# CURRENT US PLANS FOR DEVELOPMENT OF FUELS FOR ACCELERATOR TRANSMUTATION OF WASTE

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## Abstract

The United States is currently investigating the feasibility of proposed technologies for the Accelerator Transmutation of Waste (ATW) concept, which is funded as part of the U.S. Department of Energy's Advanced Accelerator Applications (AAA) Program. The ATW concept is proposed as a means to transmute transuranic isotopes and, perhaps, long-lived fission products removed from light water reactor spent fuel to shorter-lived fission products. To attain maximum possible transmutation rates, no fertile material (i.e., U-238 or Th-232) is to be incorporated into the fuel. Fuel forms currently proposed for ATW application include non-fertile dispersions of metal alloy or nitride fuel particles in a metal matrix, a non-fertile metal alloy, or non-fertile nitride pellets for a fast-spectrum, liquid metal-cooled transmuter, and non-fertile TRISO-coated particles dispersed in graphite compacts for a thermal-spectrum, gas-cooled transmuter. There is little or no experience with these non-fertile fuels, so an extensive fuel development program is envisioned. Current plans call for initial effort to demonstrate feasibility of the proposed fuel forms by the end of 2005, consistent with AAA program decision milestones. Feasibility research and development will consist of the following:

- Development of fabrication processes to demonstrate fabricability of the proposed fuel forms;
- Simple irradiation tests to screen samples of each fuel type for unexpected or poor performance; and
- Determination of intrinsic properties or characteristics (e.g., out-of pile interdiffusion behavior of fuel and constituents and thermophysical properties).

If the decision is made to continue development of the ATW concept beyond 2005, then of the successful candidate forms, one or two will be selected for further development, with more extensive irradiation testing and fuel property characterization.

## 1. INTRODUCTION

The Accelerator Transmutation of Waste (ATW) concept has been proposed as a means to reduce or eliminate transuranic elements from commercial high-level nuclear waste in the U.S. As envisioned, the concept consists of a transmuter in which transmutation by fission (or by neutron capture in long-lived fission products, or LLFPs) is accomplished in a neutron flux, provided by an accelerator-driven neutron source and multiplied through a subcritical blanket containing actinides [1].

The transmuter is designed as a spallation neutron source-driven, subcritical blanket, with an environment similar to those of nuclear reactor cores. Therefore, much of the transmuter design would be based on technology already developed for nuclear reactors, including design of heat-exchange systems, configuration of transuranic-bearing blanket fuel into coolant channels, and design of blanket fuel rods, target rods and assemblies. Although the candidate technology options for an ATW system are based on reasonably-mature reactor technologies, the stated features of the system require fuel types with which there is little or no experience. Specifically, the ATW concept calls for use of non-fertile (i.e., non-uranium-bearing) fuels to allow the system to achieve maximum transuranic destruction rates. Because much of the development of reactor fuel over the past several decades was motivated by the desire for sustainable energy generation, the great majority of relevant fuel experience is with uranium-bearing fuel. The proposed transmutation system thus requires that a new and unproven fuel be developed, characterized, and qualified. This paper summarizes the current U.S. R&D plans to develop blanket fuel for an ATW transmuter, with emphasis on establishing by the end of fiscal year 2005 the feasibility of the fuel forms considered.[*] The approach reported here is described more fully in a yet-unpublished document entitled

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[*] Although technologies proposed for consideration for an ATW concept include liquid-fueled concepts as well as solid-fueled concepts, at the time of this writing, solid fuel technology options are considered more viable.

"Accelerator Transmutation of Waste: Fuel and LLFP Target Development Plan for FY'01 through FY'05". Upon successful completion of the described activities, sufficient information will have been obtained to support decisions regarding the feasibility of proposed ATW fuel technologies. Although a parallel development of several fuel types is underway, down-selection to a reference and an alternative fuel type will be necessary upon completion of the feasibility R&D phase, if not sooner.

