INL/CON-07-12159
PREPRINT

Irradiation of Metallic and Oxide Fuels for Actinide Transmutation in the ATR

Global 2007

Heather J. MacLean
Steven L. Hayes

September 2007

The INL is a
U.S. Department of Energy
National Laboratory
operated by
Battelle Energy Alliance

Irradiation of Metallic and Oxide Fuels for Actinide Transmutation in the ATR

Heather J. MacLean, Steven L. Hayes

Idaho National Laboratory: P.O. Box 1625, Idaho Falls, ID, 83415, heather.maclean@inl.gov

Metallic fuels containing minor actinides and rare earth additions have been fabricated and are prepared for irradiation in the ATR, scheduled to begin during the summer of 2007. Oxide fuels containing minor actinides are being fabricated and will be ready for irradiation in ATR, scheduled to begin during the summer of 2008. Fabrication and irradiation of these fuels will provide detailed studies of actinide transmutation in support of the Global Nuclear Energy Partnership. These fuel irradiations include new fuel compositions that have never before been tested. Results from these tests will provide fundamental data on fuel irradiation performance and will advance the state of knowledge for transmutation fuels.

I. INTRODUCTION

Transmutation of long-lived transuranic (TRU) isotopes contained in spent nuclear fuel is one of the key components of the Global Nuclear Energy Partnership (GNEP). Under the GNEP plan, uranium and transuranic elements (plutonium, neptunium, americium, and curium) separated from spent nuclear fuel could be fabricated into fuels for fast reactors. Fast reactors would consume or destroy the transuranic elements, dramatically decreasing the volume of material requiring disposal as well as the long-term radiotoxicity and heat load of the high-level waste sent to a geologic repository.[1]

Successful implementation of the GNEP plan requires development and demonstration of transmutation fuel forms for fast reactors. The two leading fuel forms under current development and testing are metallic and oxide fuels. Both metallic and oxide fuels have previously been used as driver fuels in fast test reactors (e.g., EBR-II and FFTF in the U.S.). The U.S. plans to make a down-selection between metallic and oxide fuels in the FY 2012 timeframe to select the fuel form for the first GNEP fast reactor core.

I.A Transmutation Fuel

Little data exists on the irradiation performance of fuels containing significant minor actinide constituents. The Advanced Fuel Cycle Initiative (AFCI, now the technology research and development component of GNEP) has been and is currently conducting irradiation tests of transuranic-bearing fuels for transmutation of minor actinides. Results from this irradiation program will provide performance data on fuel compositions that have never before been fabricated and tested. These data will improve the state of knowledge of transmutation fuels and will provide information to support fuel modeling efforts.

The previous test series, Advanced Fuel Cycle 1 (AFC-1), contained metallic and nitride fuels with varying amounts of transuranic elements, some in non-fertile (uranium-free) compositions for potential accelerator-driven systems and others in low-fertile compositions for reactor-based transmutation. The AFC-2 test series consists of metallic and oxide fuels with plutonium and minor actinides for fast reactor-based transmutation. The metallic fuel compositions are an evolution of the previously tested AFC-1 metallic fuels; the AFC-2 oxide test is the first oxide test in the AFC test series, but the compositions under investigation are an evolution of historic fast reactor oxide fuel experience.

The TRU composition in the AFC-2 test series is based on the expected TRU content by group extraction from recycled spent fuel from light water reactors with an average burnup of 50 MWd/MT and cooled for 40 years. The relative ratio of TRU elements is presented in TABLE I. The Am and Np concentrations selected for the AFC-2 test series represent an upper bound on the expected TRU compositions (the ratio of Am and Np to Pu has been increased slightly over the expected ratio in order to provide a bounding composition).

TABLE I. TRU Elemental Ratio

Element	Ratio	20% Pu	30% Pu
Pu	1	20	30
Am	0.138	3	5
Np	0.086	2	3

I.B Irradiation Test Assembly

The AFC-2 test series fuels will be irradiated in the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL) using the same configuration as the AFC-1 test series. The ATR is a thermal reactor with a maximum thermal power of 250 MW_{th}. The AFC-1 and AFC-2 test capsules are irradiated in the East Flux Trap, one of nine high flux test locations. The East Flux Trap has a maximum thermal flux of 4.4×10^{14} n/cm^2-s and a maximum fast flux of 9.7×10^{13} n/cm^2-s (E>1 MeV).

