCSs, there are a number of types of transactions which are done through ASCII terminal sessions. These include starting and stopping the MTG Servers, the initial data entry for items (e.g., containers and feed material) which are to be introduced into the facility, and transactions which are a part of the procedure for revising compositions of process material when analytical-chemistry results become available.

The Material Control and Accounting (MC&A) System forms an integral part of the MTG system. It consists of software for estimating measurement errors, controlling the measurement errors, calculating material balances and their associated uncertainties. In addition, central to the MC&A system of the fuel cycle facility (FCF) are the balances for mass measurements. The remote operation of the in-cell equipment requires remote in-cell calibration and control of the balances and limits their direct adjustment.

The estimation of errors, both random and systematic, is performed at calibration, which consists of repeated measurements of standard masses. Five balances for mass measurement have been installed in the FCF for startup operation. Three of these are for masses up to 32 kg and two for masses up to 120 kg. In preparation for pin casting operation with depleted uranium, these instruments have been remotely calibrated in-cell, taking into account as much as possible the environment characteristic of routine cell operation. Based on these data a SAS® regression procedure is used to determine the absolute and relative bias together with the random and systematic errors, and the lack of linear fit for a particular instrument. These data form the basis for the corrections to the instrument reading and the uncertainty to be associated with measurements performed with the instrument.

Measurement control is maintained on two levels. At least twice a day, prior to the use of the instrument, a measurement of a standard mass, randomly selected from a set, is made, and the measured value compared to the predictive interval. The randomization of the selection and the use of the predictive interval minimizes the number of measurements at the first level of control. These data are maintained in the MTG and decision rules are in place which will, when necessary, initiate more detailed statistical tests to assure the instrument

is in control or requires recalibration. This second level of control consists of a sequence of SAS® procedures which test normality and the lack of autocorrelations, that is the assumption underlying the control chart methodology. Control charts for mean and variance are generated for individual measurements and weekly averages. For timely detection, cumulative sum charts are generated with appropriate decision limits.

To assure that all material is accounted for, material balances are performed over unit process areas and the facility as a whole. These balances consist of inventory differences, between two time periods, together with an associated uncertainty. As a part of startup checkout of the FCF, material balances were calculated for the initial pin casting with depleted uranium. This tested the data handling between instruments and MTG, and the interface with the MC&A software. For the casting operation, taking into account not only mass measurements but also results of chemical analysis, the total uranium inventory difference was 6.5 g with a standard deviation of 59 g. The total throughput during this operation was 20 kg.

At the front and end of the FCF operations, data files describing input and output fuel elements and subassemblies are passed between the MTG System and reactor operations. In an effort which is separate from the MTG System, an extensive database (the Physics Analysis Database) has been developed which describes the compositions and structures of fuel elements which have been irradiated in EBR-II and which are scheduled to be processed in FCF.

All of the software mentioned above are either in production use or are in test awaiting the installation of process equipment. Qualification testing has been completed on software required for casting operations (basic transfers and casting-specific transactions), compliance with operating limits, sample-data entry and processing, and material control and accountability.

To date, more than 130,000 lines of code have been written. Additional software will be developed to support the operations of the electrolyzer, the cathode processor, and the fuel-element chopper.

E. Criticality Safety in FCF Design and Operations

The Fuel Cycle Facility (FCF) is designed to demonstrate the feasibility of commercial-scale remote pyrometallurgical process equipment for metallic fuels from liquid metal-cooled reactors and to show closure of the Integral Fast Reactor (IFR) fuel cycle. Requirements for nuclear criticality safety impose the most restrictive of the various constraints on the operation of FCF. The upper limits on batch sizes and other important process parameters are determined principally by criticality safety considerations. To maintain an efficient operation within appropriate safety limits, it is necessary to formulate a nuclear criticality safety program that integrates equipment design, process development, process modeling, conduct of operations, a measurement program, adequate material control procedures, and nuclear criticality analysis. The nuclear criticality safety program for FCF reflects this integration, helping to ensure that the facility can be operated efficiently without compromising safety. The experience gained from the conduct of this program in FCF will be used to design and safely operate IFR facilities on a commercial scale.

Nuclear criticality safety at FCF is achieved by carefully controlling the amounts, forms, locations, and configurations of fissionable, moderator, and other process materials within the facility. This is done by accounting for all controlled materials within the facility and by dividing the facility into criticality hazards control zones. Each zone has a specific set of operations that will be conducted within the zone, and each operation within the zone has a specific set of rules and limits that govern all aspects of the operation. The specific rules and limits detail the types of containers, the forms and types of controlled materials, and the masses of controlled materials allowed within the zone during the given operation. Implementation of the nuclear criticality safety program is achieved through integration of physical criticality controls, administrative criticality controls, the Mass Tracking System, process models, and nuclear criticality analysis.

1. Physical Criticality Controls

A physical criticality control is any design or modification that physically limits or controls the amount and/or configuration of process

materials such as fissile material, moderators, or chemicals. FCF and the process equipment within FCF have been designed to maximize the number of physical controls inherent in the system. Physical controls include container designs, storage rack designs, equipment designs, exclusion of a water-based fire suppression system in the hot cell areas, and division of the facility into specific zones referred to as criticality hazards control (CHC) zones.

Container design is a major aspect of criticality safety. Containers which house large quantities of fissile material or which house the more reactive forms of fissile material are designed to satisfy specific criticality safety criteria. These criteria include limiting container capacity, limiting the type of material that can be placed in a container, limiting the geometric configuration of a container, and limiting the interaction between adjacent containers. Nuclear criticality analysis was used to guide container design and to verify that final designs provide adequate criticality safety controls.

Each storage rack is designed to limit the number and types of containers that can be stored in that rack and to rigidly control container configurations in the rack. Process equipment is designed to limit the number and types of containers that can be used in the process.

Process equipment design was driven largely by process requirements, e.g., operating temperature requirements, but nuclear criticality analysis was used to verify that the final designs of process equipment and storage racks did not pose any criticality hazards.

The FCF was divided into a number of criticality hazards control (CHC) zones, setdown areas, and transfer corridors. The CHC zones separate areas where different operations are performed and neutronically isolate the zones (and process equipment items) from one another. The setdown areas and transfer corridors provide areas where containers can be transferred, weighed, or examined. Setdown areas and transfer corridors are isolated from operations within CHC zones, so the interaction between a container in a setdown area or transfer corridor and the various pieces of process equipment is minimal. Interaction among zones was still considered for credible scenarios such as dropping a container in transit.

Operations in the FCF hot cells are based on minimizing the amount of moderator present in any CHC zone in these cells. The fire suppression system is specifically designed to prevent the introduction of a large amount of water into the hot cells. The practical value of limiting the amounts of water and other moderators in the hot cells is that process batch sizes can be increased due to the larger critical masses in the absence of significant moderation.

2. Administrative Criticality Controls

Administrative controls are any rules, limits, or restrictions imposed which reduce the probability or consequences of an accident due to operator error or procedural violations. While physical controls are based on actual physical constraints; administrative controls are restrictions on the conduct of operations based on safety analysis and operational requirements. Physical controls have been relied upon whenever possible in FCF. However, administrative controls are required for many of the operations in FCF, and adherence to these administrative controls is an important component of criticality safety in FCF.

Administrative controls require compliance of operations personnel with a strict set of rules, limits, and procedures. A system of checks and balances at all levels of facility operations is used to minimize the probability of noncompliance. Because a violation of administrative controls is still possible, multiple administrative controls are imposed on each operation in FCF. The criticality analysis included consideration of events such as double batching that could result from violation of administrative controls. The criticality analysis ensures that occurrence of a potentially unsafe condition requires multiple violations of the administrative controls.

