1 of 1

DISCLAIMER

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

Actinide Behavior in the Integral Fast Reactor

Progress Report for the Period

May 1, 1992 to April 30, 1993

J. C. Courtney

Louisiana State University
Nuclear Science Center
Baton Rouge, LA 70803-5230

May 1993

Prepared for

The U. S. Department of Energy
Agreement Number DE-FG07-89ER12896

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

THE GOAL OF THIS PROJECT IS TO DETERMINE THE CONSUMPTION OF NP-237, PU-240, AM-241, AND AM-243 IN THE INTEGRAL FAST REACTOR (IFR) FUEL CYCLE. THESE FOUR ACTINIDES SET THE LONG TERM WASTE MANAGEMENT CRITERIA FOR SPENT NUCLEAR FUEL; IF IT CAN BE DEMONSTRATED THAT THEY CAN BE EFFICIENTLY CONSUMED IN THE IFR, THEN REQUIREMENTS FOR NUCLEAR WASTE REPOSITORIES CAN BE MUCH LESS DEMANDING. IRRADIATIONS IN THE EXPERIMENTAL BREEDER REACTOR II (EBR-II) AT ARGONNE NATIONAL LABORATORY'S SITE NEAR IDAHO FALLS, IDAHO, WILL BE CONDUCTED TO DETERMINE FISSION AND TRANSMUTATION RATES FOR THE FOUR NUCLIDES. THE EXPERIMENTAL EFFORT INVOLVES TARGET PACKAGE DESIGN, FABRICATION, QUALITY ASSURANCE, AND IRRADIATION. POST IRRADIATION ANALYSES ARE REQUIRED TO DETERMINE THE FISSION RATES AND NEUTRON SPECTRA IN THE EBR-II CORE.

INTRODUCTION

The irradiation data package for EBR-II is now in the draft stage. This is a massive document that provides all of the details necessary for review of the hardware to be inserted into the reactor. After some iterations, the design of the vanadium capsules has been finalized. Because of the difficulties encountered in loading the Am_2O_3 powders, the capsule has been increased in diameter. This forced a change from a standard 61 pin driver subassembly to a 37 pin subassembly; there is no way to fit the larger diameter pins into the 61 pin unit. Multigroup diffusion calculations show that the neutron spectra are not changed by this substitution in hardware. Designs of the subassemblies, vanadium capsules, and capsule holders are complete, and all related engineering drawings have been generated. All calculations done by LSU have been repeated by ANL to provide independent verification of important parameters. No significant discrepancies with the LSU analyses have been identified. The thermal analysis for the "beginning of life" irradiation conditions has been completed. The ANL work has been used to determine the isotopic and elemental purity requirements of the targets. Such a determination is necessary to forecast interferences that might compromise the gamma spectrometric analyses.

Preirradiation gamma spectroscopy and chemical analysis have been delayed by the

refurbishment of the ANL-W Analytic Laboratory. To meet the project schedule, this work has been shifted to the Analytic Chemistry Laboratory in the Chemical Technology Division at Argonne's main site near Chicago, IL. This unit operates on a strict cost recovery basis; that is, the experimenter's Division must fund the tasks completed. To help defray the \$25,000 dosimeter characterization costs, LSU has issued Purchase Order R178911 in the amount of \$5500 to cover the isotopic analysis of the ^{241}Am samples. Table 1 outlines the sensitivities required and provides the justification for sole-source specification of Argonne's laboratory to accomplish this task. At this time, we are working to resolve a problem associated with the indemnity clause in Argonne's standard "Work for Others" contract; it is in conflict with Louisiana law.

Another detailed task that has been completed is the development of the welding parameters for the vanadium capsules. Two types of dosimeters as shown in Figure 1 will be used. The Oak Ridge National Laboratory (ORNL) design holds the light metal wires; the LSU design holds the powders and actinide metal foils. Since this figure was drawn, the diameter of the LSU design has been increased to facilitate accurate loading of target materials. However, the shape and general proportions remain the same. A diligent search of Argonne records turned up several uranium and plutonium dosimeters already in capsules of the ORNL design. These have been X-rayed and pressure tested for use in this project. Since a larger diameter fuel element is needed, more of the ORNL designed capsules can be placed at the same axial location in the core. As long as the flux perturbations are low, uncertainties associated with core position are reduced; this is especially important for the sets of dosimeters at the top of the EBR-II core where the flux intensity and spectra vary rapidly. Because of the low cross-sections of all the targets for fast neutrons, no significant perturbations of neutron spectra associated with the dosimetry packages have been identified.

