

Fuel Design for the U.S. Accelerator Driven Transmutation System

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Abstract - *The U.S. concept for actinide transmutation is currently envisioned as a system to destroy plutonium as well as minor actinides in a single or two tier system. In order to maximize the actinide destruction rate, an inert matrix fuel is used. The effectiveness of transmutation in reducing the actinide inventory is linked to the development of a robust fuel system, capable of achieving very high burnup. Very little fuel performance data has been generated to date on inert matrix systems, and there are several issues specific to the behavior of higher actinides that do not allow extension of the existing uranium-plutonium fuel database to these new fuels. These issues include helium production, fuel-cladding-chemical-interaction, and americium migration. In the early 1990's, two U-Pu-Zr metal alloy fuel elements containing 1.2 wt.% Am and 1.3 wt.% Np were fabricated and irradiated to approximately 6 at.% burnup in the Experimental Breeder Reactor-II. Postirradiation examination results were not published; however the recent interest in fuel for actinide transmutation has prompted a reexamination of this data. The results of the postirradiation examination of this experiment, including gas sampling, metallography, and gamma scanning are discussed. Available data on inert matrix fuels and other fuels incorporating actinides are used to assess the implications of minor-actinide specific issues on transmuter fuel. Considerations for the design of nitride and oxide fuels, metallic fuels, and metal-matrix dispersion fuels are discussed.*

I. INTRODUCTION

The development of fuels for higher actinide transmutation systems presents many technical challenges. Fuel must behave in a benign manner during core off-normal events, maintain integrity to high burnup, lend itself to low-loss recycling processes, and be easily fabricable in a remote environment. A transmutation system cannot function effectively without a fuel that meets these criteria.

Little data, however, is currently available to support the selection of a fuel type for an accelerator driven transmutation system. The results of only two experiments on americium and neptunium-bearing fuels have been published; only one of these used an inert-matrix fuel form.^{1,2} In order to add to the available database this paper presents summary results of the X501 minor actinide burning experiment conducted in the EBR-II (Experimental Breeder Reactor) during the early 1990's. The known

data on minor actinide irradiation behavior and properties is then used to comment on potential fuel design issues for dispersion fuel, ceramic fuel types, and metallic fuel.

II. THE X501 MINOR ACTINIDE BURNING EXPERIMENT

The X501 experiment was conducted in EBR-II as part of the IFR (Integral Fast Reactor) program to demonstrate minor actinide burning through the use of a homogeneous recycle scheme. The X501 subassembly contained two metallic fuel elements loaded with relatively small quantities of americium and neptunium. Interest in the behavior of minor actinides (MA) during fuel irradiation has prompted further examination of existing X501 data, and generation of new data where needed in support of the U.S. waste transmutation effort. The X501 experiment is one of the few minor actinide-bearing fuel irradiation tests conducted

worldwide, and although it did not use an inert matrix fuel, knowledge can be gained by understanding the changes in fuel behavior due to addition of MA's. Of primary interest are the affect of the MA's on fuel-cladding-chemical-interaction, and the redistribution behavior of americium. The quantity of helium gas release from the fuel and any effects of helium on fuel performance are also of interest. It must be stressed that information presented at this time is based on the limited PIE conducted in 1995-1996, and currently represents a set of observations rather than a complete understanding of fuel behavior.

II.A. Fabrication and Irradiation

Elements were fabricated by using differential pressure to inject molten metal from a casting crucible into a vitreous silica mold. Details of the casting process and resulting microstructure are given in reference [3]. Considerable americium was lost due to volatilization during the fabrication process, which was not designed for use with am-bearing alloys. Fuel slugs were fabricated into the EBR-II MkIV fuel element configuration; the chemical analysis and physical attributes of these elements are given in Table I.

Figure 1 shows optical images of fuel cross-sections taken from locations near the top and bottom of a pin from the same casting batch as the irradiated fuel pins. Figure 1(a) was taken from the top section of the fuel pin, and shows the inhomogeneous structure typical of U-Pu-Zr fuel produced by this casting method. Wavelength dispersive spectrometry (WDS) analysis shows that the distribution of americium and neptunium is uniform across the section. Figure 1(b) shows a micrograph of a section taken from the lower part of the fuel pin, in a region where cooling is slower after solidification due to proximity with the furnace internals. In this region of the fuel, WDS indicates that a few americium-rich precipitate phases are beginning to form, and that these precipitates are uranium depleted relative to the matrix.