The criticality safety analysis and the formulation of the specific administrative controls were closely linked. The administrative controls formed part of the basis (along with equipment design, container design, and process development) for the criticality analysis. The results of the criticality analysis were then used to refine the administrative controls. This cycle was continued until an acceptable regime of safety and control was achieved. The final result is a set of criticality hazards control rules and a set of facility

operating procedures which govern and limit all operations within FCF. These rules and procedures minimize the potential for an unsafe condition to develop in FCF.

A key aspect of the administrative controls is the limits imposed on each operation in each CHC zone. For each operation in each zone, a list exists which specifies the number and types of containers permitted in the zone. This list also specifies the forms of material permitted in each container type as well as the mass limit for each allowed material form/container combination for each operation. In addition, the list includes mass limits for all controlled materials in a zone during a given operation. The administrative controls include supplemental rules to address any unique aspects of an operation that cannot be adequately covered by the limits on containers, material forms, and masses.

3. Mass Tracking System

The Mass Tracking System (MTG) tracks the location and movement of containers and items of process material in FCF and automatically updates a database with current locations, amounts, and compositions of tracked items. This system records the amount of fissile material assigned to each container and equipment item as holdup as well as the amount of fissile material assigned to each zone as zone difference (a quantity which is functionally equivalent to holdup but which applies to the CHC zone as a whole). In addition, the MTG System generates mass balances for zones over arbitrarily specifiable time periods and carries estimated errors for subsequent statistical analysis. In addition to meeting the material control and accountability requirements for FCF, these MTG System functions are important to the criticality safety program. The material control features of the MTG System reduce the probability of violating criticality safety limits during operations.

The MTG System has several additional features which relate specifically to criticality safety. The MTG System maintains a database of criticality operating limits against which proposed activities involving controlled materials are compared prior to actual movement of a controlled material. For each operation in each zone, this database contains the

criticality safety mass limits for controlled materials permitted in the zone as well as lists of containers and material forms that are permitted within each zone during each operation. The list extends one level further to specify the types of materials permitted in each container during each operation as well as the criticality safety mass limits for each container during the operation. In addition, the MTG software includes coding to apply special criticality safety rules that are only relevant to specific operations, e.g., casting or pin-processing.

The MTG System is interrogated for verification that operating limits will not be exceeded by a proposed transfer of controlled material prior to the transfer. The MTG System is also interrogated before starting a process to verify that criticality safety requirements will be met.

4. Process Model

A process model is a model or simulation of various of the chemical and physical processes that can occur during the operation of a particular piece of equipment. The process model for a particular operation is based on the following factors: a) chemical and physical laws which govern the process, b) models and relationships developed during earlier phases of FCF operations, c) information about previous operating history of and present physical conditions in the equipment at the start of the operation, d) information about the locations and quantities of process materials, e) analysis of previous states of process materials, and f) prior experience with similar operations. A process model could, in principle, be constructed for each operation in FCF. In practice, however, it is not necessary to construct a formal model for the more routine operations such as subassembly fabrication.

A process model serves several purposes that relate to criticality safety in FCF. First, the process model can be used to detect irregularities during normal operations. Persistent or significant deviations of measured process parameters from model predictions indicate a potential abnormal situation that needs to be investigated. Second, the process model can be used to run simulations of proposed changes in operations to identify promising proposals. Third, the process model is used to specify process parameters and parameter

limits for some operations. Finally, because of the nature of the electrorefining process, it is not possible to measure precisely the mass or composition of the actinides removed from the electrorefiner in an inhomogeneous cathode product until several steps later (cathode processing) in the overall reprocessing procedure. The initial mass and composition estimates required for criticality control and to meet limits for exposed pyrophoric materials in FCF must be based on the process model.

An accurate and conservative process model for each major piece of process equipment in FCF is an important aspect of the criticality safety program. These process models are used with the nuclear criticality analysis methodology to establish the criticality limits for each operation, and these limits establish one set of constraints on facility operation (batch sizes, compositions, etc.). Process models are used as predictive tools to show that current operations are within facility operational and safety limits and that proposed operational changes will remain within the safety envelope. Finally, process models are used to specify operational parameters for the Process Work Sheets that actually govern a specific operation.

5. Nuclear Criticality Analysis

The basic approach taken in the FCF criticality analysis is mechanistic. Five broad categories of abnormal events were defined. These categories were mass events (generally overloading or excess cathode deposition), material events (material substitutions or production of abnormal compositions), density events (abnormally high densities or packing fractions), reflection events (changes in reflection around equipment or containers), and geometry events (abnormal geometric configurations of process material). Accident configurations which did not fit into these broad categories such as certain consequences of the design basis earthquake, were treated as special cases.

Each operation in FCF was analyzed to determine the credible events in each of these categories. Four criteria were used to determine and categorize credible abnormal events. First, the chemistry and physics of the process were considered. Second, equipment and container designs were analyzed to determine any physical constraints on volume, configuration, etc. Third, the nature of the

process and the materials used in the process were examined. Fourth, the preparations for and conduct of the operation were examined to determine the likelihood of certain events. Events were defined with sufficient generality and conservatism to adequately cover process uncertainties and the novel aspects of the processes. Ranges of very similar events or event initiators were combined into a single, conservatively defined event to reduce the volume of required calculations. Arbitrary or unnecessarily conservative assumptions were avoided in modeling processes and in event definitions. The events and modeling were based on a conservative but realistic analyses of FCF operations and on a conservative assessment of process uncertainties.

Each credible abnormal event in each category was classed as unlikely or extremely unlikely on the basis of the numbers of physical and procedural barriers that must be violated before the event can occur. The criticality safety criterion adopted for FCF is that criticality shall not result from the concurrent occurrence of one unlikely event and one extremely unlikely event from two different categories of abnormal events. Event pairs in the same category were eliminated for the following reasons: a) some pairs are self-contradictory, b) the unlikely event may be a subset of the extremely unlikely event, c) the extremely unlikely event may be a repeated occurrence of the unlikely event, and d) in some cases it is not credible for two events in the same category to occur within the limited scope of a single operation. The KENO V.a Monte Carlo code[16] was used to analyze the event pairs.

Criticality analysis was used to guide container and equipment designs and to confirm the criticality safety of the final designs. The criticality analysis established the boundaries of the operating envelope for processes in FCF. This analysis determined the upper limits on process batch sizes, the mass limit for each container/material form combination, and the mass limit for each operation in each zone. The criticality analysis was also used to refine and improve the operating procedure for each operation to ensure criticality safety. Over 2000 cases were evaluated.

6. Summary

The operations in FCF range from novel processes such as electrowinning and cathode processing to routine procedures such as fabrication of fuel elements and subassemblies. Because of the diversity of processes and operations in FCF, no single form of control was adequate to ensure criticality safety on a facility-wide basis. The FCF nuclear criticality safety program integrates physical design, administrative controls, nuclear criticality analysis, the Mass Tracking System, and process models into a comprehensive program that maximizes facility efficiency and batch throughput within the envelope for safe operations.

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VI. FUEL CYCLE DEMONSTRATION

A. Status of Facility Modifications

Facility modifications progressed steadily during this report period and significant milestones were achieved. Many construction activities were completed and installation of several key facility systems was accomplished. Checkout and readiness review of the facility systems, modifications, procedures and safety documents required for safe operation of FCF using depleted uranium were completed. Authorization to begin FCF process equipment operations using depleted uranium was granted by DOE on schedule April 30, 1993.