The first task in the study is to determine the effect on pure samples of ^{237}Np , ^{240}Pu , ^{241}Am , and ^{243}Am of exposure to the highest fast neutron flux available in EBR-II. These four minor nuclides are considered to be the controlling materials for the long-term radiation hazard of spent fuel.^(1,2) The original proposal did not include ^{243}Am ; however, this nuclide has been added after

a review of the experimental protocol. Those specimens irradiated for extended periods will be characterized by alpha, gamma, and mass spectrometry. A short (ten day) exposure in the reactor is currently scheduled for 1994. These specimens will be characterized by gamma spectrometry to determine fission rates relative to those of ^{235}U , ^{238}U , and ^{239}Pu which are well known at the axial core centerline. Neutron activation of light metal wires will be used to help characterize the neutron spectra in all irradiations. Figure 2 shows the arrangement of the dosimeters in the central (1A1) subassembly for the short dosimetry run. Measurements above and below the axial centerline of the core are valuable because commercial IFR designs will have neutron spectra somewhat softer than that of the EBR-II. The ten-day exposure will provide other experimenters an opportunity to use the reactor for short term exposures and provide early physics data to designers of larger IFRs. There is a strong possibility that all the desired information can be obtained in this irradiation. If not, several extended exposures may be required; this would drive up the experimental cost significantly.

The target materials will be placed in standard pins inserted in the center (1A1) EBR-II 37 pin driver subassembly. If extended irradiations are necessary, this subassembly will be inserted into the driver core and removed from the reactor in the normal operating cycle after an appropriate burnup is achieved. Nominally, this would involve a full power year; at the present schedule of reactor use, the subassembly would be in the core about 18 months.

Faculty and students from LSU are working with ANL personnel to provide the required analyses of the safety and reactivity considerations associated with this irradiation. All operations must meet the requirements including the most current revision of the Guide for Irradiation Experiments in EBR-II.⁽³⁾ The point depletion codes ORIGEN-II and ORIGEN-S have been used to estimate the fission and capture rates of the four target actinides in EBR-II.^(4,5) The results indicate that the relatively hard spectrum of a metallic fueled reactor is more efficient at consuming the minor actinides.⁽⁶⁾ All of the current efforts are being directed toward supporting the experimental phase of the project. Table 2 lists the LSU students who have been supported by the first three years of this grant. All of the student projects have contributed to the IFR Program; it should be noted that much of their work has been directed toward the

activation of the Fuel Cycle Facility at ANL-W. This facility is necessary to demonstrate the IFR concept by separating the actinides for recycle to EBR-II. In fact, it has equal importance with EBR-II because spent fuel processing and actinide recycle are corner stones of the IFR.

PROGRESS AND ACCOMPLISHMENTS

The major milestone in the first year of funding was the development of a test specification for the actinide burning experiments. On February 21, 1990, the EBR-II Experiment Planning and Scheduling Committee (EPSC) reviewed the specification and found it to be programmatically and technically feasible. Subassembly numbers X-502 and X-503 have been assigned to the project. We are continuing to develop the supporting documents needed for an Engineering Level II design review; this is a major effort that involves personnel at ANL-W who are familiar with the specific procedures needed to satisfy quality assurance requirements. As of late April 1993, ten Argonne professional staff members are spending 15% to 20% of their time to support the project. The manpower and funding commitments from Argonne have increased significantly as we approach the short term dosimetry exposure. Involvement of LSU personnel in the early phases of this project was a cost-effective way of developing the experimental plan.

In 1992, I reported that the selection of irradiation target materials and the design of encapsulation, the second milestone, was completed. During 1993, we modified the designs somewhat and increased the number of targets to be used in the ten day run. In addition to the four minor actinides of interest, eight other targets will be used to characterize the reactor spectra and serve as references for fission rates. Table 3 lists the reactions of interest for all twelve target materials. The original proposal addressed only measurements at the axial centerline. In 1992, it became apparent that the benefits of this experiment could be increased greatly at a small incremental cost. A better knowledge of the axial flux and spectral distribution of neutrons in EBR-II are important to the global core neutronic prediction, as axial leakage is a major component of the global neutron balance. Such predictions have a direct bearing on the calculated isotopic content of the spent fuel. Predictions of the axial flux profile in the EBR-II core are particularly difficult because of massive transport effects that influence

neutron leakage. Neutron backscattering from the lower steel reflector strongly influences the spectrum. There are little experimental data available to validate theoretical predictions for energy-dependent fluxes in the top and bottom regions of the core. No data are available for modern core configurations. Measurements made over 15 years ago are difficult to interpret because of the lack of detailed information on core loadings. Much of the earlier work at the reactor supported development of oxide fuels which have a much softer core spectra. Also, past core loadings included nonfuel materials, such as stainless steels, that further reduced neutron energies. By spanning the whole height of the pin as shown in Figure 2, spectral details important to code verification would be obtained.

