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H. J. M. Chichester, W. J. Carmack

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Development of Metallic Fuels for Actinide Transmutation

S. L. Hayes, R. S. Fielding, M. T. Benson, H. J. M. Chichester, and W. J. Carmack
Idaho National Laboratory
P. O. Box 1625, Idaho Falls, ID 83415-3835 USA
Tel: +001 208-526-7255, Fax: +001 208-526-2930, Email: steven.hayes@inl.gov

Abstract – *Research and development activities on metallic fuels are currently focused on their potential use for actinide transmutation in future sodium fast reactors. This application necessitates a near zero-loss fabrication process and includes a strong motive to demonstrate a multifold increase in burnup potential. Development and testing of a new casting method to more efficiently utilize the melted fuel charge and suppress volatilization of fuel constituents is presented. Irradiation testing of a wide variety of metallic transmutation fuel compositions is reviewed, including tests conducted in the Advanced Test Reactor and the Phénix Fast Reactor. Finally, an overview of activities related to metallic fuel alloy optimization and performance enhancement is provided.*

I. INTRODUCTION

Metallic fuels have a history of use that spans the entire nuclear age, especially in association with the development of the LMFBR. Initially, metallic fuels were employed due to their ease of fabrication and high heavy metal density (which led to the most favorable breeding efficiencies in fast reactors). During subsequent decades of use, metallic fuel technology has matured significantly, leading to the realization of major benefits in the areas of fuel reliability and burnup, proliferation-resistant recycling, remote fabrication, and passive reactor safety.[1] Such characteristics continue to make metallic fuels a highly attractive technology, especially for use with future sodium fast reactors.

Current research and development activities on metallic fuels are focused on their potential use for actinide transmutation in advanced SFRs. In such a mission, metallic fuels would be employed as part of a closed fuel cycle into which plutonium and minor actinides (Am, Np) recovered from LWR spent fuel would be incorporated. All actinides in this metallic fuel would be fissioned or transmuted in a fast reactor to eliminate the extremely long-lived, heat-producing actinides in order to reduce the volume, heat load, and radio-toxicity burden on a future geological repository.[2] Thus, the need exists to demonstrate reliable performance of metallic fuels with Pu and minor actinide constituents to very high burnup. As part of this application, there is also a need for a near zero-loss fabrication process, since losses during fuel fabrication are a potential source of long-lived actinides destined for geologic disposal.

This paper presents the current state and future direction of the development of metallic fuels for actinide transmutation in the US. A brief historical background is also provided to indicate what the reference metallic fuel technology was at the close of the 20th century, from which the US began a renewed and redirected fuel cycle research and development program in the early 2000's.

II. HISTORICAL BACKGROUND

In the 1980s and 1990s the composition of metallic fuels under serious development evolved from binary to ternary alloys (i.e., U-xPu-10Zr, with x = 20-30 wt%) in support of the Integral Fast Reactor program. The IFR concept sought to develop a sodium fast reactor that was passively safe and that made use of a fuel that was easily reprocessed and remotely refabricated, but in a way that was inherently proliferation-resistant (i.e., one in which Pu was never isolated from other recovered actinides, and even some residual fission products). Metallic fuels fit well with such a concept owing to their high thermal conductivity and thermal expansion which contributed to reactor safety, compatibility with an electrochemical reprocessing scheme, and their demonstrated fabrication at engineering scale in a remote hot cell environment.

A substantial plutonium content was incorporated into the metallic fuel alloy to approximate an equilibrium, closed fuel cycle in which plutonium was bred in the blanket region of a fast reactor and added back into the driver fuel at the reprocessing stage. In the latter phase of the IFR program, it became important to demonstrate the concept of transmutation (i.e., 'burning') of plutonium and minor actinide isotopes (especially Am, Np, and Cm) that could be recovered from LWR spent fuel in order to obtain benefits for a future geologic repository. Zirconium was selected as the alloying element because it offered benefit in reducing fuel-cladding chemical interaction and increasing fuel solidus temperature.