## 2. TRANSMUTER REQUIREMENTS FOR FUEL

### 2.1. General fuel requirements

Specific requirements for blanket fuel performance will be derived from the requirements for ATW system performance, such as actinide destruction rates, safety requirements, and fuel processing. However, some general fuel requirements can now be identified and are listed as follows:

- ATW fuel must be fabricable using remote processes. The majority of the head-end processes being considered to prepare LWR spent fuel for transmutation and the processes being considered for recycle of transuranics back into the transmuter would leave residual fission products in the blanket fuel feed. Therefore, fuel fabrication will likely be conducted in a hot cell environment, demanding that fabrication processes be amenable to remotization.
- ATW fuel must be compatible with the fuel recycle process. If transuranics are to be recycled back into the ATW transmuter, then some type of chemical or mechanical processing of the irradiated blanket fuel will be required.
- The blanket fuel must maintain the fissile material in a predictable configuration and location in the transmuter.
- The blanket fuel must provide robust containment for fission products and radionuclides, as a first barrier for safety, during normal operations; i.e., cladding failure with leakage of fission products must be a low-probability event.
- The blanket fuel must retain a coolable geometry during normal operations and all design basis accidents.

### 2.2. Emerging specific requirements

Two primary types of transmuters have been proposed for ATW application [1]. One type of system is a fast-spectrum system, cooled by either sodium or a eutectic composition of lead-bismuth alloy. Two proposed fast-spectrum systems are loosely based on the U.S. concept for the sodium-cooled Advanced Liquid Metal Reactor (ALMR), developed by General Electric using Integral Fast Reactor technology [2, 3], and the Russian lead-cooled BREST reactor system [4]. Another proposed fast spectrum-system would be cooled by helium and would be similar to the Gas Cooled Fast Reactor concept (e.g., [5]).

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Furthermore, technical issues requiring resolution for solid and for liquid fuels are quite different in nature, and therefore necessitate separate treatment. Therefore, efforts to address liquid fuel R&D have been deferred until an appropriate time.

The second type of system is a gas-cooled, thermal-spectrum system proposed by General Atomics (GA) in the U.S.

Although these system types are not described here, requirements for fuel for some of the transmuter concepts are listed in Table 1. Many of the requirements are tentative, based on assumptions used for calculations to support the evaluation of system point designs. Such requirements are likely to change or evolve as limits to fuel characteristics (e.g., fissile loading) or performance (e.g., operating temperature or burnup) are determined or as the tramsmuter concepts evolve.

TABLE 1. EMERGING REQUIREMENTS OR DESIRED CAPABILITIES OF PROPOSED ATW TRANSMUTER CONCEPTS

| Sodium-cooled transmuter fuel requirements [6] | |
|---|---|
| Burnup potential: | > 30% desired |
| Linear heat generation rate: | 400 W/cm, possibly up to 500 W/cm |
| Fuel TRU density: | 3.2 to 4.2 g/cm$^3$ in fuel slug as described |
| | 2.7 to 3.5 g Pu/cm$^3$ inside cladding I.D. of 0.632 cm |
| | 2.0 to 2.5 g/cm$^3$ within fuel pin of O.D. 0.744 cm |
| **Lead-bismuth-cooled transmuter fuel requirements [7]** | |
| Burnup potential: | > 30% desired |
| Linear heat generation rate: | 300 W/cm |
| Fuel TRU density: | 3.2 to 4.6 g/cm$^3$ in fuel slug as described |
| | 2.7 to 3.9 g Pu/cm$^3$ inside cladding I.D. of 0.440 cm |
| | 1.5 to 2.2 g/cm$^3$ within fuel pin of O.D. 0.580 cm |
| **Gas-cooled, thermal-spectrum transmuter fuel requirements [8]** | |
| Achieve very high burnup of the initial loading of plutonium and minor actinides. | |
| Maintain fuel particle integrity throughout the fuel cycle and into disposal, if possible, avoiding the need for intermediate reprocessing. | |
| Maintain high operating temperatures needed to achieve a high net thermal efficiency. | |

## 3. SELECTION OF CANDIDATE FUEL FORMS

Development of new, non-fertile fuel forms is best initiated with consideration of non-fertile analogues to more-familiar uranium-bearing fuel forms. A varied experience base exists with several chemical and physical forms, which are listed in Table 2. Considerable effort will be required to establish feasibility by 2005 for any of the potential fuel types. Technical judgement and experience provide the basis for limiting the fuel candidates to those that appear most attractive and are most likely to be successful.