Operation of the fuel fabrication equipment using unirradiated enriched uranium and plutonium was scheduled to begin in early FY 1994. This assumed that DOE would authorize this startup phase rapidly following completion of the applicable construction and documentation in September. This milestone was delayed, however, because reactivation of systems which were unmodified and presumed to be operational caused several key delays. The argon atmosphere cooling and purification system reactivation was especially difficult because of leaks and poor performance. In addition, preparation of the documentation packages required for the independent Argonne and DOE readiness reviews required significantly more engineering effort than had been anticipated.

The following describes the significant activities which were accomplished in FY 1993.

1. General Facility Improvements

Improvements of the general facility for support of the fuel cycle demonstration continued. The most significant single modification to the general facility in FY 1993 was the provision of a large new opening in the operating floor to permit transfer of large equipment to and away from the new Hot Repair Facility bag transfer rooms in the basement. Construction of this opening (approximately 2 by 2.5 m) required cutting out a large section of the operating floor, including a support beam, and replacing the section with a new floor and beam structure. The task took significant structural analysis and several weeks

of construction. Figure VI.1 shows the hole cut in the operating floor for installation of a new hatch. Other improvements included installation of support beams in the ceiling of the air cell operating corridor to enable maintenance of the master-slave manipulators, remodeling of four FCF staff offices, roof modifications to the personnel entryway, completion of construction of the radiation safety office and the new women's locker room. The north wall of the women's locker room forms a portion of the new isolation barrier between the argon cell operating corridor and the air cell corridor.

2. Transfer Systems

The two new transfer systems (the small equipment lock and the transfer tunnel cart, ram, hatch shield and platen restraint system) between the air and argon cells were completed and checked out. The most significant part of this activity was completion of the Platen Safety Restraint installation and checkout of the computer control system software. The operational checkout included integrated operation of the hydraulic rams, the east/west transfer cart, the platen safety restraint and the shielded hatch for the air cell. All of this equipment operated satisfactorily after a few bugs in the software and hardware were resolved. Figure VI.2 shows the equipment transfer lock platen safety restraint.

3. Hot Repair Facility (HRF)

Relatively little work was done in the HRF during this report period because of lower priority. This facility is needed for irradiated fuel processing support (to remove high gamma radiation contamination from in cell equipment needing repair) and is not essential for fabrication of cold fuel. It is scheduled for completion as one of the final Project tasks. However, acceptance testing of the crane in the Suited Entry Repair Area (SERA) was completed, the glove port shielding was installed and a hole in the SERA ceiling left by removal of an old air duct was filled with concrete for shielding.

Fig. VI.1. Cutting FCF Operating Floor for a New Hatch (ANL Neg. No. 103-W12153)

Fig. VI.2. Equipment Transfer Lock Platen Safety Restraint (ANL Neg. No. V12222)

4. Argon Cell

In preparation for filling the argon cell with argon in December 1993, several final cell closure tasks were completed. Installation and acceptance testing of the new lighting system for the cell was completed. This change provides sodium vapor lamps in place of the original mercury vapor lamps and decreased the heat load from lighting by about a factor of two. Unused utility penetrations were inspected, seal welded, and leak checked. Floor penetrations were capped for later remote installation of the process equipment feedthrus that make the final seal as well as provide the necessary electrical power and instrumentation for the equipment. The master-slave wall feedthrus were installed. The cell windows were cleaned and the window which had been removed to allow personnel entry was reinstalled in the wall cavity.

5. Argon Cell Atmosphere Systems

The combined argon cell atmosphere system includes three reactivated systems and three new systems. The reactivated systems include the cell cooling system, the atmosphere recirculation system and the purification system. The new systems include a normal feed/bleed system, an emergency argon supply system, and a computerized temperature/pressure control system. Another new system, the Safety Exhaust System, was added to provide for filtration of exhaust under design basis accident conditions. This system is covered separately below.

The feed/bleed system is the normal system used for control of the nitrogen concentration and the mass of argon in the cell through a continuous but variable purge of fresh argon into the cell. The emergency argon supply system provides a backup supply of argon which can automatically be introduced into the cell in case of rapidly decreasing pressure. The pressure/temperature control system is a computer control system which monitors the pressure and temperature of the cell and controls the recirculation, cooling, feed and bleed as necessary to maintain the cell pressure within the normal operating range below atmospheric pressure. It also records the feed and bleed of the system to provide a record of the amount of argon exhausted to the atmosphere as required by the Idaho Air Quality permit. Each of these new systems was completed, tested for operation and is now in operation. Problems with these systems have been relatively minor with

acceptable but lower than desired exhaust flow from the feed/bleed system being the most significant

Reactivation of the old systems which were functional at the time FCF was shutdown for refurbishment became the most significant unanticipated problem for the Project during this report period. Recirculation system problems included bearing failures in the blowers, instrumentation failures, and leakage in the piping. Purification system problems were similar but included flow anomalies. The most serious, time consuming and expensive problems encountered were associated with the freon refrigeration system for cell cooling. By the time all of the problems were discovered, this system had been completely overhauled. The condensers were retubed, a crack in the outer wall of a condenser was repaired, the compressors were rebuilt including redesign of the bypass flow to prevent overheating and numerous freon leaks had been fixed. In hindsight, the system should have been replaced but the problems were encountered sequentially rather than at the onset.

As a result of these problems, filling of the cell with argon was delayed from September 1993 until December 1993.

6. Safety Equipment Building and Safety Exhaust System

Construction of the new Safety Equipment Building (SEB) was completed including the building heating and ventilating and fire sprinkler systems. This is a concrete building designed to survive the design basis earthquake and winds at ANL-West. It houses and protects the equipment that provides for filtered exhaust from the argon cell in the extremely unlikely case that the cell containment boundary is breached. Because of process equipment location, no other part of FCF will contain exposed fuel or process materials with potential for significant release to the environment from such an event. In the SEB there are six separate rooms divided by concrete walls to provide complete isolation of the redundant safety systems. A tunnel running under the original FCF basement floor provides for connection of the SEB equipment to the main FCF building systems. The tunnel is also divided into two separate sections to isolate the redundant equipment from common hazards such as fire. Figure VI.3 shows the new FCF Safety Equipment Building.

Fig. VI.3. New FCF Safety Equipment Building
(ANL Neg. No. W12623)

There are three basic systems in the SEB and each is redundant. Each Safety Exhaust System (SES) consists of a blower, two stages of DOP testable HEPA filters, an oil seal pot to prevent back flow of air into the cell, an automatic valve which opens if the cell pressure increases above a set point which is above the operating range but below atmospheric pressure, and all the necessary instrumentation and controls. Both of the SES systems were connected to the FCF exhaust stack and subjected to rigorous acceptance tests to qualify them for operation. Operating performance to date has been good including appropriate response to some unplanned pressure transients in the argon cell due to atmosphere system testing. Installation, testing and calibration of this system was all completed. Figure VI.4 shows testing of the Safety Exhaust System. Each Emergency Power system consists of a 320 kW diesel generator (seismically qualified by shake test), fuel tanks and switch gear. Load test equipment is located outside the SEB and can be connected to either system as required. Installation of these generator systems was completed and they were tested to full load conditions. They are operating as expected. The third redundant system

Fig. VI.4. Testing the Safety Exhaust System
(ANL Neg. No. V12885)

in the SEB is the exhaust stack monitoring system. This system monitors for radioactive gases and particles in the exhaust from the FCF. The system includes some existing equipment which was interconnected with new detectors, isokenitic sampling manifolds and pumping equipment.

