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Overview of the FUTURIX-FTA Irradiation Experiment in the Phénix Reactor

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Abstract – *A joint fast reactor irradiation experiment, known as FUTURIX-FTA, was conducted in the Phénix reactor to evaluate the transmutation of minor actinides in metallic and nitride fuels, among others. The experiment was a collaboration between the Department of Energy of the United States of America and the Commissariat à l'Energie Atomique et aux Energies Alternatives of France. Fuels with the same compositions were also irradiated in AFC-1 experiment series in the Advanced Test Reactor using a modified neutron spectrum to create fast reactor thermal profiles in the fuel. Comparisons of results from both sets of experiments will be used to validate fuel performance in the ATR experiments.*

I. INTRODUCTION

Transmutation of minor actinides in fast neutron spectrum reactors is being investigated as a means of reducing the inventory of used nuclear fuel requiring geologic disposal. Multiple international programs are studying transmutation fuels, including programs supported by the Department of Energy (DOE) in the U.S. and the Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA) in France. A joint agreement between DOE and CEA was established in 2004 to conduct an irradiation experiment of metallic and nitride fuels in the Phénix reactor at CEA. Companion experiments were irradiated in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL).

Research and development activities on transmutation fuels for fast reactors have been in progress in the U.S. since the early 2000s. Efforts in the U.S. have been conducted without a fast reactor for irradiation testing. An experiment test train using a cadmium shroud to filter out thermal neutrons in ATR provides a thermal environment which is nearly prototypic of that experienced by fuels in a fast reactor.[1] However, a comparison between irradiation testing using this modified ATR spectrum and a real fast neutron spectrum was desired. Therefore, irradiation experiments using the same fuel compositions were conducted in both ATR and Phénix in order to compare fuel behavior in both spectra. The irradiation experiment in Phénix is named FUTURIX-FTA and the matching irradiation experiments conducted in ATR are part of the AFC-1 series.

This paper presents a summary of the FUTURIX-FTA experiment design, test matrix, and irradiation in the Phénix reactor.

II. EXPERIMENT TEST MATRIX

At the time the AFC and FUTURIX-FTA experiments were designed, the U.S. was investigating both metallic and nitride fuels for transmutation of actinides from light water reactor (LWR) used fuel. In addition, both fertile (uranium bearing) and non-fertile fuels were being developed for actinide transmutation in fast reactor and accelerator systems, respectively.

Previous fast reactor metallic fuel development in the U.S. had focused on binary (U-10Zr) and ternary (U-xPu-10Zr, with x = 20-30 wt%) alloys. The design of transmutation fuels started with this base composition and made adjustments to add minor actinides and increase the alloying agent to stabilize the fuel phase.[2]

The fertile fuel compositions selected for AFC and FUTURIX-FTA irradiation testing contained a substantial amount of plutonium as expected in an equilibrium closed fuel cycle where plutonium is bred in the blanket region of a fast reactor, then incorporated in the driver fuel after reprocessing. Americium and neptunium are added to the fuel as minor actinides that could be recovered from LWR fuel and transmuted (i.e., burned) in a fast reactor and removed from the geologic repository waste path. Zirconium is used as an alloying agent in order to increase the fuel solidus temperature and reduce fuel-cladding chemical interaction (FCCI).

The non-fertile fuel compositions selected for irradiation testing contained high levels of plutonium and americium to demonstrate burning in an accelerator-driven system. These compositions were free from uranium in order to avoid the production of plutonium. As with the fertile fuels, the non-fertile metallic fuels were alloyed with zirconium to increase the fuel solidus temperature and reduce FCCI. The non-fertile nitride fuels were fabricated with ZrN as the inert matrix phase.

Nominal compositions for the four FUTURIX-FTA fuel pins are listed in Table I. Also listed are the companion AFC-1 series experiments irradiated in ATR that had the same nominal fuel compositions.