The irradiation test assembly consists of an experiment basket and a capsule assembly. The experiment basket for the AFC-2 test series is an aluminum-sheathed cadmium tube designed to act as a thermal neutron filter. The capsule assembly contains six rodlet assemblies stacked vertically. The fuel specimens are encapsulated in the rodlet assemblies, which are designed as miniature fast reactor fuel pins with a prototypic diameter but reduced length. The metallic rodlet assemblies utilize a sodium bond, and the oxide rodlet assemblies utilize a helium bond. Fuel rodlet design data are shown in TABLE II.

TABLE II. Fuel Rodlet Design Data

Design Parameter	AFC-2A AFC-2B	AFC-2C AFC-2D
Cladding Material	HT9	HT9
Cladding O.D.	5.84 mm	5.84 mm
Cladding I.D.	4.93 mm	4.93 mm
Bond Material	Sodium	Helium
Fuel Type	Metallic	Oxide
Fuel Smear Density	75%	81%
Fuel Porosity	0%	15%
Fuel O.D.	4.27 mm	4.88 mm
Fuel Height	38.1 mm	50.8 mm

The experimental capsules containing the AFC-2 fuel rodlets are inserted into ATR inside cadmium baskets that filter the thermal flux, producing a somewhat hardened neutron spectrum in the test region. Although the irradiation environment does not include a prototypic fast-neutron spectrum and does not produce prototypic cladding damage, ATR is the best domestic irradiation facility currently available in the U.S. ATR provides a sufficiently large test volume for conducting the AFC irradiations, AFC irradiation tests fit easily within the ATR safety envelope, and transportation between ATR and the post-irradiation examination (PIE) facilities at INL is straightforward.

Although the AFC-2 tests will not be conducted in a prototypic fast reactor environment, the test results will provide meaningful data on TRU-bearing fuel behavior and help develop fundamental understanding of the effect of TRU constituents on fuel performance. Due to the lack of a prototypic neutron spectrum, however, cladding damage is accumulated at a much lower rate than in a fast neutron environment. Fast reactor testing will be required to supplement ATR irradiations and confirm these results.

Fast reactor experiments are underway in the Phénix reactor in France. These tests, containing fuel compositions from the AFC-1 test series, will provide confirmation of the results obtained from ATR irradiations. Additional fast reactor irradiation experiments are in the planning stage.

II. METALLIC FUEL

II.A Experiment Fabrication

Metallic fuel development was restarted in the U.S. in 2002. The AFC-1 series of irradiation tests included non- and low-fertile metallic fuels with varying amounts of plutonium, neptunium, americium, and zirconium. AFC-1 metallic fuel irradiations have achieved burnups above 25% (of fissile content) for fertile alloys and nearly 40% for non-fertile alloys. The AFC-2 series of irradiation tests is focused on prototypic fuel compositions for a fast-reactor transmutation fuel containing recycled transuranics from light water reactor spent nuclear fuel.

The compositions for the AFC-2 metal irradiations, shown in TABLE III, consist of two base compositions containing minor actinides and variations with rare earth elements added.[2] The amount of actinides included in each composition is based on the elemental ratios of TRU expected from group extraction (of Pu, Am, Np) from LWR spent fuel cooled for approximately 40 years. An alloy of rare earth (RE) elements is added to the base compositions to simulate fission product carry-over resulting from the pyroprocessing of fast-reactor fuel. The RE alloy was specified based on chemical analyses of TRU products recently extracted from EBR-II spent fuel using a liquid cadmium cathode. The ^{235}U enrichments were selected to achieve the linear heat generation rate (LHGR) design objective of 350 W/cm.

Two identical capsules, AFC-2A and AFC-2B, will contain rodlets with compositions 1-6, as shown in TABLE III.