Using the design innovations developed for the metallic fuel pin in EBR-II over 20 years (especially low smear density to accommodate swelling and a large plenum to accommodate high fission gas release), U-Pu-Zr metallic fuel pins were reliably irradiated to 20 at.-% burnup without cladding breach. When the IFR program ended and EBR-II operations were terminated in 1994, EBR-II was on the verge of a total core conversion from U-10Zr to U-20Pu-10Zr metallic fuel. Furthermore, the first experiment was underway to study the effects of adding americium and neptunium to the metallic fuel alloy. A low-burnup experiment was completed in which U-20Pu-2.1A-1.3Np-10Zr was irradiated to 7.6 at.-% burnup.[3]

In the renewed Fuel Cycle Research & Development program initiated in the early 2000's, research and development activities on metallic fuels are focused on their potential use for actinide transmutation in future sodium fast reactors. Early factors that led to metallic fuels being identified as a strong candidate for such a mission included: 1) traditional ease of fabrication, especially considering their prior fabrication at engineering scale in a remote, hot cell environment[4]; 2) compatibility with the proliferation-resistant electrochemical recycle scheme; 3) very high burnup potential; and 4) passive reactor safety features that derive from physical attributes inherent to metallic fuels.[5]

III. FABRICATION DEVELOPMENT

The incorporation of Am and Np into traditional U-Pu-Zr metallic fuel alloys was attempted during the IFR Program in the early 1990's. However, the conventional counter gravity injection casting method performed under vacuum, previously used to fabricate these metallic fuel alloys for EBR-II, was not optimized for mitigating loss of the volatile Am constituent in the casting charge; as a result, approximately 40% of the Am casting charge failed to be incorporated into the as-cast fuel alloys.[6] In addition, the counter gravity method was inherently inefficient, as a significant fraction (typically 30-40%) of the melted charge remained as a heel in the crucible after casting. Limited recycle of the heel into subsequent casting batches was possible. However, interaction of the molten fuel alloy with crucible wash materials (typically a Y_2O_3 slurry) and quartz molds increasingly introduced impurities into the fuel alloy, eventually making continued recycle undesirable; discarded heels became a significant actinide waste stream. Furthermore, the use of one-time, disposable quartz molds gave rise to yet another actinide waste stream.[7] There existed a need for an advanced casting system to address these issues.

Fabrication development efforts of the recent past have pursued an optimized bottom-pour (gravity) casting method to increase utilization of the melted charge to near 100%, a differential pressure casting approach, performed under an argon overpressure, to mitigate Am volatilization during fabrication, and the use of permanent molds. A small, experimental casting furnace was designed, fabricated, and installed in a radiological fume hood in order to evaluate the proposed casting changes relative to melt utilization, loss of volatile constituents, and mold performance. It was operated for several years for casting experiments on U-10Zr and using Mn as a volatile constituent with properties reasonably similar to Am. These experiments indicated that excellent improvement could be achieved in every respect using this casting approach.[8]

The next step was to design and build a casting system that could be installed into a glovebox in order to demonstrate these improvements using actual U-Pu-Am-Np-Zr fuel alloys. The new furnace, designated the Glovebox Advanced Casting System, became operational near the end of 2013. A solid model of the GACS and photograph of its glovebox installation are shown in Fig. 1.

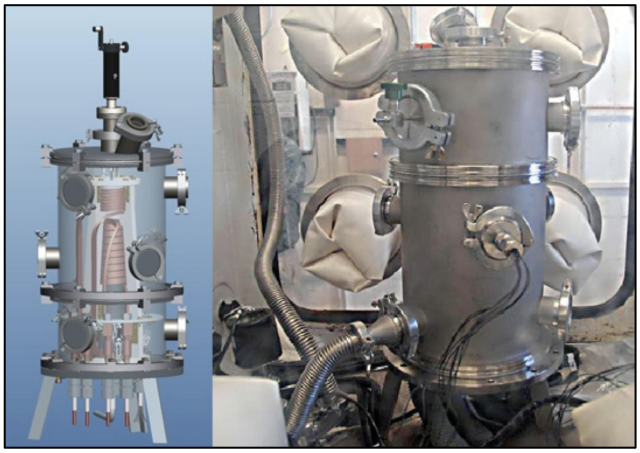


Fig. 1. Schematic and photo of the installed GACS.

A series of three experiments have been performed using the GACS. The first set tested the operability and feasibility of using the furnace for minor actinide fuel fabrication by simultaneously casting three U-TRU-Zr pins of 4.2mm in diameter. The next two sets of experiments focused on retention of Am in the fuel melt during a prototypic heating cycle.

In the experiment which attempted to simultaneously cast three U-TRU-Zr fuel pins, the crucible was charged with U, a Pu-Am-Zr alloy, and Zr feed materials and heated to a casting temperature of 1450°C. The casting produced two pins of approximately 100 mm in length and a third pin of approximately 75 mm in length. The cast fuel pins appeared to be sound and free of substantial surface flaws. A photograph of the three as-cast fuel pins is shown in Fig. 2.