TABLE I

Nominal Fuel Compositions for FUTURIX-FTA and Companion AFC-1 Experiments

Nominal Fuel Composition*	FUTURIX-FTA Pins	AFC Rodlets
U-29Pu-4Am-2Np-30Zr	DOE-1	AFC-1F: 1, 4 AFC-1H: 1, 4
Pu-12Am-40Zr	DOE-2	AFC-1B: 1, 4 AFC-1D: 1, 4 AFC-1G: 1, 4
$(U_{0.50},Pu_{0.25},Am_{0.15},Np_{0.10})N$	DOE-3	AFC-1Æ: 1, 3, 4
$(Pu_{0.5},Am_{0.5})N$-36ZrN	DOE-4	AFC-1Æ: 6 AFC-1G: 3

* DOE-1 and DOE-2 composition expressed in weight %.
DOE-3 and DOE-4 composition expressed in mole fraction.

III. FABRICATION

Fuels for the FUTURIX-FTA experiment were fabricated in the U.S. The metallic fuels were fabricated at INL and the nitride fuels were fabricated at Los Alamos National Laboratory (LANL). Short experimental fuel pins were assembled and welded at INL and shipped to CEA in 2006 where extensions were welded onto the short pins to make them the same length as standard Phénix fuel pins. A schematic of the fuel pins with extensions is shown in Fig. 1.

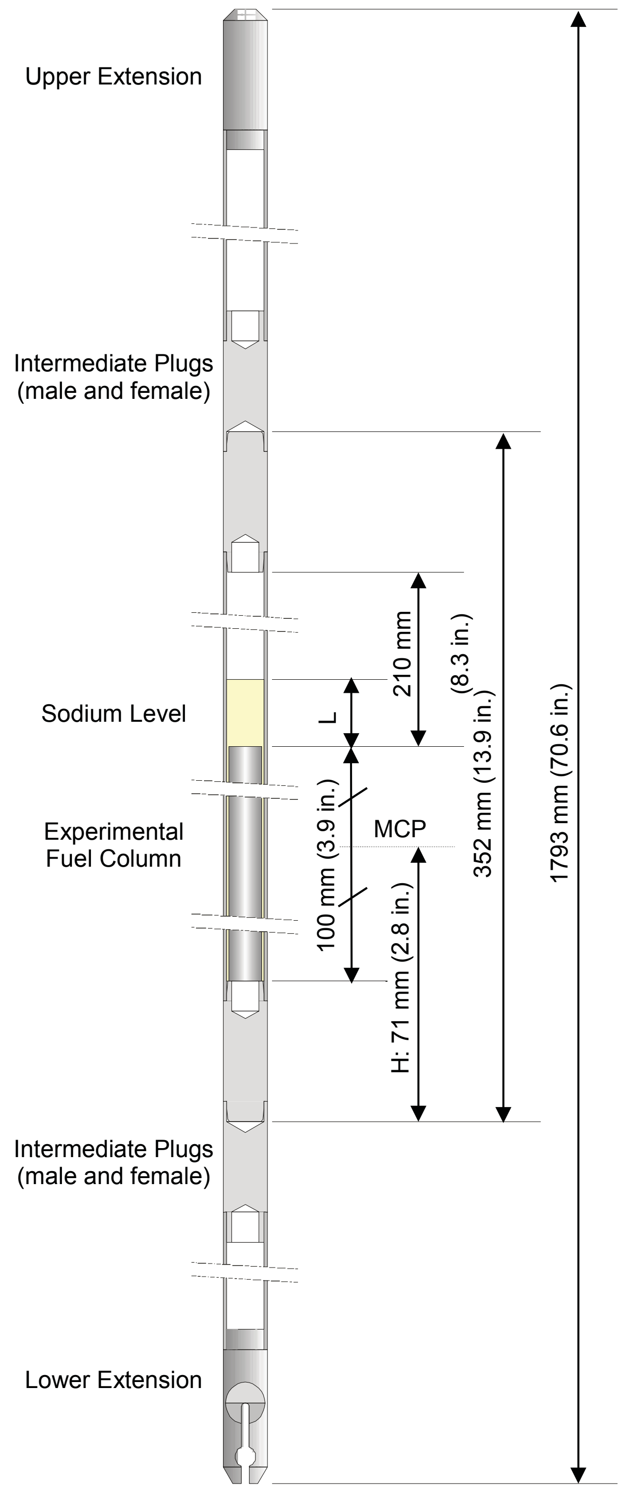

Fig. 1. Schematic of fuel pin with extensions.