TABLE III. AFC-2 Metallic Fuel Alloys

Rodlet	Composition †	^{235}U Enrichment
1	U-20Pu-3Am-2Np-15Zr	93%
2	U-20Pu-3Am-2Np-1.0RE*-15Zr	55%
3	U-20Pu-3Am-2Np-1.5RE*-15Zr	45%
4	U-30Pu-5Am-3Np-1.5RE*-20Zr	55%
5	U-30Pu-5Am-3Np-1.0RE*-20Zr	65%
6	U-30Pu-5Am-3Np-20Zr	93%

† Alloy composition expressed in weight percent

* RE designates rare earth alloy (6% La, 16% Pr, 25% Ce, 53% Nd)

Fabrication of the metallic fuel alloys has been completed at Idaho National Laboratory. Fabrication was performed by arc-casting, with a portion of the work completed in a new glovebox line (shown in Fig. 1) recently started in the Fuel Manufacturing Facility. A representative metallic fuel slug from the AFC-2 campaign is shown in Fig. 2.

Fig. 1. AFCI glovebox in the Fuel Manufacturing Facility at INL.

Fuel characterization is currently in progress to measure alloy chemistry and isotopic composition, density, and thermal diffusivity, along with microstructural characterization. The thermal diffusivity measurements will be used to assess the thermal conductivity of the different fuel alloys. These measurements will be the first performed on the fuel compositions described above. The measurements will be used to compare the properties and characteristics of transuranic-bearing metallic fuels with the historic database to develop a better understanding of the effect of transuranic constituents, particularly the minor actinides, on fuel properties.

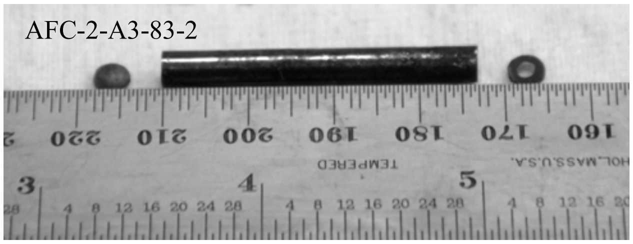

Fig. 2. AFC-2 metallic fuel slug

II.B Experiment Irradiation

The AFC-2A and AFC-2B capsules are scheduled to be inserted into ATR in August 2007 (ATR cycle 140A). Neutronic analyses have been performed to estimate the operating conditions for each of the six fuel rodlets in each experimental irradiation capsule. TABLE IV lists the predicted linear power and fission density for each of the rodlets in capsules AFC-2A and AFC-2B.[3]

TABLE IV. AFC-2 Metallic Fuel Predicted Linear Heat Generation Rate (LHGR) and Fission Density for ATR Cycle 140A

Rodlet	Capsule 2A		Capsule 2B	
	LHGR (W/cm)	Fission Density (fissions/cm^3)	LHGR (W/cm)	Fission Density (fissions/cm^3)
1	308	1.18 E+20	320	1.22 E+20
2	315	1.20 E+20	334	1.27 E+20
3	334	1.26 E+20	**343**	1.30 E+20
4	329	1.24 E+20	338	1.27 E+20
5	317	1.20 E+20	334	1.26 E+20
6	307	1.16 E+20	306	1.16 E+20

The target burnup for capsule AFC-2A is ≥10 at.% and for capsule AFC-2B is ≥25 at.% (burnup defined to be percent depletion of initial ^{235}U and ^{239}Pu). The peak cladding temperature during normal operation is 550°C.

III. OXIDE FUEL

The AFC-2 oxide fuel tests are focused on restarting fast-reactor oxide fuel development in the U.S. The initial oxide fuel irradiations will consist of two identical capsules (AFC-2C and AFC-2D), each with two rodlets of three compositions. The baseline oxide composition was selected to have 20% plutonium.

The compositions for the AFC-2 oxide fuel irradiations, shown in TABLE V, consist of a baseline fast-reactor MOX composition [$(U_{0.8},Pu_{0.2})O_{1.98}$] and two compositions with minor actinide additions (americium and neptunium). As with the metallic alloy fuels, the ratio of the transuranic elements in the oxide fuel is based on the expected ratio of TRU in recycled legacy LWR spent nuclear fuel. Of the two actinide-bearing compositions, one will have a baseline oxygen/metal (O/M) ratio of 1.98 and the other will have a lower O/M ratio.