In the set of experiments investigating Am volatility, the three previously cast pins were broken up, loaded in the crucible, and heated to 1450°C for 15 minutes under a constant purge of approximately 0.2 m^3/hr of argon, then allowed to cool. The resulting ingot was sampled, and the remainder of the ingot was re-melted according to the same procedure. This second ingot was again sampled. Chemical analyses of these two ingots are shown in Table I, with a reported uncertainty in the analyses of 5% (2σ). As seen in the table, the Am content appears to be fairly consistent between the two ingots and the initial charge.

Fig. 2. U-TRU-Zr pins cast in the GACS (calipers set at 25.4 mm).

TABLE I

Chemical Analyses from Volatility Experiments

Material	Initial Charge* (wt.%)	After Run 1 (wt.%)	After Run 2 (wt.%)
U	80.96	81.90	81.10
Pu	6.05	7.66	7.54
Am	1.21	1.18	1.18
Zr	8.86	9.97	11.1

*Analysis resulted in 2.92% undissolved/unknown material.

Certainly much additional experimentation, testing, and optimization of the casting process is needed. However, these initial results demonstrate essentially no loss of Am due to volatilization during fabrication. In short, a path toward zero-loss fabrication of metallic fuels including minor actinides appears to be feasible.

IV. IRRADIATION TESTING

The Fuel Cycle Research & Development program in the early 2000's renewed the development of metallic fuels for actinide transmutation in future fast reactors, but it was hindered by the fact that there was no longer a domestic (U.S.) fast reactor available for irradiation testing of fuels. Since early metallic fuels were developed using experiments in thermal test reactors (such as the Materials Test Reactor in Idaho or CP-5 in Chicago), an irradiation test program was developed for implementation in the Advanced Test Reactor in Idaho.

Irradiation testing of advanced metallic fuels in the ATR has been underway since 2003. Testing in the ATR is performed inside of cadmium-shrouded experiment positions to remove >99% of the thermal flux incident on the test fuels, resulting in an epi-thermal driven fuel test that is free from gross flux depression[9] and produces an essentially prototypic radial temperature profile inside fuel rodlets (i.e., miniature test rods).

To date, two ATR test series (AFC-1,2) on transmutation fuels have been completed. As summarized in Table II, almost 20 different metallic fuel alloys have been tested to burnups as high as 30% with constituent compositions of Pu up to 30%, Am up to 12%, Np up to 10%, and Zr between 10 and 60%. In general, the performance of all of these substantially disparate metallic fuel alloys has been observed to be excellent, and their irradiation behaviors are generally consistent with historic norms for metallic fuels without minor actinide additions and having lower Pu or Zr contents.[10]

Although irradiation of these transmutation fuels inside the cadmium-shrouded experiment positions in ATR generally achieve temperature conditions that are fairly prototypic of fast reactor conditions, the test fuels do not experience neutron damage conditions prototypic of a fast-spectrum reactor. For this reason, cladding performance is not genuinely being challenged in these tests. Furthermore, even the fuel performance under these non-prototypic irradiation conditions must be validated. To this end, two metallic fuel compositions (Pu-12Am-40Zr and U-29Pu-4Am-2Np-30Zr) were irradiated for 235 EFPDs to approximately 12% burnup in the Phénix fast reactor as part of the FUTURIX-FTA experiment.[11] Postirradiation examination of these test pins is just beginning. Comparison of these Phénix irradiated metallic fuels with identical compositions irradiated under nominally the same conditions in ATR will be an important part of the validation process.

V. FUTURE DIRECTION

Future work is being undertaken with a view toward increasing the burnup potential of metallic fuels even more. Design innovations under investigation include: 1) lowering the fuel smear density below the traditional 75% in order to accommodate more swelling, 2) an annular fuel geometry in intimate contact with the cladding to eliminate the need for a sodium bond (with the central hole accommodating fuel swelling), 3) coatings/liners on the cladding inner surface to mitigate fuel-cladding chemical