TABLE 2. PREVIOUSLY-INVESTIGATED CHEMICAL AND PHYSICAL FUEL FORMS

| Chemical forms | Familiar examples |
|---|---|
| Metallic (usually alloy) | U-Fs, U-Zr, U-Pu-Zr |
| Oxide | $UO_2$, $(U,Pu)O_2$ |
| Nitride | UN, (U,Pu)N |
| Carbide | UC, (U,Pu)C |
| Intermetallic compound | UAlx, Uranium silicides |
| Physical forms | Familiar examples |
| Monolithic | |
| Metal alloy slug | Na-bonded U-Zr for EBR-II |
| Pressed pellets | $UO_2$ for LWRs |
| Particulate | - Vipac<br>- Sphere-pac |
| Dispersion (with metal or ceramic matrix, with metallic or compound fuel dispersant) | - $UAl_x$ in AL matrix for research reactors<br>- Coated $UO_2$ particles in graphite compacts used for gas-cooled reactors |

The advantages and disadvantages of each of the fuel option is not addressed here; however, some rationale for selection of candidate fuel forms for development is provided. Some of the available fuel options were chosen as candidates based on programmatic considerations. For example, oxide fuel, including oxide fuel particles in a dispersion fuel, is not a candidate fuel form because of considerations related to chemical processing. (Pyroprocessing techniques – though feasible – are more complicated for oxide fuel than for other fuel types). Aqueous recycle processes that are currently established for oxide fuel are considered undesirable for ATW due to proliferation concerns. Carbide and nitride fuels are equally attractive in terms of properties. Although a more extensive database exists for carbide fuels than for nitride fuels, of the two, only nitride fuels are considered for ATW due to compatibility with the pyroprocess and due to interest in nitrides in Japan, Europe, and Russia. Particulate fuel types, such as vi-pac, are not considered because it is unlikely that the cost associated with development of a fuel type with which the U.S. has little experience could be supported by the ATW budget. The selected fuel forms are described in the following subsections, which include a description of current fuel element design and issues to be addressed by R&D.

### 3.1. Metallic and ceramic dispersions

A dispersion of TRU-Zr alloy fuel particles in a zirconium matrix has been proposed for application to a liquid metal-cooled transmuter. The TRU-Zr alloy is proposed based on experience with alloy fuel from the Integral Fast Reactor (IFR) program. Zirconium was selected as the alloying element because of the fabrication, reprocessing, and irradiation experience obtained with U-Zr and U-Pu-Zr fuel during the IFR program. Zirconium is the component of the U-Pu-Zr fuel alloy that raises the alloy solidus temperature and provides resistance against fuel-cladding chemical interaction [9] and dimensional stability during irradiation [10]. Cubic phases are stable in the Pu-Zr system at reactor temperatures for compositions of interest for ATW fuel. Although the behaviour of multi-element alloys is not known, TRU-Zr alloys may also form alloys with cubic crystal structures that are likely to be more stable against growth under irradiation.

If successful, this fuel form would combine the performance advantages of a dispersion fuel (including high-burnup potential, low fuel temperature, and retention of fission gas) with a low temperature fabrication route that will minimize the amount of volatile actinide loss during fabrication. Using metallic fuel particles in a metal matrix will also impart favorable neutronic safety characteristics to the fuel that are typical of metal fuel. These include high thermal expansion, which provides substantial negative reactivity feedback as fuel temperatures rise during transient events, and low heat capacity, which reduces the amount of stored energy in the core that must be dissipated in the event of a loss-of-flow accident. A metallic dispersion is expected to be compatible with pyrometallurgical processing concepts now being considered for recycle of ATW blanket fuel.

To minimize the potential for fuel-cladding chemical interaction, it is proposed that the compacted fuel "meat" (i.e., the Zr-matrix rod bearing the fuel particles) be placed inside of an enclosing Zr sheath. The Zr-enclosed fuel slug would then be co-extruded inside an enclosing cladding tube, which would ensure good thermal contact between the fuel slug and cladding. The fuel slug could be either a rod or a shorter "pellet." If irradiation testing indicates that the Zr-matrix fuel swells sufficiently to stress the co-extruded cladding, then a radial gap inside the cladding can be incorporated as a backup option. The Zr-enclosed fuel slug would then be bonded to the cladding using a liquid-metal bonding alloy similar to that used for the metal alloy fuel. The fuel would be clad in a ferritic-martensitic stainless steel, and the fuel rods would be loaded in hexagonal-shaped subassemblies similar to those used in Experiment Breeder Reactor-II and the Fast Flux Test Facility.