The short fuel pins were 352 mm (in.) in length with ~100 mm (in.) fuel column. Sodium was included in the fuel-cladding gap to improve heat transport between the fuel and the cladding and improve the fuel thermal behavior. The fuel pin cladding is AIM1, an austenitic stainless steel provided by CEA as the standard Phénix cladding. Fuel pin parameters are listed in Table II.

TABLE II

FUTURIX-FTA Pin Parameters

Parameter	Value
Cladding	AIM1
Fuel Pin Inner Diameter	5.65 mm (0.222 in.)
Fuel Pin Outer Diameter	6.55 mm (0.258 in.)
Fuel Column Length	100 mm (3.93 in.)
Short Fuel Pin Length	352 mm (13.8 in.)
Full Fuel Pin Length	1793 mm (70.6 in.)

The FUTURIX-FTA experimental fuel pins were assembled into two Phénix fuel capsules and placed into adapted assemblies. Each Phénix fuel capsule contained 19 fuel pins. Capsule KCI 6908 housed two metallic fuel experimental pins, DOE-1 and DOE-2, and capsule KCI 6909 had two nitride fuel experimental pins, DOE-3 and DOE-4. The remaining 17 fuel pins in each capsule were Phénix standard fuel pins.

III.A. Metallic Fuels

Two metallic fuel compositions were fabricated at INL in 2006. The metallic fuel slugs were fabricated using an arc-casting method where the individual feedstock materials are melted together and homogenized into a “button.” The button was melted, an uncoated quartz tube mold was dipped into the liquid, and the liquid was drawn up into the model via suction using a syringe.[3] The final metallic fuel slugs are shown in Fig. 2 and Fig. 3

Samples from representative casts of the fuel alloy compositions were characterized for phase formation by X-ray diffraction (XRD), microstructure by scanning electron microscopy (SEM), heat capacities and thermal phase transitions by Differential Scanning Calorimetry (DSC) and Differential Thermal Analysis (DTA) measurements, thermal expansion from Thermomechanical Analysis (TMA) measurements, thermal diffusivity by the laser flash method, thermal conductivity, and fuel-cladding-chemical-interaction (FCCI) with the AIM1 stainless steel cladding material.

Fig. 2. Fuel column of the U-29Pu-4Am-2Np-30Zr composition prior to DOE-1 fuel pin loading.

Fig. 3. Fuel column of the Pu-12Am-40Zr composition prior to DOE-2 fuel pin loading.

III.B. Nitride Fuels

Two nitride fuel compositions were fabricated at LANL in 2006. The nitride fuel pellets were fabricated using carbothermic reduction/nitridization (CTR/N) to convert feedstock material and solutions from an oxide to a nitride. The nitride solution feedstock was then milled, pressed into pellets and sintered.[4] Selections of the nitride fuel pellets and property measurement samples are shown in Fig. 4 and Fig. 5.

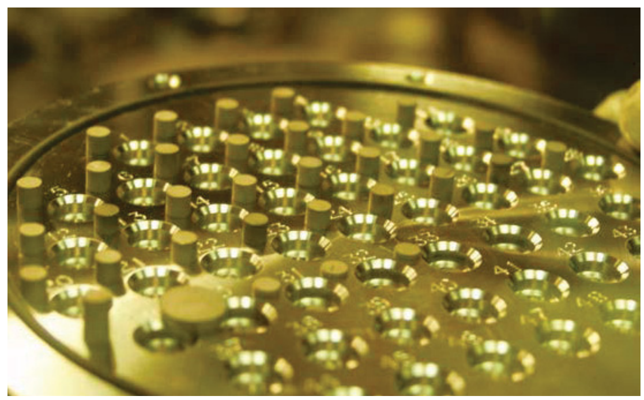

Fig. 4. Low-fertile nitride pellets and property measurement samples for the DOE-3 fuel pin.