TABLE II

Metallic Transmutation Fuel Alloys Irradiated in ATR

Metallic Fuel Alloy	Irradiation Time (EFPDs)	Peak LHGR (W/cm)	Peak FD (fiss/cm^3)	Peak BU (% HM)
Pu-40Zr	582	300	3.30E+21	22.6
Pu-60Zr	582	300	2.25E+21	26.7
Pu-12Am-40Zr	582	300	2.84E+21	22.6
Pu-10Np-40Zr	644	300	2.91E+21	20.3
Pu-10Am-10Np-40Zr	582	300	2.24E+21	17.7
U-25Pu-3Am-2Np-40Zr	653	330	3.54E+21	30.2
U-28Pu-7Am-30Zr	653	330	3.96E+21	26.1
U-29Pu-4Am-2Np-30Zr	653	330	3.91E+21	26.7
U-34Pu-4Am-2Np-20Zr	653	330	3.48E+21	17.2
U-20Pu-3Am-2Np-15Zr	364	350	2.30E+21	10.4
U-20Pu-3Am-2Np-1.0Ln*-15Zr	364	350	2.43E+21	11.5
U-20Pu-3Am-2Np-1.5Ln*-15Zr	364	350	2.22E+21	9.6
U-30Pu-5Am-3Np-20Zr	364	350	2.07E+21	11.5
U-30Pu-5Am-3Np-1.0Ln*-20Zr	364	350	2.21E+21	11.5
U-30Pu-5Am-3Np-1.5Ln*-20Zr	364	350	2.49E+21	11.1
U-20Pu-10Zr	483	350	3.25E+21	11.3
U-20Pu-3Am-2Np-10Zr	483	350	3.09E+21	11.6

*Ln is the designation for a lanthanide alloy comprised of 6% La, 16% Pr, 25% Ce, and 53% Nd (by weight).

interaction and enable higher temperature operation, 4) alternative major alloying elements to Zr that can stabilize the gamma phase of uranium under irradiation conditions, while increasing fuel alloy melting temperatures or offering other fuel performance benefits, and 5) minor alloy additions to immobilize lanthanide fission products where produced inside the fuel and prevent their transport to the cladding where they make up the primary species responsible for FCCI.

V.A. Alternative Alloy Systems

As the TRU content of a metallic transmutation fuel alloy increases, the fuel alloy melting temperature (solidus) decreases below that of U-10Zr or even U-20Pu-10Zr. This decrease can be offset by increasing the Zr content above 10 wt.%. As can be seen in Table II, metallic fuels with much higher Zr contents have been the subject of extensive testing. While the performance of these high Zr content metallic fuels has been observed to be very good, it comes as the price of a decreasing actinide (i.e., fissile) density in the fuel. As an alternative to dramatically increasing the Zr content, other major alloying element(s) are under investigation. Two such alloy systems that are showing potential are Mo-Ti-Zr and Ta.

For example, although U and Ta are immiscible, success in alloying U-Zr-Ta has been achieved. Fig. 3 shows an SEM micrograph of a U-10Zr-5Ta as-cast alloy with the chemical compositions of all phases present determined by EDX (Table III). Modest additions of Ta could significantly increase the fuel alloy solidus while stabilizing the gamma phase of U to even lower temperatures than are achieved by Zr alone.

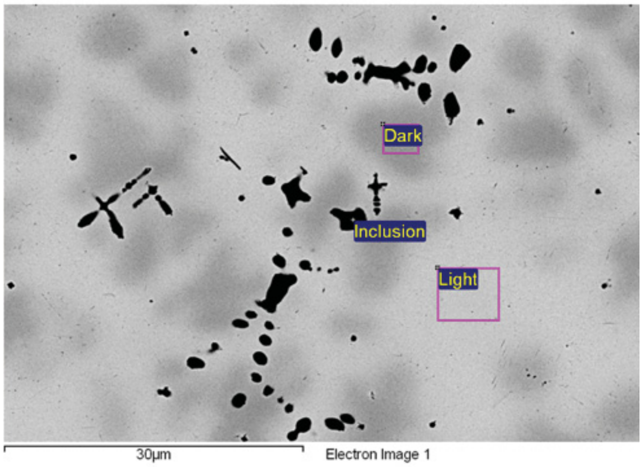


Fig. 3. SEM micrograph of U-10Zr-5Ta alloy.