The two MA-bearing pins were inserted into a standard EBR-II subassembly with the remainder of the fuel element locations filled with U-10Zr driver fuel. The X501 subassembly was inserted into EBR-II beginning

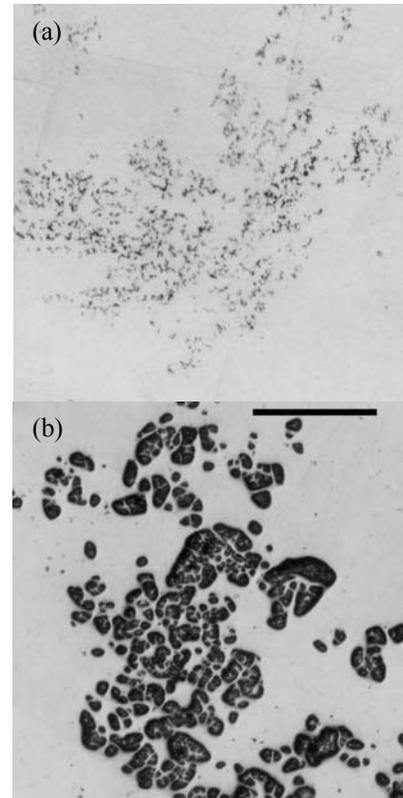


Figure 1. Microstructure of as-cast fuel pin. (a) top section, (b) lower section. Bar = 500 μ m.

TABLE I

Attributes of X501 minor actinide bearing fuel elements

Composition (average analyzed wt%)	U-20.2Pu-9.1Zr-1.2Am-1.3Np
Major impurities (average wt%)	Si:0.26, Al:0.089, Ca:0.067, Cr:0.0025, Mg:0.009, Fe:0.001, Mn:0.001
U-235 enrichment (nominal)	60%
Fuel mass	77.5 g
Fuel length	34.3 cm (13.5 in.)
Element length (nominal)	74.9 cm (29.5 in.)
Cladding type	HT-9 steel
Cladding OD	5.84 mm (0.230 in.)
Cladding wall	0.457 mm (0.018 in.)
Slug diameter	4.27 mm (0.168 in.)
Plenum volume	7.1 cm ³
Plenum gas	75He-Ar
Smear density	75%
Fuel slug density, % theoretical	99.5%

in February 1993, and withdrawn just prior to EBR-II shutdown in August 1994. Total irradiation time was 339 EFPD's. Burnup, calculated on the basis of REBUS/RCT/ORIGEN calculations⁴ was 7.6% HM with transmutation of 9.1% of ²⁴¹Am. Peak linear heat generation rate was estimated to be 45 kW/m (13.7 kW/ft) and peak fuel centerline and cladding inner surface temperatures were approximately 700°C and 540°C, respectively.

II.B. Postirradiation Examination

A partial postirradiation examination was completed on X501, including gamma scanning, optical microscopy, microprobe analysis, and metallography. Gamma scans (not shown) showed normal metallic fuel fission product behavior, with ¹³⁷Cs alloying with the bond sodium and migrating to the region near the top of the fuel slug.

A microscopic examination of the inside cladding surface was made to determine if the inclusion of the MA's to U-Pu-Zr fuel has an effect on FCCI (Fuel-Cladding Chemical Interaction). The HT-9 cladding used for the X501 experiment is also the reference cladding for U.S. transmuter fuel. Optical microscopy (Fig. 2) showed no evidence of reaction layer formation on the inner cladding wall or the outer surface of the fuel slug. A gap is visible between the fuel and the cladding wall at all locations. These preliminary results indicate that under typical metal fuel operating conditions, FCCI (fuel-cladding-chemical-interaction) of HT-9 is not strongly affected by small amounts of americium or neptunium.

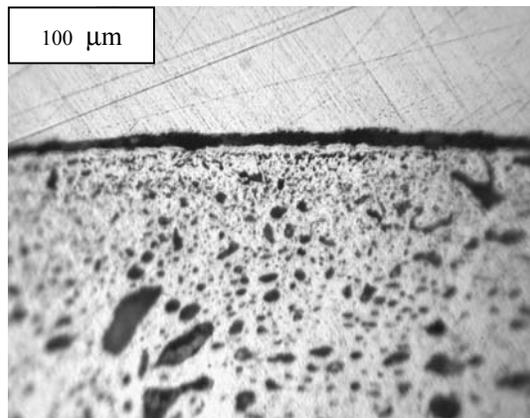


Figure 2. Optical micrograph of fuel/cladding interface.