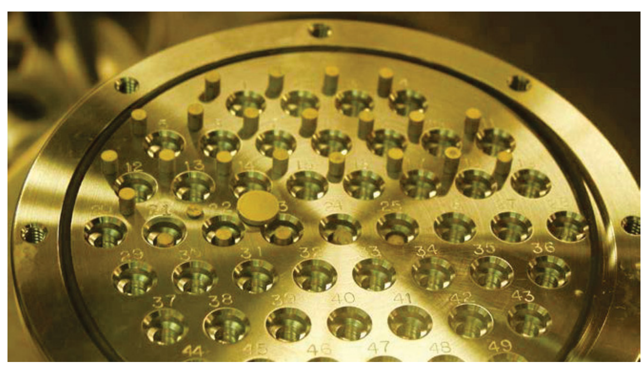

Fig. 5. Non-fertile nitride pellets and property measurement samples for the DOE-4 fuel pin.

In addition to the fuel pellets, several fuel samples were fabricated for material property measurements. Characterization of fuel pellets and/or property samples included visual inspection, dimensional inspection, density, thermal diffusivity, thermal expansion, calorimetry, optical microscopy, phase formation by X-ray diffraction, and microstructure by scanning electron microscopy.

Fabrication development for nitride fuel pellets containing minor actinides encountered significant challenges. The americia feedstock for the FUTURIX-FTA fuels appeared to be different than the feedstock previously used for the AFC-1 fuel fabrication, so additional heat treatments were applied to the feedstock to remove moisture and improve flowability. Smaller batch sizes were used during the fabrication process to reduce handling and storage time for the feedstock.

Limited fabrication development time prevented fuel pellet fabrication process optimization. Lamination and endcapping were observed in the non-fertile fuel pellets. Process improvements were identified and tested and the final pellets were fabricated using the recommended process parameters. Details of the fertile and non-fertile nitride fuel fabrication are described in a fuel fabrication report.[4]

All pellets selected for the irradiation experiment met fuel fabrication specification criteria for density and physical defects; however, given that the fuel densities were on the lower end of the specification range, pellets were prone to cracking during fabrication, and the fact that irradiation performance data for actinide-bearing nitride fuels was limited, the irradiation duration was constrained to prevent fuel swelling and hard contact with the cladding.

IV. IRRADIATION

The metallic fuel pins met all of their fabrication and inspection acceptance criteria and were accepted for their planned irradiation. The metallic fuel pins, DOE-1 and DOE-2, began irradiation in May 2007 and completed irradiation in May 2009 for a total of 235 EFPD. No sign of fuel pin failure (i.e., loss of tightness) was detected during irradiation.

Fabrication of the minor actinide-containing nitride fuels produced pellets that were less robust than desired and prone to cracking. Based on concerns about the integrity of the fuel, the irradiation duration was limited to prevent fuel swelling.

The nitride fuel pins, DOE-3 and DOE-4, began irradiation in July 2008 and completed irradiation in November 2008 for a total of 56 EFPD. No sign of fuel pin failure (i.e., loss of tightness) was detected during irradiation.

A summary of burnup for the four FUTURIX-FTA pins is shown in Table III.

TABLE III

FUTURIX-FTA Fuel Pin Burnup

Pin	Nominal Fuel Composition*	Burnup (at% HM)
DOE-1	U-29Pu-4Am-2Np-30Zr	9.1
DOE-2	Pu-12Am-40Zr	15.5
DOE-3	$(U_{0.50},Pu_{0.25},Am_{0.15},Np_{0.10})N$	1.6
DOE-4	$(Pu_{0.5},Am_{0.5})N$-36ZrN	4.1

* DOE-1 and DOE-2 composition expressed in weight %. DOE-3 and DOE-4 composition expressed in mole fraction.

Companion fuel pins with the same nominal fuel compositions were irradiated in ATR between 2003 and 2008 as part of the AFC-1 experiment series. The design for AFC experiments in ATR uses cadmium-shrouded experiment positions to remove >99% of the thermal flux incident on the test fuels.[1] The remaining flux is primarily epi-thermal and results in a fuel experiment that is free from gross flux depression and produces a radial temperature profile inside the fuel that is essentially prototypic of that in a fast reactor.

A summary of the burnup of the FUTURIX-FTA and AFC-1 experiment series pins is shown in Table IV.