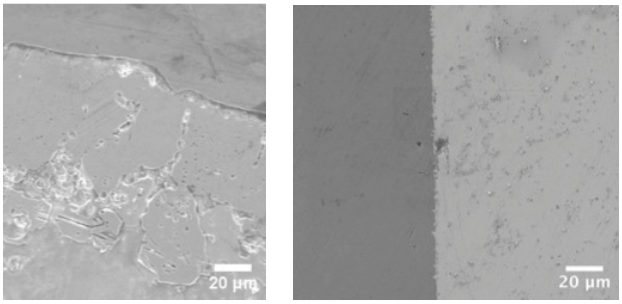
TABLE III

Chemical Compositions of Phases in U-10Zr-5Ta Alloy

Phase	U (wt.%)	Zr (wt.%)	Ta (wt.%)
Light	93.13	4.88	1.99
Dark	70.07	18.61	11.31
Inclusion	3.98	83.94	12.08

V.B. Minor Alloy Additions to Immobilize Lanthanides

In U-Zr and U-Pu-Zr metallic fuels, lanthanide fission products migrate to the fuel periphery and become the primary species responsible for fuel-cladding chemical interaction. FCCI thins the cladding wall, increasing cladding stress. The rate of FCCI attack increases with temperature. As a result, FCCI caused by the lanthanide fission products (on stainless steel cladding) limits cladding temperatures to approximately 600°C. Development of coatings/liners (e.g., Zr, V, TiN, Cr) that could be applied to the inner surface of the cladding to act as a diffusion barrier, mitigating FCCI, are under investigation. However, a more elegant solution to the FCCI issue would be the addition of a minor alloying element that would react with the lanthanide fission products in situ, immobilizing them in the fuel matrix and preventing their migration to the cladding. Preliminary alloy casting and interdiffusion experiments suggest that palladium may be able to achieve the desired effect in relatively small quantities. In casting studies in which both Pd and lanthanide elements were included in the fuel alloy, Pd reacted with the lanthanides to form a separate, stable intermetallic phase.[12] In separate experiments, diffusion couple studies showed that although Pd and Fe (the major constituent in stainless steel cladding) strongly react, including liquefaction, at 700°C, a PdNd intermetallic (i.e., 1:1 atomic ratio) showed no reaction with Fe at the same temperature[13] (see Fig. 4).



(a) (b)

Fig. 4. (a) Liquifaction of Nd-Fe diffusion couple at 700°C; (b) no reaction between Nd/Pd-Fe diffusion couple at 700°C.

Thermodynamic calculations suggest that 2 wt.% Pd would be needed in the metallic fuel alloy to stabilize the lanthanide fission products produced per 10% burnup. Metallic fuel alloys with Pd additions are currently under irradiation in the ATR.

VI. CONCLUSIONS

Metallic fuels have had a long history of use in sodium-cooled fast reactors. Continued research, development, and design innovation over a period of decades have demonstrated this fuel technology to be safe and reliable to extremely high burnups over a wide range of compositions. Their simple fabrication has historically given them a significant economic advantage over other fuel forms. Research and development on metallic fuels continues, with particular emphasis on showing that this fuel technology can incorporate significant quantities of plutonium and minor actinides for the purpose of efficiently and economically transmuting these elements in a fast-spectrum reactor. New fabrication methods to improve retention of the volatile fuel constituents during the casting process (and reduce waste generation), are being developed. Irradiation testing of the new metallic fuels has so far indicated that the new compositions, although differing significantly both in the quantities of Pu and minor actinides included and in the Zr alloying addition, perform as a function of fission density very analogously to the historic U-Zr and U-Pu-Zr fuels.

In the next few years it is anticipated that a new casting furnace prototype will demonstrate that these new metallic fuel alloys can be fabricated at engineering scale, in a remote environment, with no significant fuel loss or waste generation. Irradiation tests currently underway will provide the performance data needed to assess fuel reliability/burnup capability and identify any unique behaviors that are a result of the new compositions. If these future results are consistent with the intermediate data available at present, metallic fuels are expected to continue to be a strong contender for the fuel technology choice of future fast reactors.

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NOMENCLATURE

ATR	Advanced Test Reactor
EBR-II	Experimental Breeder Reactor II
EDX	Energy Dispersive X-ray Spectroscopy
EFPD	Effective Full Power Day
FCCI	Fuel-cladding Chemical Interaction
GACS	Glovebox Advanced Casting System
HM	Heavy Metal
IFR	Integral Fast Reactor
LHGR	Linear Heat Generation Rate
LMFBR	Liquid Metal Fast Breeder Reactor
LWR	Light Water Reactor
SEM	Scanning Electron Microscope

SFR	Sodium Fast Reactor
TRU	Transuranic

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