7. Radioactive Liquid Waste System

There are two aqueous waste streams associated with FCF operations. One is from removal of sodium from EBR-II spent fuel and the other is from decontamination of in-cell process equipment for hands on repair or disposal. There are no liquid wastes from the in-cell fuel refining and refabrication

processes. The Radioactive Liquid Waste System (RLWS) provides for collection, retention, filtration, ion exchange, sampling and evaporation of the two aqueous waste streams. The system consists of five tanks (selected tanks are shielded and contain neutron poisons), filters, ion exchange units, an integral evaporator, connection to the ANLW radioactive liquid waste treatment facility and all associated piping, valves, instruments, controls (standard FCF computer control system) and other supporting equipment. It is located in the basement of FCF in place of the old contaminated and inadequate system. A new concrete shielding wall was constructed around the area to provide shielding and ventilation control.

The old system was removed for disposal. A new floor with tank anchor embedments adequate for restraint of the tanks from earthquake loads was constructed and the five new tanks were installed. Neutron poison rods and shielding were installed for the Interbuilding Cask (IBC) Holdup tank. Piping, valves, blowers and pumps necessary for restart of the IBC wash stations for removal of sodium from EBR-II spent fuel were all installed. The final connections to the existing wash station piping was accomplished and the tank vents were connected to the Air Cell exhaust system. Figure VI.5 shows installation of the IBC Holdup Tank for the Radioactive Liquid Waste System.

8. Miscellaneous

An operator control station (OCS) terminal was installed in the radiation safety office for facility radiation instrumentation monitoring.

Removal of the original building exhaust fans was completed and they were replaced with new fans to provide a significant improvement in reliability and reduce maintenance. The normal and emergency power connections to these new fans were completed and acceptance tests were successfully conducted.

The structure of the new cask cart was completed and it was transferred into the basement cask tunnel for final assembly. This allowed the cask tunnel isolation wall to be completed as required for balance of the building ventilation system.

Fig. VI.5. Installing the IBC Holdup Tank for the
Radioactive Liquid Waste System
(ANL Neg. No. W12265)

B. Status of Process Equipment

During FY 1993, no major changes in the process equipment functional requirements or locations were implemented. The process equipment was being designed, fabricated, and tested. Figures VI.6 and VI.7 shows the various stages of completion for each piece of equipment. The different phases are design, fabrication, assembly qualification phase (Phase I), the remote qualification (Phase II), transfer in-cell and in-cell qualification (Phase III). The design and fabrication phases are the traditional engineering definition of these activities. In the assembly qualification phase, the various components of the machine are configured and tested as an integral unit. This phase is usually performed under the supervision of the designers. During the remote testing, the equipment is assembled and tested for remote operability and maintenance using only master slaves and electro-mechanical manipulators identical to those available in-cell. This testing is performed in the mock-up shop in FCF. Typically, the assigned system engineer and technicians will perform this work.

Fig. VI.6. Refining Equipment Status

Fig. VI.7. Fabrication Equipment Status

This testing familiarizes them with the equipment's characteristics and provides training. The transfer of the equipment in-cell involves the physical movement of the equipment into the cell where it is reassembled. The in-cell qualification demonstrates the equipment meets its functional requirements before being declared operational.

All the equipment made significant progress and should be operational by the end of FY 1994. The following sections discuss the process equipment in the order that the fuel is processed and fabricated into new fuel.

1. Vertical Assembler Disassembler (VAD)

The Vertical Assembler Disassembler removes the individual fuel elements from the irradiated subassemblies. This machine is an existing machine that needed some modifications to meet new requirements. Several minor modifications were completed but most work has been postponed until FY 1994 since the equipment is not needed until operation with irradiated fuel begins.

2. Element Chopper

The Element Chopper shears the fuel elements into fuel segments which expose the fuel for dissolution in the Electrorefiner. This equipment was installed in FY 1991 and tested during FY 1992. No activities were completed during FY 1993.

3. Electrorefiner

The Electrorefiner is the key process equipment which separates the actinides from the fission products. The electrorefining operations will occur at workstations P3 and P4. P3 contains the Electrode Assembly and Disassembly Machine (EADM). This machine provides remote handling support to the electrorefining operations. The fuel dissolution baskets containing the element segments are attached to the electrode shafts. After the electrorefining operations, the solid cathode is harvested from the electrode mandrel, the liquid cathode is removed from the liquid cathode crucible and the cladding hulls are emptied from the dissolution baskets. Also, electrode assembly maintenance

operations and other support operations such as handling and packaging of Electrorefiner samples are performed with this equipment.

Design, acquisition, and qualification activities related to the FCF Electrorefiner System continued in FY 1993, and culminated in the shipment of all of the Station 4 equipment and control cabinets, except for the Cover Gas System, in mid-June.

Major design work included the Liquid Cadmium Cathode Assemblies, the Electrode Assembly and Disassembly Machine (for Station P3) and special fixturing for this machine. Three separate final design reviews were held for these items and preparation of fabrication packages was initiated, although resource limitations brought Liquid Cadmium Cathode activities to a virtual standstill after the review. An Elevator and Rotator Assembly Transfer Lift Stand, for use in moving equipment through the Suited Entry Repair Area and into the FCF process cell, was designed. Many smaller, but nonetheless essential, component designs and design revisions were also accomplished, including new Valve and Latch Assembly valve actuators, Instrument Assembly components (primarily Level Detectors, Reference Electrodes, and Corrosion Samples), and Cover Gas System Components. A final design review also was held for the OCS/PLC software systems, and final wiring drawings were completed for all Electrorefiner systems except the Electrode Assembly and Disassembly Machine and its control cabinet.

Acquisition, outfitting, and checkout was accomplished on all of the above equipment, including Control Cabinets 2 (Cover Gas System and Liquid Cadmium Cathode Assemblies) and 5 (Electrode Assembly and Disassembly Machine), as well as previously designed equipment such as the Cable Festoon Assemblies and Electrode Storage Fixtures, but not the Liquid Cadmium Cathode Assemblies themselves nor the Instrument Assembly Reference Electrodes. Plans were prepared and Phase I qualification testing of the entire Electrorefiner was completed, both as subassemblies and as an integrated system, before shipping. The Cover Gas System was qualified as a separate integrated system, since the Electrorefiner was shipped and already undergoing Phase II remote qualification testing in the FCF mockup shop at ANL-West, before the Cover Gas System was ready. At the end of the fiscal year, the Cover Gas System was poised for

shipment and the Electrode Assembly and Disassembly Machine was ready for initiation of Phase I qualification testing.

The main Electrorefiner vessel and electrode assemblies were assembled in the FCF mockup shop as shown in Fig. VI.8. All the final cabling was completed so the equipment could be tested in its final configuration. The remote assembly of the equipment was tested and several necessary modifications were identified and implemented. Remote maintenance testing was started and will continue during FY 1994.

Seismic analysis packages were completed for the Electrode Storage Fixtures, Electrode Assembly and Disassembly Machine and continued for the Electrorefiner assembly itself. Numerous structural and thermal analyses for other components were completed and reviewed.

4. Cathode Processor

FY 1993 was a very productive period for the IFR-FCF Cathode Processor with the completion of a number of major milestones. The first major milestone was the completion of the design and fabrication of both in-cell and prototype Cathode Processors. The second major milestone was achieved in February of 1993, the completion of the Phase I qualification on the in-cell Cathode Processor. The third major milestone was completed in March of 1993, the shipment of the in-cell unit to ANL-W. Reassembly of the in-cell Cathode Processor (shown in Fig. VI.9) in the FCF mockup area was completed and the Phase II qualification testing was initiated in May of 1993. The remainder of FY 1993 was spent on remote handling. All of the Phase II software validation efforts were completed with the exception of the integrated system functional testing.