A (TRU)N (i.e., nitrides of TRU elements) fuel particle in a Zr matrix is proposed as an alternative to the metallic dispersion. This nitride offers a higher fuel-particle margin to melting combined with the thermal and neutronic advantages of the metal matrix. Nitride compounds are currently being investigated as fuels by researchers in Japan, Europe and Russia (including some work with non-fertile nitrides). Additional complexity associated with fabricating a nitride dispersion might be balanced against the advantages of mitigating problems of americium volatility (americium nitride is less volatile than americium metal).

Alternative matrix metals will be considered as required to provide chemical stability with the fuel phase, with an emphasis on identifying candidates based on compatibility with ATW recycle processes, acceptable irradiation performance characteristics, fabricability, high temperature strength, and compatibility with potential coolants. Another design variant that could be considered is a coating on the fuel particle, which may enhance the ability of the fuel particle to retain fission products and reduce interdiffusion of fuel constituents and matrix constituents. Fabrication of other design characteristics will also be considered, including means for placing a barrier the fuel slug and cladding.

A number of issues must be investigated to establish feasibility for this fuel type:

- What is the optimum particle loading that balances competing requirements for fabricability and low fuel swelling against the desire for a relatively high fissile density?
- Which fuel alloy composition best balances fissile density with fuel performance?
- Will fuel co-extruded with cladding (with no fuel-cladding gap) perform acceptably, or must a radial gap be incorporated into the design?
- If co-extruded cladding is used, will unacceptable stresses be induced in the cladding due to thermal expansion of the fuel upon startup and shutdown or accelerator transients?
- If a gap is required, what is a suitable bond metal?
- Can the fuel slugs be fabricated as long rods, or must shorter pellets be used?
- Can the fuel be fabricated using remotely-adaptable methods?
- Can the fuel be fabricated with sufficient fissile density to satisfy transmuter performance requirements or goals?

- Do the fuel particles remain intact during the irradiation, or do fuel constituents interdiffuse with the matrix?
- Is a matrix material other than Zr required?
- Does the fuel retain integrity and dimensions during irradiation?
- Do metal fuel particles melt during overpower transients, and what are the consequences?
- What is the behavior of high-burnup ceramic fuel particles during overpower transients?
- Will a Zr matrix dissolve at unacceptable rates upon contact with lead-bismuth coolant (e.g., during failed-fuel operation)?
- Is a barrier between the fuel “meat” and cladding required to limit fuel-cladding chemical interaction?
- Must fuel particles be coated to achieve desired performance?
- Can alternative matrix metals that are more compatible with lead-bismuth be identified?

### 3.2. Metal alloy

A TRU-Zr alloy fuel has been proposed as an alternative, based on experience with metal alloy fuel during the Integral Fast Reactor (IFR) program. This fuel form would consist of a slug of TRU-Zr alloy similar to the metallic fuel alloy envisioned for use in the dispersion fuel option, but with a higher Zr content.

The fuel rod design proposed is the same as that employed most recently for the IFR concept. A fuel smear density of 75% and a plenum-to-fuel volume ratio of 1.4 are proposed to accommodate radial swelling and fission gas release, respectively. Experience with metal fuel performance has demonstrated that accommodation of radial swelling and fission gas release from the fuel into the fuel-cladding plenum enables relatively high-burnup. The fuel would be clad in a ferritic-martensitic stainless steel, and fuel rods would be loaded in hexagonal-shaped subassemblies similar to those used in Experiment Breeder Reactor-II and the Fast Flux Test Facility.

Use of a liquid metal bond (selected to be compatible with fuel, cladding and coolant) would be required to ensure adequate heat transfer from the fuel to the cladding, in a manner similar to the Na bond used for previous fast reactor metal fuel designs. For the Na-cooled transmuter, Na would be used as the bond metal. It is not clear that Na is suitable as a bond metal for the lead-bismuth transmuter; interactions between Na and both lead and bismuth must be investigated to understand implications for failed-fuel operation.