The third milestone is to obtain the target nuclides. All of the light metal targets have been purchased from a commercial vendor, Reactor Experiments, Inc., and are on-hand at ANL. Our efforts to obtain the actinides had been thwarted by the loss of the Isotope Distribution Center at Oak Ridge National Laboratory (ORNL). A major task in the throughout the past two years has been locating the target materials and obtaining firm cost and delivery schedule quotations. Fortunately, we have obtained all the needed actinides from a variety of laboratories at Argonne. All of these nuclides have been assigned to this project at no cost. An order for the ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu ceramic wires by ANL from Oak Ridge in February 1992 was canceled because of the extremely high administrative and shipping charges.

The high-purity vanadium capsules or cans to contain all of the target materials must bear one or more markings to identify its contents. Two designs are to be used in the project. To contain the wires, a 0.05 inch diameter can with a 0.34 inch length designed by ORNL has been selected. Two hundred of these cans have been obtained from ORNL in 1992 at a cost to the project of \$20,550. To contain the other targets, another can has been designed by LSU and modified by ANL. The ^{240}Pu and ^{243}Am are in the form of thin metal foils which will be sliced to obtain nominal 5 mg targets. The ^{241}Am targets will be made from the Am_2O_3 powder now being characterized by the Chemical Technology Analytical Laboratory.

In addition to target material costs, the charges for loading the targets into the capsules must be

considered. Argonne personnel have performed the necessary quality assurance checks on the vanadium cans and have loaded almost all of the light metal wires. A semimicro balance has been purchased by ANL and has been dedicated to the project. As part of this effort, they will produce the appropriate quality assurance documents to satisfy Reference 3. It is estimated that the encapsulation work will cost \$150,000. This work began in February 1992 and is continuing as of the date of this report, May 1993. One reason for the slow pace of work is that glove boxes at Argonne have needed modifications to bring them up to current USDOE standards. Also, significant planning has gone into the transfer of the americium samples from their storage locations to the glove box line. Photon dose rates of several hundreds of millirem per hour have been measured outside of their containers. The 59.5 KeV gamma ray from the ^{241}Am and the photons from the decay of ^{239}Np , the daughter of ^{243}Am , are the major sources of radiation exposure.

Thus the cost of this experiment has increased dramatically over the initial estimates. We have received significant support from Argonne's IFR project budget. Many of the required tasks must be accomplished by the experienced technical staff members from Argonne rather than University personnel. However, continued involvement of LSU will be required to assure project continuity; the USDOE grant has served to build a true collaborative relationship between the ANL technical staff and LSU students and faculty.

The need for an additional set of samples to be irradiated in light water test reactor, NRAD, has been questioned. After some study, it has been decided to eliminate this exposures because the spectra in the NRAD reactor is so different than that of EBR-II. However, sets of un irradiated samples will be retained for quality control purposes. They will be retained at ANL-W as an archival set of references. Previous work in measuring neutron energy distributions indicates the importance of accuracy in determining the composition of target nuclides.⁽⁷⁾

A fourth milestone, development of irradiation schemes, was met in the first year, but has been modified throughout the project duration. The 10-day exposure in EBR-II will involve the exposure of ten to twelve targets in three rows of a 37-pin driver subassembly. Because of the

low burnup, the fission product decay heating is small so the subassembly can be removed from the core and transferred to the hot cells at the Hot Fuel Examination Facility (HFEF) with a cooling time of only two days. However, the activity in the 34 fueled pins is such that it will take at least 10 days before the subassembly can be transferred to HFEF for recovery of the irradiated targets for transfer to the ANL-W Analytic Laboratory.

Gamma spectrometry conducted in the ANL-W Analytic Laboratory will be used to determine the neutron spectrum and relative fission rates of the heavy elements in EBR-II. Mass and alpha spectroscopy will not be done on these samples because of the low burnup; the number of nonfission captures will be too low. If the delay between reactor shutdown and gamma spectroscopy is minimized, there is a possibility that we may be able to determine capture rates in ^{238}U by measurement of the ^{239}Np that is formed. When combined with the spectral measurements, this reaction rate can be used to predict breeding ratios in various IFR designs.