Shown in Fig. 3(a) is a postirradiation optical micrograph of a cross section near the center of element G582. The fuel shows a microstructure typical of U-20Pu-10Zr, where constituent radial redistribution has resulted in the formation of three microstructural zones within the fuel. Fig. 3(b) is a comparison of the WDS traces of

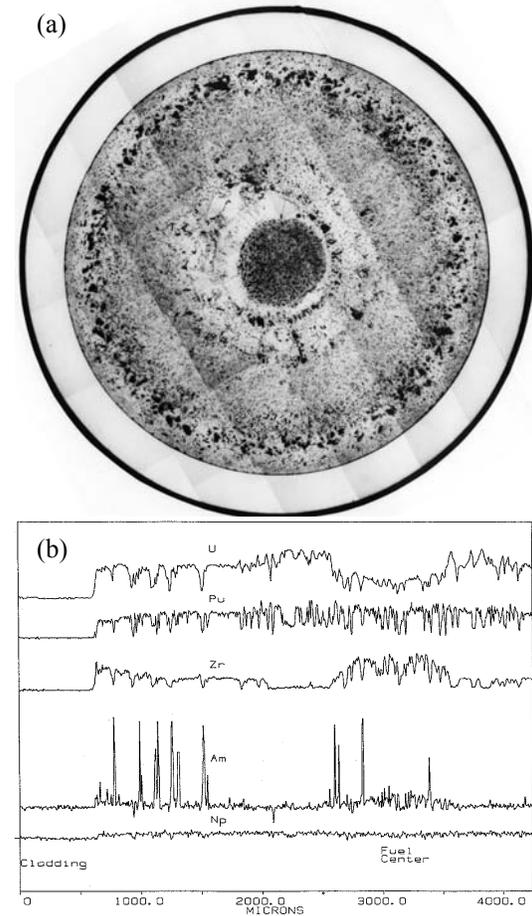


Figure 3. (a) Postirradiation optical image of MA-bearing fuel. (b) WDS line scans showing distribution of actinides and Zr.

neptunium, americium, zirconium, plutonium, and uranium across the section shown in Figure 3(a). The outer zone, near the cladding, is enriched in zirconium, the intermediate zone is zirconium depleted, and the central zone is enriched in zirconium and depleted in uranium. Plutonium content remains relatively uniform. Americium appears in the WDS traces as features with high elemental concentrations, generally depleted in U, Pu, and Zr (the other major elemental constituents). The morphology of the americium-rich phases cannot be determined from existing micrographs, and other

elemental constituents, such as fission product and impurity elements were not analyzed during the examination. Comparison of the preirradiation and postirradiation WDS line scans suggests that the americium-rich features formed during irradiation. Americium is present only in the uranium-depleted central and outer zones, indicating that americium migration has occurred along with the migration of uranium and zirconium. Local radial redistribution of americium to the cladding inner wall does not occur; however these results do not rule out the possibility of condensation of americium in the plenum region above the fuel column.

Plenum gas sampling results were combined with ORIGEN calculations to estimate the fission gas and helium release rates from the fuel. Results are shown in Table II. Helium generation is principally due to a sequence of neutron capture by ^{241}Am , and subsequent decay of the ^{242}Cm product to ^{238}Pu by alpha particle emission. The amount of He introduced as plenum fill gas was estimated based on pressure ratios between fill gas and the quantity of radioactive Xe tag gas measured by counting Xe activity. Based on these estimates, it appears that approximately 90% of the helium gas produced was released to the plenum. Fission gas release was 79%, typical of U-Pu-Zr fuel at this burnup.