TABLE IV

Burnup for FUTURIX-FTA and Companion AFC-1 Fuel Pins

Nominal Fuel Composition*	Pin†	Burnup (at% HM)
U-29Pu-4Am-2Np-30Zr	DOE-1	9.1
	AFC-1F-1	3.1
	AFC-1F-4	4.4
	AFC-1H-1	18.0
	AFC-1H-4	26.7
Pu-12Am-40Zr	DOE-2	15.5
	AFC-1B-1	3.1
	AFC-1B-4	4.3
	AFC-1D-1	15.7
	AFC-1D-4	22.6
	AFC-1G-1	14.1
	AFC-1G-4	20.3
$(U_{0.50},Pu_{0.25},Am_{0.15},Np_{0.10})N$	DOE-3	1.6
	AFC-1Æ-1	4.0
	AFC-1Æ-3	5.9
	AFC-1Æ-4	6.4
$(Pu_{0.5},Am_{0.5})N$-36ZrN	DOE-4	4.1
	AFC-1Æ-6	2.9

* DOE-1 and DOE-2 composition expressed in weight %. DOE-3 and DOE-4 composition expressed in mole fraction.
† DOE pins were irradiated in Phénix and AFC pins were irradiated in ATR

V. POSTIRRADIATION EXAMINATION

Following the completion of irradiation and the shutdown of the Phénix reactor, the FUTURIX-FTA experiments were stored in the transfer drum. In March 2014 the assemblies containing the FUTURIX-FTA pins were transferred into the Phénix hot cell, washed, and disassembled. The four FUTURIX-FTA pins were visually verified and inserted into the ET-004 basket prior to loading in the TN-106 cask for shipment to the U.S.

In July 2014, the TN-106 cask with the FUTURIX-FTA pins sailed from France to the U.S. and was then transported via truck to INL and received at the Hot Fuel Examination Facility hot cell. The TN-106 cask, after arrival at INL, is shown in Fig. 5.

Fig. 6. TN-106 cask waiting to be unloaded at INL.

Postirradiation examination (PIE) of the FUTURIX-FTA fuels will be conducted to evaluate fuel behavior at both the bulk and microstructural scales.[5] Baseline PIE includes:

- visual exam (digital photography)
- dimensional measurements
- gamma spectroscopy (axial and radial tomography)
- neutron radiography
- fission gas release analysis
- optical metallography and microhardness
- analytical chemistry for burnup analysis

Visual examinations began in July 2015 and baseline PIE will continue throughout the remainder of 2015 and into 2016. Additional PIE, particularly exams to evaluate the microstructure and chemical composition of the fuel, will be conducted as new shielded equipment (e.g., scanning electron microscope, electron probe microanalysis) becomes available for irradiated fuels.

Baseline PIE has previously been completed on the AFC-1 series experiments. Among a wide variety of compositions, AFC-1 fuel behavior has generally been observed to be excellent and is consistent with historic results for metallic fuels without minor actinides and with lower plutonium and zirconium content, as studied in experiments in the Experimental Breeder Reactor II, a sodium-cooled fast reactor.[6]

Results from PIE of AFC-1 experiments will be compared to PIE from FUTURIX-FTA to identify and evaluate any significant differences in fuel behavior caused by using a modified (filtered) neutron spectrum in ATR vs a real fast neutron spectrum in Phénix.

VI. CONCLUSIONS

The FUTURIX-FTA joint irradiation experiment between DOE and CEA was a successful international collaboration. Results from irradiation testing in a prototypic fast reactor environment will provide vital data to the transmutation fuels development program in the U.S.

Results of fuel behavior from FUTURIX-FTA, irradiated in a typical sodium-cooled fast reactor, will be compared with results from the AFC-1 experiment series, irradiated in ATR, to identify and evaluate any differences in fuel behavior due to the modified ATR spectrum. Comparison of these results will be an important part of the validation process to show that the modified neutron spectrum in ATR can be used to investigate fast reactor fuel behavior.

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NOMENCLATURE

ATR	Advanced Test Reactor
CEA	Commissariat à l'Energie Atomique et aux Energies Alternatives
DOE	Department of Energy
EBR-II	Experimental Breeder Reactor II
EFPD	Effective Full Power Day
FCCI	Fuel-cladding Chemical Interaction
HM	Heavy Metal
IFR	Integral Fast Reactor
INL	Idaho National Laboratory

LANL	Los Alamos National Laboratory
LWR	Light Water Reactor
MA	Minor Actinide
PIE	Postirradiation Examination

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