With the shipment of the in-cell unit to ANL-West the Cathode Processor Group at ANL-East intensified its effort on the Prototype Cathode Processor (PCP). The PCP was moved into the Prototype Test Enclosure in March and additional instrumentation to the PCP internals was installed. The PCP Phase I qualification was completed in May 1993. The first Phase II hot qualification tests (the hot characterization testing) were initiated in June, 1993. The remainder of FY 1993 was spent learning about hot operations of a Cathode

Fig. VI.8. Electrorrefiner Being Assembled in ANL-West Mockup Shop (ANL Neg. No. 12376)

Fig. VI.9. Cathode Processor in ANL-West Mockup Shop (ANL Neg. No. 103-W12888)

Processor. The hot characterization testing completed in FY 1993 did not process any Cd, salts, or DU. A number of problems arose, and most were resolved. Some of the major problems that arose were: 1) an extreme outgassing problem that coated the interior of the PCP with a "coal tar" like substance. The source of this problem was determined to be the graphite in the process crucible and the furnace liner. A solution was proposed, namely to purify the graphite, but tests had not been completed on this solution. 2) The PCP could not attain the design temperature of 1370°C. The most promising modifications to improve the thermal behavior were determined to be improved thermal insulation and improved coupling between the induction coil and the furnace liner which is also the furnace inductive susceptor. However, these proposed solutions had not been tested by the end of FY 1993.

5. Injection Casting Furnace

The Injection Casting Furnace shown in Fig. VI.10 was transferred into the argon cell and assembled. During initial testing, several problems were discovered and solutions were implemented. One problem was the overheating and damaging of a bus bar insulator as shown in Fig. VI.11. A design modification and the use of a higher temperature insulator solved this problem. Another problem was the higher than expected heating of the casting furnace shell. A combined numerical-empirical model was developed to investigate various process and design changes. This model and the final acceptance of the equipment will be completed during the first depleted uranium casting which will be early FY 1994.

6. Fuel Pin Processor

The Fuel Pin Processor originally was designed to remove the quartz mold from the fuel pin, shear it to length, and measure length, weight, diameter, and straightness. Recent tests with ternary fuel in the Experimental Fuels Laboratory have shown that the demolding operation would not work as configured on the pin processor. For this reason, a new piece of equipment was designated to perform this function at the pin handling station P9.

Fig. VI.10. Casting Furnace Installed in the Argon Cell
(ANL Neg. No. V12377)

Fig. VI.11. Damaged Casting Furnace Bus Bar Insulator
(ANL Neg. No. V12899)

The pin handling equipment was designed, fabricated, and assembled during FY 1993. The out-of-cell qualification of this equipment was also started. This equipment shown in Fig. VI.12 was able to remove quartz molds off of glass rods. This testing provides high confidence that this new equipment will meet ternary fuel fabrication requirements.

The Pin Processor was assembled in the mockup area and completed its out-of-cell remote qualification. So the operations personnel could gain more experience with the equipment, the pin processor was transferred to another location for additional testing. Both the pin processor and pin handling equipment will be transferred in-cell early in FY 1994.

Fig. VI.12. New Pin Handling Equipment for Demolding Cast Fuel
(ANL Neg. No. W12192)

7. Fuel Element Welder

The Fuel Element Welder completed the qualification phase (Phase I) by completing the software necessary to run the equipment in automatic mode. The automatic sequence will inspect the cladding, fill the element with tag gas, insert a top end plug, weld the end plug and inspect the weld. This sequence is repeated until all elements (36) in an Element Fabrication Magazine (EFM) are welded.

The Element Welder out-of-cell qualification phase was completed and several minor modifications were made to assist in remote operations. All in-cell wiring was completed with radiation resistant wiring. After completing the out-of-cell qualification, the equipment was moved to another room so that welding parameters, gas tag filling parameters and inspection criteria could be developed. More than 300 welds were performed to support these efforts. Figure VI.13 shows a schematic of the vision system images that are produced by using the digitized images and using edge detection and data fitting routines. The sequence of the present inspection process is shown in Table VI.1. The data from this process have been used to develop 12 different acceptance criteria. Initial testing has shown these criteria work but further development is needed to finalize the parameters.

Fig. VI.13. Schematics of Pre- and Post-weld Vision System Images

TABLE VI.1. Sequence of Major Inspection Steps

Front View	
PRE-WELD	POST-WELD
1. Check insertion of the end-plug to the top of the fuel element	7. Detect the edges of the dome height through vertical measurements
2. Calibrate the front-view image	
3. Establish the base coordinate for the vertical measurements	
Top View	
PRE-WELD	POST-WELD
4. Calibrate the top-view image on perimeter points A, B, C, and D	8. Estimate the coordinates of the center of the top-view circle
5. Calculate the coordinates of the center of the top-view circle	9. Measure the coordinates for the perimeter points A', B', C', and D'
6. Perform 360 radii measurements at 1° intervals around the circle	10. Perform 360 radii measurements at 1° intervals around the weld

8. Fuel Element Settler

The Element Settler heats the finished elements to 500°C and uses an impacting force to settle the fuel slug to the bottom of the cladding and to develop a good sodium bond. The settler was assembled in the mockup shop and underwent remote testing. Also, thermal tests were performed and thermal baffles were redesigned to produce the correct thermal profile. The final thermal profile is shown in Fig. VI.14. Operations with sodium and depleted uranium fuel elements were started to develop the process parameters which will produce acceptable fuel elements. When the parameter development is completed, the settler will be installed in-cell.

9. Fuel Element Inspection

The Element Inspection verifies that the fuel slug is positioned properly at the bottom of the cladding and measures the sodium height above the slug to verify an adequate sodium bond has been made. The X-ray unit, which was received in FY 1992 and temporarily installed in an X-ray inspection cell, was further tested this year. These results show the equipment should be able to perform all the required inspections.

Fig. VI.14. Final Settler Thermal Profile

The shielding analysis and a preliminary design were completed. The design fabrication and qualification of this equipment will be completed in FY 1994.

C. Process Control and Accountability

The Fuel Cycle Facility is implementing a distributed process control system that will be integrated together to provide a process control, process monitoring, data archival and material tracking system. The interconnecting networks and all major components were installed and tested this past fiscal year. Significant progress was made on implementing the Operator Control Station (OCS)/Programmable Logic Controller (PLC) software for both the facility and process equipment. The Mass Tracking System (MTG) hardware was installed in the facility and testing started on the software.

1. Operator Control Station (OCS)/Programmable Logic Controller (PLC) Software

The Operator Control Stations (OCS) provide the principal operator interfaces to the facility systems, process equipment, and mass tracking computer. The Programmable Logic Controller interfaces with the field instrumentation, converts the signals to a digital signal and provides the direct machine control. Figures VI.15 and VI.16 show the various components in the facility and process control system.

All facility remote chassis which contain the PLCs and input/output modules were designed, built, installed and tested. Software was written to provide the necessary monitoring for the stack monitor, safety exhaust system, power systems, and ventilation system. The control and monitoring software for the argon atmosphere and transfer tunnel system was also written. The software qualifications for all these systems was started and will be completed during FY 1994.

The OCS/PLC software for the process equipment is developed in conjunction with the machine developments. The pin processor, element welder, and element settler software was independently qualified during the out-of-cell testing phase. The electrorefiner and cathode processor software was completed and the first phase of qualification was completed during the assembly qualification of the equipment.

2. Mass Tracking (MTG) Software

The Mass Tracking system has two SUN Sparc station computers that operate under the UNIX operating system. Also, electronic weighing balances will be installed in-cell so that new weights are established when material is transferred between containers and weights are confirmed to verify identity when items are transferred. During the FY 1993, the software to support casting operations was written and tested through the first two phases of qualification. The first three balances were installed in-cell and integrated testing of the mass tracking software was started. This software will be fully implemented during FY 1994.