TABLE II
X501 gas production and release

BOL ^{241}Am content	0.972 g
EOL ^{241}Am content	0.884 g
^{241}Am transmutation	0.088 g (9.1 %)
Measured He release	3.1 cm ³
Calculated He inventory, 1 yr. decay	3.4 cm ³
He release	90%
FG release	79%

III. DESIGN OF INERT MATRIX FUEL FOR THE U.S. TRANSMUTATION SYSTEM

The reference U.S. transmuter core used for fuel design is a sodium cooled 840 MWt system operating at a peak linear power of 40 kW/m.⁵ A heavy metal burnup of about 30% is desirable to minimize the number of recycle/refabrication steps and limit the waste generation inherent in these processes. The current single strata actinide destruction scenario burns LWR generated plutonium along with the minor

actinides neptunium, americium, and curium in an accelerator driven system. Also under consideration are dual strata systems which use intermediate reactors to more efficiently extract energy from LWR generated plutonium. Only the minor actinides and some residual plutonium are burned in the accelerator driven system (ADS).

In addition to the high burnup requirement, there are several other new fuel performance issues associated with the development of ATW fuel. These include exposure of fuel and cladding to a very hard neutron spectrum, the potential for americium migration, and helium generation. HT-9 has been chosen as the reference cladding for demonstration of transmuter fuel because of its high resistance to swelling under fast reactor conditions. A few fuel pins have been successfully irradiated to a fast fluence of 3.9×10^{23} n/cm² in the Fast Flux Test Facility (FFTF), thus a fluence limit of 4.0×10^{23} n/cm² has been chosen for HT-9 transmuter cladding. Although radial migration of americium to the cladding inner wall was not observed in the X501 experiment, the question of axial migration requires further analysis and verification through irradiation testing.

The generation of helium gas during transmutation of ^{241}Am is a significant issue that must be considered in the design of transmuter fuel. In fuels with high americium content, helium generation is principally due to neutron capture by ^{241}Am and subsequent alpha decay of ^{242}Cm to ^{238}Pu . There are three known experiments involving transmutation of ^{241}Am . Helium release results from two, the EFTTRA-T4 test and the SUPERFACT-1 experiment have been previously published. Some data on helium generation is available for each experiment, and results are presented in Table III, along with ORIGEN calculations for the U.S. Na-cooled point design and X501 data. The quality and completeness of data vary for each experiment; therefore the results of this analysis contain a good deal of uncertainty, and do not provide a good basis for extrapolation. A rule-of-thumb for estimating helium production from americium for the U.S. transmuter design, based on the results in Table III and calculations of compositions ranging from 6-40 wt% ^{241}Am is 50 ml He per gram of transmuted americium. The wide range of possible fuel compositions leads to a wide range in the potential for helium production. Helium production is likely to be the most important fuel design consideration for transmutation scenarios with high MA content.

Table III

Data and calculations for He production
in ^{241}Am -bearing fuels

Experiment	Specific He generation (ml per g gram ^{241}Am transmuted)
X501	38.6-44.8
SUPERFACT-1	33.3-63.4
EFTTRA-T4	44.8-59.9
Na point design	58.1

The limited amount of fuel performance data available on inert matrix fuels containing americium, and the possibility of changes in fuel performance as a function of composition, forces consideration of multiple fuel types for the transmutation mission. In the U.S., consideration is being given to four fuel types; metallic alloy fuels, nitride pellet fuel, oxide pellet fuel, and metal-matrix dispersion fuel. Each of these fuel types has demonstrated acceptable fuel performance in uranium-bearing analogues to the fertile fuels under consideration. Many fuel performance questions are raised, however, for the isotopic compositions required for transmutation.

III.A. Ceramic Fuels

There are two possible approaches to dealing with helium gas produced in oxide and nitride fuels that contain americium. The first is to design and operate the fuel under conditions which promote gas release. The second is to design the fuel to effectively retain fission gas and helium while maintaining an acceptable level of gas-driven swelling, sometimes referred to as a 'cold fuel'. The two published data points on americium-bearing oxide fuel effectively demonstrate these two approaches. The former is illustrated by the SUPERFACT⁶ experiment, which tested, in addition to other fuel compositions, two uranium oxide matrix pins containing 20 wt.% americium in the Phénix fast spectrum reactor. The fuel exhibited gas release of ~60%,¹ typical of oxide fast reactor fuel and an acceptable level of fuel swelling at 4.5 at.% burnup.

The thermal spectrum EFTTRA-T4 test⁷ used a microdispersion of americium oxide in a magnesium-aluminate spinel matrix. Gas release was a fraction of that measured in the SUPERFACT pins. Pellets in this test exhibited

volumetric swelling of ~18 vol.%, resulting in excessive cladding strain.