Six of the twelve fuel elements in the third row of a driver subassembly will be displaced by the pins containing dosimeters in the extended irradiations. When this driver achieves its target burnup, it will be removed from the core, allowed to cool in the reactor tank, and then removed to the Main Cell of HFEF for disassembly. Two of the target pins will be sent to the ANL-W Analytical Chemistry Laboratory; the other four will be reconstituted in a fresh driver subassembly and reinserted into EBR-II. This procedure will be repeated until all six pins have been irradiated. The Analytical Chemistry Laboratory will perform gamma, alpha, and isotope-dilution mass spectrometry on the irradiated samples to determine changes in composition. Three materials, iron, nickel, and titanium are not included in the extended runs because of the short half-lives of their activation products. The extended exposures require a considerable cooling time before the subassemblies can be removed from the reactor tank. So there is a high probability that statistically significant data would not be available from these targets.

Calculations of neutron spectra around the dosimeter holders irradiations to determine the perturbations and estimates of cross sections and consumption rates, the fifth milestone, have been completed. One-group cross sections for fission, nonfission capture, and (n,2n) reactions

were obtained using a spectrum for metallic fuels in EBR-II provided by ANL-W.⁽⁸⁾ These are compared to those developed in the early 1980s by ORNL.⁽⁷⁾ Except for the (n,2n) cross sections which play a very minor role, the two sets were quite close as shown in Table 4.

As reported last year, these LSU-developed cross sections were used to determine the perturbation of the spectra and flux intensities caused by the substitution of the experimental package into the 1A1 subassembly. The effects were minor; so we can conclude that the measured spectra will be appropriate for a 37-pin fueled subassembly. The comparison of the spectral intensities inside and outside of the experimental capsule is shown in Figure 3.

Additional calculations of heat generation in the experimental capsules are underway at ANL to support the safety documentation. Estimates of fission gas generation and resulting swelling of the vanadium capsules have been completed at LSU using the ORIGEN-S program.⁽⁴⁾ To assure conservative results, it was assumed that all of these gases were released from the targets. Table 5 presents a summary of the results for a 10 mg ²³⁵U target irradiated for 1149 days for three different sets of cross sections. The fission rates and associated gas generation rates are less in the other target actinides except ²³⁹Pu. Since our irradiation times are shorter, this table presents an upper bound on internal gas pressures.

Three refereed articles were published in the fourth year of this grant. One associated with shielding ²⁴¹Am sources appeared in the August 1992 issue of *Health Physics*; a reprint is included as Appendix B of this report. Another paper on characterization of actinide waste was published in the Proceedings of the Waste Management '92 Conference; it is also included in Appendix B. Support from this grant has been acknowledged in two publications of papers given at Waste Management '93; the full text will be published in the conference proceedings that will be issued in June 1993. Another manuscript on the radiological risk associated with spent fuel from EBR-II is being prepared for submission to *Health Physics* for consideration. This is the key parameter that governs the amount of cooling time in the reactor before a subassembly can be transferred to HFEF. The work on nuclear criticality safety in the Fuel Cycle Facility and HFEF accomplished at LSU will be incorporated into another article for

journal publication. An invited paper on actinide burning will be given at the Annual Meeting of the Pacific Division of the American Academy for the Advancement of Science on June 22, 1993 in Missoula, MT.

REVIEW OF FOURTH YEAR MILESTONES

Eight milestones were identified for the fourth year of this grant. These tasks were all scheduled for completion earlier in the project. They are outlined as follows with their status.

- Characterize the unirradiated dosimeter target materials by gamma, alpha, and isotopic dilution mass spectrometric techniques. **Continuing.**
- Complete estimates of all irradiation conditions to support the safety and quality assurance requirements for EBR-II. **Continuing - expected completion by July 1993.**
- Fabricate the LSU/ANL designed dosimeters. **Continuing.**
- Load the dosimeters with known amounts of target materials. **Continuing.**
- Obtain the ^{240}Pu , ^{241}Am , and ^{243}Am target materials. **Completed.**
- Obtain the ORNL fabricated dosimeters for the wire targets. **Completed.**
- Complete short term irradiation of the dosimetry targets in the NRAD reactor. **Cancelled.**
- Fabricate the samples and hardware for irradiation in EBR-II. **Continuing.**

To summarize, the analytic support work is on schedule, but the experimental phase has been delayed by the lack of target materials. A major effort throughout this project has been devoted to obtaining the required target actinides; at last, this task has been successfully completed.

The fourth year of this project has been characterized by increased involvement of Argonne personnel. Most of the Principal Investigator's time has been spent in providing continuity and coordination to the project. This has involved extensive travel and interaction with the variety of disciplines needed to accomplish the tasks. Efforts at the University have centered on a number of supporting projects that are needed to bring the Fuel Cycle Facility on line and support HFEF operations.

DATE

FILMED

12 / 29 / 93

END