Fig. VI.15. Process Monitoring and Control and Interconnecting Networks

Fig. VI.16. Facility Monitoring and Interconnecting Network

D. Preparations for FCF Operations

Throughout the year, facility operations personnel were fully involved with activities necessary for startup. The staffing level is up to 13 systems engineers, 8 chief technicians, and 19 operations technicians. Development of operating procedures, maintenance instructions, and the FCF training for startup of depleted uranium operations was completed. Engineering and technical support personnel completed work on reactivating existing facility systems and refurbishing and installing remote handling equipment necessary for argon cell closure and depleted uranium operations.

The final DOE Final Safety Analysis Report (FSAR) questions were resolved and the FSAR was approved by DOE on November 16, 1993.

The Technical Safety Requirements (TSRs) document was approved by the Nuclear Facilities Safety Committee (NFSC) and submitted to DOE for review and approval. It was also approved on November 16, 1993. The Criticality Hazards Control Statement was accepted by the NFSC and submitted to DOE for review along with the TSRs.

Preparation of a detailed plan for phased facility startup was completed. The plan covered preparations for depleted uranium processing (Phase 2), cold Pu processing (Phase 3) and irradiated fuel processing (Phase 4) with emphasis on what systems are required for each and what review and approval levels are required. The approach taken was to break the remaining work into discrete packages which will be reviewed and approved as they are completed. The process was successfully followed for the Phase 2 review and DOE authorization for Operations with depleted uranium was granted on April 30, 1993.

E. Preparation for Demonstration Experiments

As part of the preparation for the Fuel Cycle Facility (FCF) operations, the IFR Fuel Cycle Demonstration Program Plan was prepared. It describes the overall goals of the IFR Fuel Cycle Demonstration and the means by which these goals will be accomplished. The planning and review processes, including the required documentation, are described in detail.

As part of the Program Plan, the first detailed listing of the feedstocks available for the demonstration was prepared. This listing includes irradiated subassemblies from EBR-II and cold feedstocks that can be used in FCF.

Operating strategies for the pyroprocessing demonstration were updated. The goal of this work is to develop a processing campaign that provides enough fuel to meet the needs of EBR-II and that provides the data needed for the further development of the Integral Fast Reactor's pyrochemical process. The startup, cold plutonium, and irradiated fuel operations of all pieces of equipment in FCF have been considered.

Included in the report is the proposed Electrorefiner operating strategy that was modified since the 1992 Annual Report. The startup phase of the Electrorefiner still includes loading the vessel with salt and cadmium and performing pretreatment operations. The initial operations will employ depleted uranium and then plutonium will be added to test the equipment and operating procedures prior to the addition of irradiated material to the system. At the end of startup, the plutonium to uranium ratio in the salt will be appropriate for normal operations.

Because only a limited amount of irradiated ternary fuel is available, simulated material will be used to demonstrate adequately the pyrochemical process. The simulated, irradiated, ternary fuel will consist of irradiated binary fuel mixed with cold plutonium metal. With this loading, the recovery of the transuranic elements in the presence of high concentrations of fission products using liquid-cadmium cathodes will be demonstrated thoroughly. The limited amount of irradiated ternary fuel will also be processed at the appropriate time during the operating campaign.

A typical batch in the first campaign will consist of an 18 kg loading of the simulated-ternary fuel. Anodic dissolution and recovery of solid cathodes will be followed by a liquid cathode operation. The solid and liquid cathodes will be used for the production of fuel for EBR-II.

The operating strategy for the first campaign, including startup, has been modeled using the PYRO code. Work is continuing to optimize the operating

strategy and to use the results to make an estimate of the samples required during the first campaign.

The PYRO simulation is also being applied to the Electrorefiner waste operations after the first operating campaign. This model predicts that the Electrorefiner vessel alone can be used to process the bulk of the salt after the campaign. Portions of the Electrorefiner contents will be processed using more advanced equipment and techniques as part of the waste demonstration. A document describing the Waste Demonstration Strategy is in draft form and will be completed in 1994.

The Electrorefiner process chemicals, including eutectic salt and cadmium, were ordered. Delivery is expected in early FY 1994.

VII. LIQUID METAL REACTOR TECHNOLOGY RESEARCH AND DEVELOPMENT

A. Temperature Effects on Seismic Isolator Elastomers

An Elastomer Testing Facility has been established at Argonne to perform high-precision dynamic analysis of small samples of the elastomers used in seismic isolation systems, designed to protect nuclear reactors from damage caused by earthquakes. The seismic protection system uses high-damping steel-laminated elastomeric bearings to support the reactor structures and isolate them from damaging ground motion. In the United States, the current reference design for the Advanced Liquid Metal Reactor (ALMR) uses laminated bearings for seismic isolation. This base isolation technology was proven effective in the area of southern California hit by the January 17, 1994, Northridge earthquake. The USC University Hospital and the LA County Fire Command and Control Facility, two of several base-isolated structures, survived the earthquake without damage and continued operation. In contrast, several nearby buildings and hospitals suffered extensive damage and had to be closed. A typical base isolation installation may require a few hundred of these bearings which are typically about 1 m (36 in.) in diameter and 0.4 m (15 in.) high. The bearings are constructed from alternating layers of high-damping rubber ~1.25 cm (~1/2 in.) thick and steel plates. They are generally designed for horizontal shear strains between 50% and 100% and can be expected to sustain two to three times these levels for beyond design basis loading conditions. Elastomeric bearings are currently designed to provide a fundamental horizontal system frequency between 0.4 and 0.8 Hz and are expected to operate between -20°C and +40°C.

The goal of the work at Argonne is to produce a fully-validated bearing analysis code (a version of the Neptune finite-analysis code) based upon the data from dynamic tests of small samples (coupons) of the elastomer and tests of full- and scale-sized bearings. The coupon tests are carried out at Argonne and the bearing tests are carried out at the Earthquake Engineering Research Institute of the University of California, Berkeley. To assure proper performance of isolation bearings, it is necessary to characterize the elastomer's response over the expected variations of shear-strain level, temperature and frequency. In the case of frequency variation, the frequency range must span the bearing acceptance

test frequency, which may be as low as 0.005 Hz, and the design frequency. Similarly, the variation in mechanical characteristics of the elastomer must be determined over the design temperature range, which is between -20°C and $+40^{\circ}\text{C}$. Results from earlier tests to determine the strain and frequency dependencies have been reported elsewhere.[1-3] During FY 1993 tests were run to determine the variation in the mechanical behavior of a typical elastomer over a wide temperature range including the design range.

The tests are performed on one of Instron Corporation's new generation of 8500 series universal testing machines which use a computer-based system to provide full digital control of the machine. The servohydraulic machine has a 5 kip actuator and sufficient hydraulic supply to test at frequencies up to 100 Hz. The machine is connected to a 386DX computer that has been programmed for test control, data acquisition and data processing. A small environmental chamber, only large enough to contain the specimen and its holder, is mounted on the testing machine to carry out the testing at temperatures that can range from -70°C to 200°C . The chamber is heated by electrical resistance heaters and cooled using the vaporization of liquid CO_2 . This machine is set up to perform shear tests on three-bar specimens with loadings of either a stress relaxation type or cyclic type up to 375% strain using a 5-mm thick specimen. Frequencies up to 100 Hz can be handled at very low strain levels. Two sample points on the system performance limiting curve would be 5-mm cyclic amplitude at 10 Hz and 40 mm at 1 Hz.