It is clear that if gas is to be retained by the fuel, the matrix material must be carefully chosen to avoid excessive gas-driven swelling. The temperature dependence of swelling and gas release must also be taken into account, so that core thermal excursions do not result in rapid fuel swelling or rapid release of gas resulting in a catastrophic fuel failure.

Actinide nitrides are likely to be compatible with a liquid metal bond phase, including sodium and lead-bismuth alloys. The use of a high thermal conductivity metallic bond allows an additional degree of latitude in fuel design; because the temperature gradient across the fuel to cladding gap will be small, a large gap (low smeared density) can be used accommodate the volume increase of high swelling fuels.

III.B. Dispersion Fuel

Dispersion fuels are attractive for transmutation systems because of their potential for resistance to fuel failure at high burnup. Limited fuel testing done in the early 1960's showed that, in certain cases, steel matrix dispersions were capable of reaching burnups in excess of 70% ^{235}U (in HEU fuel).⁸ Ideally, a dispersion fuel is an array of particles uniformly distributed in a chemically inert, non-fertile matrix. Each particle is then, in effect, contained within a thick-walled pressure vessel. Low fuel temperatures are assured by the thermal conductivity of the continuous matrix phase. Dispersion fuel performance is, however, linked closely to the microstructural characteristics of the dispersion, the volume loading of fuel particles, and the fuel operating temperature. Microstructures that approach that of the ideal uniform dispersion of spheres allow for higher burnup.

A simple model was formulated in order to gauge the effects of helium production due to americium on the state of stress within the matrix of an ideal dispersion. In this model, the dispersion is taken as a collection of independent spherical fuel particles, each surrounded by a chemically inert thick-walled spherical pressure vessel. The wall thickness of the pressure vessel is determined by particle size and particle volume loading. Fission fragments ejected from the fuel particle are assumed to damage the matrix, so that a weak damage zone is established around each particle, according to an estimated fission fragment range. Fission gas

release from the fuel particles is calculated according to the correlation of Storms.⁹ The helium gas release rate was estimated to be five times that of fission gas, based on data from EFTTRA-T4. Gas was assumed to expand into the volume of porosity available within the fuel particle, and pressure was calculated using the ideal gas law. The relative magnitude of the effective stress components generated in the pressure vessel wall can then be compared as a function of americium content, temperature, particle size, and particle volume loading. This approach was used by Wier¹⁰ in the early 1960s, and assumes that fuel failure is due to stress in the matrix surrounding a fuel particle produced by gas pressure. This simple model does not take into account stress due to solid fission product or gas-driven fuel swelling.

Figure 4 shows the relative effects of AmN content on stress for a 25 vol.% dispersion at 500°C (773K) for spherical particle sizes ranging from 100 to 400 microns. Also shown as a horizontal line is an estimate of the gas-generated matrix stress at the failure threshold from literature data for a high burnup UO₂⁶ dispersion. The plot shows that, under the assumptions of this simple model, Am-bearing dispersion fuels have reasonable prospects for success as transmutation targets. The results, however, depend heavily on assumptions made about the release of fission gas and helium from the fuel particles, fuel particle swelling, and the failure mechanism, and depend on the ability to fabricate a fuel with an ideal microstructure.

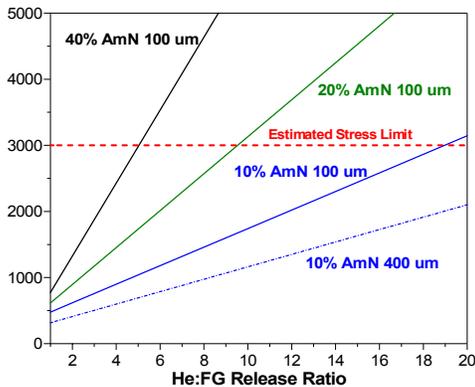


Figure 4. Gas-generated stress in dispersion fuel as a function of helium release rate,²⁴¹Am content, and fuel particle size.