Assessment of feasibility for this fuel will require investigation of the following:

- Can the fuel be fabricated without unacceptable loss of volatile transuranics?
- Are volatile actinide recovery techniques necessary and feasible?
- Is irradiation performance of TRU-Zr alloy fuel similar to that of U-Pu-Zr?
- What constituents diffuse to the fuel-cladding interface and is fuel-cladding chemical interdiffusion of consequence?
- Can fuel-cladding chemical interaction be mitigated?
- What is the dissolution behavior of TRU-Zr alloy in lead-bismuth, and what are the implications for failed fuel operation?
- Can a bond metal for the fuel-cladding gap be identified which will be compatible with fuel, cladding and lead-bismuth coolant.
- Can alloying elements be identified as potential alternatives to Zr?

### 3.3. Nitride

A non-fertile nitride fuel is proposed for application in fast-spectrum, liquid metal-cooled or gas-cooled transmuters based on prior experience with uranium nitride and mixed nitride fuels. Although, for the present time, a diluent in the fuel is assumed necessary, it is not clear which element(s) would be best suited for that purpose. (TRU,Zr)N is proposed initially, because Zr is compatible with the pyroprocess and would have small impact on neutron economy. An initial investigation of the Pu-Zr-N system indicates that PuN and ZrN are soluble, so that fabrication of an acceptable fuel mixture appears feasible. Although there is no irradiation performance experience with this fuel, non-fertile nitrides are being considered for actinide destruction in R&D programs in Japan and Europe. Therefore, international collaboration will be important for consideration of this fuel form.

The fuel rod designs proposed are those studied for (U,Pu)N fuels previously developed and tested in the U.S. Those designs consisted of pellets stacked and loaded into cladding tubes with either a gas or Na bond; gas-bonded and Na-bonded designs were each determined to perform acceptably. For ATW application, it is believed that a lead-bismuth bond could be used for a lead-bismuth-cooled transmuter, because Russian work indicates that (U,Pu)N is compatible with a lead-bismuth bond. A liquid metal bond provides for lower fuel operating temperatures, which was helpful to reduce fuel fracturing for (U,Pu)N. As stated for the other fast-spectrum fuel candidates, fuel rods would be loaded into hexagonal subassemblies in a manner consistent with U.S. fast reactor experience.

Neutron irradiation of the N-14 isotope leads to formation of C-14 through the $N^{14}(n,p)C^{14}$ reaction. For the purposes of this document, it will be assumed that enrichment of N-15 in the nitrogen will be employed for fabrication of the (TRU,Zr)N fuel. Therefore, N-15 collection and recycle techniques must be developed.

Assessment of feasibility for this fuel will require investigation of the following:

- Can (TRU,Zr)N be easily fabricated as a solid solution in the desired composition using feed material from the ATW recycle process?
- Will (TRU,Zr)N retain fission gas and resist cracking at moderate temperatures in a manner similar to that of (U,Pu)N?
- How do TRU nitrides behave in the fuel matrix while under irradiation?
- How severely do nitride fuels fragment during overpower transients, and is the fragmentation of any consequence?
- Do any issues emerge for failed fuel operation (e.g., due to interaction of (TRU,Zr)N with lead-bismuth)?
- Is a gas bond or liquid metal bond preferable?
- Is N-15 enrichment required to avoid C-14 generation issues?

### 3.4. TRISO-coated particles

Recently proposed concepts for thermal-spectrum, gas-cooled systems (critical or subcritical) call for the use of TRISO-coated oxides of uranium and/or plutonium. “TRISO” is an acronym that designates the coating configuration around the central fuel particle. Specifically, the fuel kernel is surrounded by a lower-density, pyrolytic carbon “buffer layer”, then a dense pyrolytic carbon layer and a silicon carbide layer, with an pyrolytic carbon layer. The coated fuel kernels are dispersed in graphite compacts, which are placed in arrays in graphite blocks, referred to as fuel elements. These fuels appear capable of sustaining very high burnup values, but experience has demonstrated that proper fuel design and fabrication quality are essential for good irradiation performance. Some work performed by General Atomics and ORNL in the early 1970s indicates that TRISO-coated $PuO_2$ fuel kernels can achieve burnup values as high as 75at.% [11].