To obtain data appropriate to earthquake type events, small samples of the elastomer are subject to fully-reversed cyclical shear loading. The test

Fig. VII.1. Three-bar-lap Shear Specimen

specimens consist of two elastomer pads 5-mm thick by 25-mm square mounted in the three-bar lap configuration (Fig. VII.1). The specimens are fabricated in a mold by bonding the elastomer to the steel bars during vulcanization. The elastomer tested during FY 1993 is a high-damping natural rubber compounds provided by Oil States Industries of Arlington, Texas, identified as proprietary compound No. 243-62. Each test was performed at the desired temperature, strain and frequency using a sine wave loading for six consecutive cycles. The values of shear modulus, damping, etc. reported here were calculated using the 100 data points from the hysteresis loop of the sixth cycle. Since the three-bar specimens contain two pads, the results represent an average for the two pads. In most elastomer testing, the amount the specimen has been worked before the test can have a marked effect on the results. Reference to a scragged state usually refers to a prior loading of approximately 30 cycles at 0.5 Hz and the desired strain level or extensive testing within the past hour. Reference to an unscragged state usually indicates a 24-h rest period.

Two quantities that characterize the behavior of an isolator are the shear stiffness and the energy dissipation. The shear stiffness of the isolator is a quantity that governs the fundamental horizontal frequency of a base isolated system, and the energy dissipation controls the amplitude of the system, primarily at the fundamental system frequency. Because of the relatively high shear modulus of the steel, it is the shear modulus of the elastomer that determines the shear stiffness of the bearing. There are several different definitions being used for this quantity. In this work the effective shear modulus, G_{eff} , is used, and it is given by

$$G_{\text{eff}} = \frac{\Delta\tau}{\Delta\gamma} = \frac{\tau_{\text{max}}^+ - \tau_{\text{max}}^-}{\gamma_{\text{max}}^+ - \gamma_{\text{max}}^-} \quad (1)$$

where τ_{max}^+ and τ_{max}^- are the maximum positive and negative shear stresses during a load cycle and γ_{max}^+ and γ_{max}^- are the maximum shear strains. Energy dissipation is an important property of isolation systems, and it can be characterized as the energy dissipated during a cycle, W_D . For elastomeric isolation systems, linear analysis is often performed to evaluate performance for strain levels of 100% or

less. For linear analysis, a damping ratio is useful and we are using an effective damping ratio, β , that is given by

$$\beta = \frac{W_D}{2\pi W_S} \quad (2)$$

where W_S is the energy stored during a cycle.

The influence of temperature on stiffness and damping was determined from a series of cyclic tests that were performed at a frequency of 0.5 Hz and at both 50% and 100% strain. Because of the expense of cooling the environmental chamber a group of tests at a particular shear strain level was conducted in about 5 h over the entire temperature range. The specimen was held only for about 2 min at the required temperature and the test performed. Since there was concern about the effect of these short hold times, another test sequence was run for 3 h with the specimen held at -20°C and no variation in the test results was observed. The results are presented in Figs. VII.2 and VII.3. The effective modulus values show a significant increase as the temperature is reduced. The unconnected data points (triangles) for the 100% tests illustrate the effect of temperature on recovery. The tests denoted by triangles had short rest times and at temperatures below room temperature yielded lower values of effective modulus. The material recovers rapidly at temperatures above 30°C yielding identical values of modulus. The effective damping ratio shows a significant increase with lower temperatures and this is very evident in the shape of the hysteresis loops. The hysteresis loops for the sixth cycle at two different temperatures are shown in Fig. VII.4.

The tested elastomer exhibits variations in both stiffness and damping over the design temperature range. The stiffness has been found to be 1.8 times greater at the lower design temperature limit than at the upper limit. The seismic isolation system designer must take into account these variations in properties with temperature in order to optimize the performance over the design temperature range. Data is available on this temperature related variation for compounds produced in Italy and Japan (Bridgestone Corp.). The Bridgestone compound has about the same variation in properties as presented here, while the

Fig. VII.2. Variation of Effective Modulus with Temperature

Fig. VII.3. Variation of Damping with Temperature

Fig. VII.4. Hysteresis Loops for 100% Shear Strain

Italian rubber has a much greater property variation with temperature. The producers of high-damping rubber compounds are attempting to develop new compounds that do not have so much variation in mechanical properties with temperature.

B. Uplift and Rocking of a Deformable Body Subject to Base Excitation

1. Background

The peer review of the seismic criteria for unanchored equipment in the air and argon cells of FCF raised a question concerning the possible uplift and subsequent stable rocking of equipment during the DBE.[4] The reviewer agreed that the original analysis showed that unanchored, rigid equipment would not uplift during the DBE but expressed concern that the dynamic response of deformable equipment could result in uplift. The potential problem is that the reaction forces to the dynamic response of the deformable portion of the equipment could provide additional uplift moment at the equipment base causing

uplift of the base and subsequent rocking motion of the equipment. Such rocking motion, if severe enough, could damage the base of the equipment.

ANL agreed to conduct an assessment of all unanchored equipment in the cells to determine if such uplift could occur and, if so, to assess the effect of the ensuing rocking motion on the equipment. This report summarizes the analysis methods developed for the rocking assessment; they constitute a significant advancement in analysis methods for the seismic response of unanchored, deformable equipment. The detailed development of the rocking model and the use of these methods in the assessment of equipment rocking in the FCF cells is documented in a separate report.[5]

2. Introduction

The rolling and sliding motions of a rigid body subject to gravity and supported by a plane surface are treated in elementary texts on dynamics. Rocking of a rigid body supported by a horizontal surface which experiences oscillatory accelerations due to an earthquake has been discussed by Housner.[6] If the body is deformable there is a potential for the dynamics of the body deformations to couple with the rocking mode; in particular, resonances in the deformation response can develop sufficient reaction moment at the base to cause base uplift which would not occur if the body were rigid. The purpose of this work was to develop a model suitable for studying this phenomena including both overturning and the magnitude of the uplifts and impacts occurring during stable rocking motions.

3. Mathematical Model

The equations governing the plane motion of a deformable body with rocking boundary conditions supported by a horizontal flat surface subject to vertical and horizontal accelerations were derived based on consideration of the fundamental principles of mechanics, impact conservation assumptions, and assumed mode methods. These equations depend on dynamic parameters of the body which are defined in terms of integrals of assumed modes of deformation. The number of assumed modes is arbitrary. The dynamic parameters defining a specific body include a geometric parameter related to the rigid body stability, deformation

parameters associated with mass, stiffness, dilation, shear and Coriolis properties, and participation factors for the base excitation. The model includes expressions for the location and magnitude of the reaction forces and impulses at the supporting surface and conditions for uplift and overturning. Motions which involve uplift but not overturning are termed rocking motions and are characterized by impacts with the supporting plane and loss of energy for the body. The model includes expressions for changes in the generalized velocities and energy during impact. Integration of the equations requires some care because arbitrarily high frequency rocking motions can occur.

4. Applications

Under most circumstances the fundamental deformation mode will dominate the uplift moment; higher modes may contribute to the internal stress state of the body but normally they will require minimum reaction moment at the base. Thus if the normal modes in a pinned base support configuration are known, a single mode of deformation is sufficient for a tipping analysis of a deformable body. With this observation in mind a FORTRAN computer code was written to integrate the rocking equations for a deformable body with a single deformation mode. A Runge-Kutta integration scheme was used along with the logic to determine the time of impacts, check for overturning, eliminate chatter, and retain records of extreme values of uplift, impulse, and energy loss. The code runs efficiently on a 386 PC computer.