III.C. Metallic Fuel

Similar to oxide fuel, there is an extensive database on the behavior of uranium-bearing metallic alloy fuel under fast-spectrum irradiation conditions. Metallic U-Pu-Zr alloys were the reference fuel for the U.S. Integral Fast Reactor (IFR) program. Metal fuel has a propensity toward high gas-driven swelling. Early metal fuel designs attempted to restrain swelling, inevitably resulting in unacceptably large cladding deformation at low burnup. Later designs allowed for fuel swelling by increasing the gap size, resulting in a fuel with 75% of the cross-sectional area of the interior of the cladding tube being occupied by fuel at beginning of life. This configuration allows the fuel to freely swell about 30% by volume before contacting the cladding wall. At this point the fuel develops a network of interconnected porosity, resulting in a weak mass that cannot exert substantial mechanical force on the cladding. The network of porosity leads to a high gas release rate, on the order of 80%. Metal fuel pins of this design reached burnups approaching 20% HM (Heavy Metal) in EBR-II.^{11,12} Since metal fuel is engineered to allow for swelling and promote gas release, the issue of helium generation is a matter of (1) sizing the gas plenum to prevent excessive gas pressure driven cladding creep and (2) ensuring that gap size is adequate to accommodate solid fission product swelling.

Data on the irradiation behavior of inert-matrix metal fuels is scarce. Waldron reported a swelling rate of 6.5%/burnup for an extruded Pu-36.4Zr in the δ -Pu phase, irradiated at 500°C to 0.8 at.% burnup, including 144 thermal cycles.¹³ Another study showed that cold-rolled Zr-5Pu and Zr-7Pu specimens showed little swelling but grew anisotropically 200~500% after irradiation to 0.8-1.3 at.% burnup at 430°C-530°C.¹⁴ The latter alloy exhibits the hexagonal α -Zr equilibrium crystal structure. It is probable that texture introduced by the fabrication process was responsible for the extreme change in shape.

Perhaps the most important question relating to the use of Pu-Am-Np-Cm-Zr alloys (U is also present in fractional percentage quantities as carryover from MA separations and is produced in the fuel by transmutation) as transmuter fuels are the unknown phase equilibria in the multi-component alloy system. Reasonably complete information is available on the Pu-Zr, Pu-Am, Pu-Np,¹⁵ and Pu-Cm¹⁶ systems. Americium stabilizes the desirable cubic δ -Pu phase, and Am and Pu form a wide range of solid solutions with this structure. Pu and Zr form a (δ -Pu, β -

Zr) solid solution from 0-40 wt.% Zr. Above 40 wt.% Zr, a two phase mixture of α -Zr and δ -Pu forms below 597°C, which is likely to be less desirable based on the limited data on Pu-Zr irradiation behavior. From temperatures above the δ -Pu containing regions of the phase field to the solidus, there is complete mutual solid solubility of Pu and Zr in the body-centered-cubic (ϵ -Pu, β -Zr) phase. There is also complete solid solubility of Np in ϵ -Pu; however, solubility of Np in δ -Pu is limited to a few percent. There is conflicting information on published Np-Zr phase equilibrium data obtained by different methods. According to Rodriguez, et. al.¹⁷, who investigated the Np-Zr system using dilatometry and metallography, β -Zr and γ -Np exhibit complete mutual solid solubility and regular solution behavior at high temperature. Gibson and Haire,¹⁸ however, identified endothermic events consistent with low-melting phases in a study using thermal analysis. The potential for immiscibility and formation of an inhomogeneous microstructure is not a fuel performance issue, as shown by the excellent performance of multi-phase U-Pu-Zr fuel.¹⁹ Rather, the formation of low melting Np-based phases in the complex alloy system is an issue that must be experimentally determined. If such a phase forms, the addition of a minor alloying element may be required to suppress formation of a liquid phase.

IV. CONCLUSIONS

The U.S. fast reactor fuel program demonstrated the use of americium-bearing fuel in the early 1990's. Limited PIE results from the X501 experiment indicate that the addition of 1.2 wt.% of americium did not alter the behavior of metallic U-Pu-Zr fuel.

The lack of MA-bearing inert-matrix fuel performance data forces the consideration of multiple fuel types for MA transmutation. Currently in the U.S., consideration is being given to four fuel types; metallic TRU-Zr alloys, nitride pellets, oxide pellets, and metal-matrix dispersion fuel. Each of these fuel types has demonstrated acceptable fuel performance in uranium-bearing analogues to the fertile fuels under consideration. Many fuel performance questions are raised, however, for the isotopic compositions under consideration and the use of an inert matrix. Foremost among these issues is the generation of large amounts of helium due to capture and decay processes in americium.

Experimental results are needed to clarify the issues of gas retention versus release in ceramic fuel, the presence or absence of low-solidus phases in metal fuel, and the capability of dispersions to retain large amounts of helium.

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