TABLE V. AFC-2 Oxide Fuel Compositions

Rodlet	Composition †
1	$(U_{0.75},Pu_{0.2},Am_{0.03},Np_{0.02})O_{1.95}$
2	$(U_{0.8},Pu_{0.2})O_{1.98}$
3	$(U_{0.75},Pu_{0.2},Am_{0.03},Np_{0.02})O_{1.98}$
4	$(U_{0.75},Pu_{0.2},Am_{0.03},Np_{0.02})O_{1.95}$
5	$(U_{0.8},Pu_{0.2})O_{1.98}$
6	$(U_{0.75},Pu_{0.2},Am_{0.03},Np_{0.02})O_{1.98}$

† Fuel composition expressed in mole percent

The ^{235}U enrichment for the oxide fuel compositions will be selected such that the peak rodlet will operate near 350 W/cm LHGR. Due to fabrication process constraints, it is expected that the oxide fuel rodlets will all have the same enrichment. The upper and lower rodlets, therefore, will operate at linear powers significantly less than 350 W/cm.

Capsule AFC-2C has a target burnup of ≥10 at.%, and capsule AFC-2D has a target burnup of ≥25 at.% (burnup defined to be percent depletion of initial ^{235}U and ^{239}Pu). The oxide fuel is currently being fabricated at Los Alamos National Laboratory.

More details regarding the final design and irradiation conditions for the oxide fuel irradiation are expected as the experiment progresses.

IV. POST-IRRADIATION EXAMINATION

Upon discharge from ATR, each experiment will remain in the ATR canal for a minimum cooling period of 60 days. After cooling, experiments are shipped from ATR to the Hot Fuels Examination Facility (HFEF) at the Materials and Fuels Complex (MFC) at INL for post-irradiation examinations. PIE will include visual examination, dimensional inspection, neutron radiography, gamma scanning, gas plenum assay and analysis, optical and electron microscopy, and burnup/actinide transmutation determinations. Results of PIE will be used to assess the irradiation performance of metallic and oxide fuel forms for actinide transmutation in advanced fuel cycles. Fuel irradiation performance data generated from PIE will include irradiation growth and swelling, fission gas and helium release, fission product and fuel constituent redistribution, fuel phase equilibrium, and fuel-cladding chemical interaction.

V. SUMMARY

Metallic fuels containing minor actinides and rare earth additions have been fabricated and are prepared for irradiation in the ATR, scheduled to begin during the summer of 2007. Oxide fuels containing minor actinides are being fabricated and will be ready for irradiation in ATR, scheduled to begin during the summer of 2008.

TABLE VI. AFC-2 Test Series Anticipated Schedule

Capsule	Insertion	Discharge	Target Burnup
AFC-2A	August 2007	June 2008	≥10 at.%
AFC-2B	August 2007	May 2009	≥25 at.%
AFC-2C	June 2008	May 2009	≥10 at.%
AFC-2D	June 2008	April 2010	≥25 at.%

PIE of the low-burnup metallic fuel rodlets is anticipated to begin before the end of 2008; PIE of the low burnup oxide fuel rodlets is anticipated to begin in 2010.

The AFC-2A and AFC-2B experiments are a continuing evolution of the AFC series irradiation tests to study the fuel performance of TRU-bearing metallic fuel forms for transmutation in fast neutron spectrum systems. The AFC-2C and AFC-2D experiments are the first TRU-bearing oxide fuel forms for transmutation to be irradiated in the U.S. AFCI/GNEP program. Both the metallic and oxide fuel irradiations include new fuel compositions that have never before been tested. PIE from these tests will provide fundamental data on fuel irradiation performance and will advance the state of knowledge for transmutation fuels.

ACKNOWLEDGMENTS

The authors acknowledge the assistance of T. Hyde of INL and S. Voit and K. McClellan of LANL in fuel fabrication and G. Chang, M. Lillo, and D. Utterbeck of INL in experiment analyses and coordination. Work supported by the U.S. Department of Energy under DOE Idaho Operations Office Contract DE-AC07-05ID14517.

REFERENCES

1. U.S. DEPARTMENT OF ENERGY, "Global Nuclear Energy Partnership Strategic Plan," GNEP-167312, Rev. 0 (January 2007).
2. S. L. HAYES, "Irradiation of Metallic Fuels with Rare Earth Additions for Actinide Transmutation in the Advanced Test Reactor," INL/EXP-06-11707 Rev. 1 (December 2006).
3. G. S. CHANG and M. A. LILLO, “Neutronics Analysis LHGR Projections for AFC-2A, -2B, -1G, and -1H Experiments Planned for Irradiation in the ATR EFT,” INL EDF-7399, April 2007.