Experience with the New Production Reactor (NPR) program conducted in the U.S. during the late 1980's and 1990's indicates that uranium oxycarbide fuel kernels performed better than $UO_2$ fuel kernels. Furthermore, considering that oxygen stoichiometry was determined to be an important parameter for achieving high-burnup performance of $PuO_2$ kernels, the multiple valences of TRU oxides might have some effect on the fabrication or performance of those fuel kernels. Use of oxycarbide fuel kernels would eliminate the variance of oxidation states for the TRU elements, perhaps simplifying fabrication or aiding fuel performance. Therefore, it is proposed that TRU oxycarbide [(TRU)OC] be considered as a fuel kernel option for thermal-spectrum gas-cooled transmuter fuel in addition to TRU oxide $(TRU_x)O_y$. Programs in the U.S., Russia, South Africa and France are currently considering gas-cooled reactor systems and/or accelerator-driven systems for a variety of applications; therefore, potential exists for effective international collaboration in developing this fuel form.

Current proposals from General Atomics, an industrial developer and designer of thermal gas-cooled reactors, call for using SiC layers in the TRISO coatings. However, recent work with ZrC coatings in Japan indicates that the ZrC coatings may have better resistance to failure and to palladium corrosion than the SiC coatings. Therefore, ZrC will be considered as an alternative to SiC as a pressure-restraining coating.

Fabrication and irradiation performance for this fuel type must be demonstrated, using bench-top scale samples, to establish feasibility. There is no known experience with TRU oxycarbide TRISO-coated fuel, so fabrication techniques must be developed and irradiation performance investigated. Some specific feasibility issues include:

- Can transuranic fuel particles be synthesized and coated with acceptable loss (or no loss) of volatile actinides?
- Is either $(TRU_x)O_y$ or TRU(OC) a superior fuel kernel material?
- Do the multiple oxide valences of TRU elements effect fabrication or performance of $(TRU_x)O_y$ fuel kernels?
- Will TRISO-coated TRU fuel perform with acceptable failure rates?
- Is SiC the best pressure-bearing coating, or is ZrC better?
- Can the bonding strength between coating layers and the layer thickness be optimized for better high-burnup performance?
- What is the overpower transient response of high-burnup TRISO-coated particles?
- Is post-failure behavior of TRISO-coated TRU fuel acceptable?

## 4. R&D DESCRIPTION

The R&D described in this plan is intended to determine feasibility of the proposed fuel concepts in support of ATW technology decisions. General feasibility issues are best categorized into four areas of R&D:

- Fabrication;
- Property measurement and out-of-pile experiments;
- Irradiation performance;
- Modeling of behavior and phenomena.

### 4.1. Fabrication development

Fabrication development activities will address the two primary needs of the ATW program. These are:

1) fabrication of specimens suitable for screening irradiation tests in the ATR and for fast flux irradiation testing, and
2) identification and development of fabrication processes that can be successfully deployed for large scale ATW fuel manufacturing.

Initially, small representative fuel specimens will be fabricated for use in ATR irradiation testing. Fabrication techniques employed to make these specimens may be chosen for expedience rather than being representative of a large-scale fabrication process, facilitating an aggressive irradiation testing schedule. Experience gained during fabrication of small specimens will be applied to production of more prototypic fuel rods for fast flux irradiation testing and to the conceptual design of a large-scale fabrication process.

An important consideration for all fuel types is volatilization of americium, leading to loss or redistribution during fabrication. It is partially for this reason that dispersion fuels were chosen as a candidate fuel form. Dispersion fuels are fabricated using powder metallurgical processes that are well developed and currently used for the production of research reactor fuel elements. Powder metallurgical processes allow for lower fabrication temperatures than melt processing, providing an opportunity to reduce americium volatilization during fabrication. These same techniques could also be applied to the production of monolithic metal fuel. The advantages of powder processing come at the expense of the need to remotely handle powder. This issue is of equal or greater concern for a nitride pellet fuel.

Fuel particles required for a dispersion can be fabricated by any number of methods. However, the metal feed from the reference electrorefining process fits well with both hydride-dehydride (-nitride) and mechanical powder production techniques. The ability to perform the hydride-dehydride process may depend on the purity of actinide metal feed. Thermo-mechanical consolidation of fuel meat compacts into clad, rod-shaped dispersion fuels by swaging, drawing, and extrusion will be evaluated. Methods to characterize the integrity of the core to clad bond will be developed.