The computer simulation of potential rocking for FCF in-cell equipment showed that rocking did occur under design basis earthquake conditions for most equipment. The few components that did lift had very small uplift, less than 3 mm, with minimal energy absorbed. The subsequent motion could be better described as "chattering" than as rocking. No damage to any in-cell equipment was indicated.

C. Irradiation of EM Pump Samples in EBR-II

EM Pump Insulation Irradiation Test Program

The objective of this program is to assess and confirm the adequacy of candidate insulation materials to perform the electrical insulating function in the EM pump at conditions expected in the ALMR environment. The work involves the irradiation of selected EM pump insulation bar samples in a representative neutron flux for a sufficiently long period of time and then to determine the resulting insulating performance. Exposure of selected insulation materials to a radiation field at elevated temperatures will provide the irradiation resistance performance data necessary to insure that the insulation meets the physical and electrical requirements of the EM pump being designed for the ALMR.

The program is comprised of essentially three phases: a pretesting phase, an irradiation phase, and a posttest phase. The pre and posttest phases are designed to establish the effects of radiation on the physical and electrical properties of the insulating materials. The bar samples evaluated in this program consist of a copper bar wrapped with an insulation material and held together with a binder material. Five types of bar samples, approximately 10 in. (25.4 cm) long, using four insulation materials, three binder materials and two copper materials comprise the materials test matrix. The two copper materials are GlidCop AL-15 (C15715) and Electrolytic Tough Pitch (ETP) Copper (C11000). The four insulation materials are splitting mica, muscovite (white mica), phlogopite (amber mica) and Nextel™ (a ceramic tape from 3M Corp.). Three types of potting compounds will be used as a binder to hold the insulation together on the copper bar. One binder developed by the Corning Co. consists primarily of silicon and aluminum, the second binder is Secon-5, whose main constituents are oxides of silicon, lead and aluminum, and the third binder is boron nitride which is comprised primarily of boron nitride and aluminum oxide.

To isolate the effects of irradiation, four groups of samples are being subjected to various combinations of pretest, irradiation exposure, and posttest conditions. The four groups of samples are to be tested in the following test combinations:

- Aged, Irradiated/Temperature Tested and Aged
- Aged, Temperature Tested and Aged
- Irradiated/Temperature Tested
- Temperature Tested

The aging tests will subject the samples to an applied voltage in an air atmosphere for a specified time (i.e., up to a maximum of 500 h) at a temperature of 1346°F (730°C). These accelerated aging tests will be performed on some samples before and after irradiation. Some samples, to aid in the determination of the effects of irradiation, are not irradiated but are rather subjected to the same duration and temperature in a nitrogen atmosphere as those samples being irradiated (Temperature Test).

The major feature of the test program is the irradiation phase in which 39 bar samples contained in three canisters are irradiated for a scheduled 90 full power days in the J2 thimble of EBR-II. The test temperature specified for the in-reactor test was 850°F (455°C). In the EBR-II J2 test location (i.e., in radial shield outside the reactor core), a neutron fluence in the range of 1.7×10^{16} to 3×10^{17} n/cm² for the samples is expected with a maximum fluence not to exceed 2×10^{18} n/cm².

1. Preirradiation Test Phase

The purpose of the pretesting is to characterize the insulation material both visually and electrically. These examinations and tests serve as the reference baseline from which the subsequent irradiation induced changes will be determined in the posttest phase of the program. In addition, the preirradiation test phase serves as a screening evaluation process for selecting the most promising insulation materials to be irradiated.

The preirradiation testing consists of the physical and electrical tests needed to benchmark the insulation samples prior to the irradiation phase of the program. These tests are essentially identical to those conducted in the postirradiation test phase and include visual examinations, photographs, physical measurements, voltage/temperature aging tests and electrical tests. The examinations and physical evaluations include determining the weights and

dimensions of each bar sample. The electrical tests that were performed included both a direct current resistance test as well as an alternating current leakage current test.

The accelerated aging tests were short duration high temperature and voltage tests performed in an oven. The test setup and conditions are similar to those used in the past for the ALMR EM pump insulation materials life testing program at ANL-E. The bar samples are placed in an oven in an air atmosphere. A high voltage (e.g, either 600 V AC or 1500 V AC depending on the bar sample type) is applied to the insulation and the leakage to ground current monitored. The temperature for these accelerated aging tests is 1346°F (730°C) with the test duration being 500 h (approximately 3 weeks) or 350 h (approximately 2 weeks), depending on the sample type.

As part of the preirradiation test phase several shorter duration (~1 day) aging tests were conducted. These tests were essentially conducted to checkout the performance of new combinations of insulating materials. Based on the results a selection of the most promising insulation materials were selected for irradiation.

The preirradiation test phase was conducted during this past year with all of the physical and electrical examinations and tests being completed. A draft report was written which summarizes the testing procedures and results. This report is presently in the final review stage.

2. Irradiation Testing

The irradiation program was conducted in the J2 thimble of EBR-II with the test designated NI-10. This irradiation facility, called the Nuclear Instrument Test Facility (NITF), is located outside the core in the shield region and is an air-cooled thimble. The 39 selected bar samples were contained in three stainless-steel canisters (13 samples in each canister). The canisters were filled with nitrogen gas and sealed initially at 1 atm pressure. The temperature histories of the three canisters were recorded by two thermocouples attached to each canister. The neutron flux at the three canister locations was determined by a neutronic calculation.

The irradiation phase of the program was initiated in EBR-II on July 31, 1993 when full power was reached at the beginning of Run 165A. Completion of the irradiation phase was achieved on December 1, 1993, after 92 full power days of irradiation at the end of Run 166A. The desired test temperatures were obtained. The upper canister operated at a temperature of about 850°F (455°C), the middle canister at about 995°F (535°C), and the lower canister (closest to the core) at about 1140°F (616°C) during the irradiation duration. The NI-10 test assembly was removed from the reactor on December 10, 1993, and placed in the storage pit. After several months of activity decay the canisters will be removed from the J2 assembly and the bar samples removed from the canisters for examination in the postirradiation phase of the program.

In conjunction with the irradiation of the bar samples, a temperature test was conducted in an oven at ANL-E. Using an identical fourth canister, 13 bar samples were inserted and sealed in a nitrogen atmosphere. This canister was placed in an oven at the highest irradiation canister temperature observed in the EBR-II irradiation (1140°F, 616°C) and was operated at this temperature for the same duration as the irradiation samples (~92 days). A comparison of the insulation electrical performance of these samples with that of the irradiated samples should help identify the influence of the irradiation on electrical insulation behavior.

3. Postirradiation Testing

The postirradiation testing phase essentially is a repeat of the series of examinations and tests conducted during the preirradiation phase. The major difference between the two phases, of course, being the fact that in this testing phase the samples are slightly radioactive. This factor influences the resulting methods and test procedures that are employed in the posttest phase. The activity level of the irradiated bar samples has been estimated analytically. In addition, three typical insulation bar samples were irradiated in a special test capsule in the NRAD test facility at ANL-W. Based on an assessment of these results the required decay time and adjusted test procedures have been established. It has been decided that these postirradiation examinations, accelerated age tests and electrical tests will be performed at ANL-W, which is better equipped to deal with radioactive materials than is ANL-E.

An aging oven has been shipped from ANL-E to ANL-W along with other testing equipment that is required for the postirradiation testing. This testing equipment has been assembled with the testing scheduled to be completed by the end of this Fiscal Year 1994.

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VIII. LIST OF PUBLICATIONS

The technical papers on the IFR Program activities, which have been published through the end of FY 1993 are listed below. The publications are categorized into the following subject areas: IFR Concept, Metal Fuel Performance, Pyroprocessing, Safety, Neutronics and Core Design, and Fuel Cycle Demonstration.

IFR Concept

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