At present, little is known about non-fertile nitride fuels. The R&D program for this fuel type will include fabrication of (TRU,Zr)N pellets and characterization of their microstructures, as well as preparation of specimens for irradiation testing. Both hydride/dehydride/nitride and carbothermic reduction processes are possible routes to production of nitride fuel powder. The inert element might be incorporated in an alloy or oxide precursor, or by solid state reaction of nitride powders to produce either homogeneous or heterogeneous fuel microstructures. Powder will likely be sintered using conventional techniques to produce pellets appropriate for irradiation testing.

### 4.2. Property measurements and out-of-pile experiments

Given that the non-fertile compositions proposed for use as an ATW blanket fuel have only recently begun to be seriously considered as nuclear fuels, little to no experimental data exist on the thermophysical properties of most of these materials. During the first phase of the ATW program undertaken to demonstrate the feasibility of the proposed fuel forms, an experimental program has been initiated to characterized the candidate fuels and measure important material properties. This effort is being undertaken to support the analysis and design of upcoming irradiation experiments, support fuel modeling efforts and aid in the ultimate evaluation and selection of the final ATW blanket fuel. In general, the property testing program will include investigation of: the thermal conductivity, specific heat and thermal expansion characteristics of each fuel composition; the thermal equilibria for each fuel type; and the materials compatibility of each fuel-cladding-coolant system. In each case, the existing literature is reviewed and evaluated, followed by actual experimental measurements where needed.

For both the metallic and ceramic dispersion fuels, properties to be measured or characterized include:

- Thermal conductivity and specific heat;
- Thermal expansion;
- Phase equilibria and interdiffusion behavior of TRU-Zr and matrix alloy system, including melting temperature determination;
- Compatibility and dissolution kinetics of dispersion fuel and individual fuel constituents in Pb-Bi eutectic; and
- Fuel particle-cladding interdiffusion behavior.

For the metal alloy candidate fuel, properties to be measured or characterized include:

- Thermal conductivity and specific heat;
- Interdiffusion behavior of fuel alloy constituents;
- Phase stability and presence in as-fabricated fuel alloy and evolution with time at elevated temperatures;
- Fuel-cladding interdiffusion behavior; and
- Compatibility and dissolution kinetics of the fuel alloy in Pb-Bi eutectic and bond metals.

For the nitride candidate fuel, properties to be measured or characterized include:

- Thermal conductivity and specific heat;
- High-temperature dissociation behavior;
- Phase stability and presence in as-fabricated fuel pellet and evolution with time at elevated temperatures;
- Fuel-cladding interdiffusion behavior; and
- Compatibility with cladding and with Pb-Bi eutectic.

For the TRISO-coated particle fuel option, properties of non-fertile, TRISO-coated fuel particles in graphite compacts may be more readily determined from data from similar fuels (e.g., from well characterized TRISO-coated UOC fuel particles) than for the other fuel options. New property measurements for TRU oxycarbides and oxides of ATW–relevant compositions would be required for the following:

- Thermal conductivity and specific heat; and
- High temperature phase stability.

### 4.3. Irradiation performance

Irradiation testing needs are very similar for each of the fuel types under consideration. Since no irradiation performance data exist for these non-fertile, plutonium-based and minor actinide-laced fuel forms, the approach undertaken consists of initially simple steady-state irradiation tests to identify potential irradiation performance behavior that may limit use of the fuels; these tests will serve as quick screening tests performed simultaneously on all the candidate fuel forms. Follow-on irradiation tests will narrow the variety of fuels being tested and demonstrate behavior characteristics under expected nominal irradiation conditions.

The initial steady state irradiation tests will be performed in the Advanced Test Reactor (ATR), located at the Idaho National Engineering and Environmental Laboratory. Although the neutronic conditions available in the ATR will not be prototypic of fast-spectrum transmuters, prototypic fission rates and fuel temperatures can be attained. This will allow a timely and qualitative assessment of key phenomena such as fuel swelling, fission gas release, and fuel constituent interdiffusion. Some phenomena cannot be evaluated in a thermal spectrum test, such as the matrix swelling and creep behavior of dispersion fuels and cladding performance for fuel designs intended for fast-spectrum transmuters. Therefore, subsequent irradiation tests will be conducted at conditions that are representative of those expected in particular transmuter designs, which will require irradiation in fast-spectrum test reactors; the identity of the fast-spectrum test reactor to be used remains unknown. Other tests may be required to better investigate specific phenomena, as identified in initial irradiation tests or out-of-pile characterization, such as irradiations at higher-than-nominal temperatures or power levels. ATR can provide prototypic conditions for irradiation of the TRISO-coated particle fuel proposed for use in a thermal-spectrum, gas-cooled transmuter. Eventually, transient testing of both as-fabricated and pre-irradiated fuel samples could be performed in the Transient Reactor Test Facility (TREAT) located at the ANL-West site, depending on the availability of this facility.

Post-irradiation examination of domestically irradiated test fuel will be performed at hot cell facilities such as the Hot Fuel Examination Facility (HFEF) at ANL-West or the Alpha-Gamma Hot Cell Facility (AGHCF) at ANL-East. Examination of test fuel that might be irradiated in non-U.S. fast flux test reactors would likely be performed in the country where the test reactor resides, dependent upon negotiation.

Currently, at least three irradiation tests are envisioned for the FY'01 to FY'05 time period. Irradiation test ATW-1 has as its objective an assessment of irradiation performance characteristics of fuel forms proposed for Pb-Bi-cooled and Na-cooled fast-spectrum transmuters. ATW-1 will include samples of the dispersion, metal alloy, and nitride fuels irradiated in the ATR. This initial test is intended to be a quick-turnaround, inexpensive screening test including a large variety of fuel samples.

Irradiation test ATW-2 has as its objective an assessment of irradiation performance characteristics of non-fertile TRISO-coated fuel compacts and particles under conditions prototypic of a gas-cooled, thermal-spectrum transmuter. ATW-2 will incorporate fuel particles, both loose and in graphite compacts, fabricated with ATW-representative TRU contents, irradiated in the ATR. SiC and ZrC coatings will be employed and various coating thicknesses will be evaluated.

Irradiation test ATW-3 has as its objective an assessment of irradiation performance characteristics of fuel forms proposed for Pb-Bi-cooled and Na-cooled fast-spectrum transmuters under representative spectrum and temperature conditions. ATW-3 will incorporate prototypic fuel rods of metallic and ceramic dispersions, metal alloys, and/or nitrides, depending on which of those forms emerge from ATW-1 and out-of-pile experiments as promising candidates, with compositions and fabrication variables to be determined. It will be desirable to irradiate samples with representative quantities of minor actinides, as well as samples that are primarily Pu-bearing, to allow reference to the samples from ATW-1 expected to have low quantities of minor actinides. The irradiation test will be performed in a fast-spectrum test reactor.

### 4.4. Modeling of behavior and phenomena

Modeling to support the ATW blanket fuel development effort will focus on applying (or developing, as needed) mechanistic models of key phenomena that are observed in irradiation tests. During the initial feasibility phase of the program, modeling work will emphasize developing an understanding of observed phenomena; gaining a predictive capability through development of mechanistic models will take place outside the framework of an all-encompassing fuel performance code, since such a code is generally quite fuel system-specific.

Initially, existing and applicable fuel behavior data, models or correlations for each fuel type will be identified from the literature. Although it is most desirable to employ models based on mechanistic understanding of phenomena, development of such models can require considerable effort over long periods of time. Therefore, a set of empirical correlations that can be used in initial fuel performance calculations will be prepared, and existing models will be used as appropriate. It is anticipated that empirical correlations and models based on uranium fuel analogues (or Pu, where available) to the ATW fuel systems will be used initially, with subsequent work to refine these models to describe non-fertile TRU fuel systems. Finally, longer-term work will begin to develop mechanistic models for these fuel phenomena from theoretical considerations, data from the literature and results of PIE from the irradiation test program.

For TRISO-coated particle fuel, the models developed by General Atomics and the Idaho National Engineering and Environmental Laboratory as part of the New Production Reactor program will be employed. Those models are reasonably mature and will provide a suitable beginning to ATW-related model development.

## 5. EXPECTED RESULTS

At the end of FY’05, it is expected that feasibility will be established for one reference fuel form and one alternate (resources permitting) for each ATW transmuter system remaining under consideration. Establishing feasibility will require that acceptable irradiation performance be demonstrated (at least to moderate burnup values) under nominal, representative conditions and that other potential problems have reasonable engineered solutions to allow implementation (e.g., cladding liners to address incompatibility between fuel or target materials and cladding). In addition, key property measurements will be documented, having sufficient quality to support safety analyses for more extensive irradiation testing and licensing of a potential test transmuter unit.

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