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**THORIA-BASED CERMET NUCLEAR FUEL: CERMET FABRICATION AND
BEHAVIOR ESTIMATES**

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ABSTRACT

Cermet nuclear fuels have been demonstrated to have significant potential to enhance fuel performance because of low internal fuel temperatures and low stored energy. The combination of these benefits with the inherent proliferation resistance, high burnup capability, and favorable neutronic properties of the thorium fuel cycle produces intriguing options for advanced nuclear fuel cycles. This paper describes aspects of a Nuclear Energy Research Initiative (NERI) project with two primary goals: (1) evaluate the feasibility of implementing the thorium fuel cycle in existing or advanced reactors using a zirconium-matrix cermet fuel, and (2) develop enabling technologies required for the economic application of this new fuel form.

Critical elements in the demonstration of this new fuel form include developing low-cost fabrication methods and characterizing the cermet properties and important performance parameters. A powder-in-tube drawing and heat treatment process is being evaluated as an alternative to hot extrusion. In this method, zirconium metal and ceramic microspheres are mixed, poured into a Zircaloy shell, and compacted into simulated fuel pins. Important processing variables being

evaluated include the amount of compaction required to achieve a desired matrix density and the inter-drawing thermal treatment temperature required to achieve adequate matrix fusion and grain growth.

NOMENCLATURE

ANL	Argonne National Laboratory
ID	Drawing Sheath Inner Diameter
NERI	Nuclear Energy Research Initiative
PWR	Pressurized Water Reactor
OD	Drawing Sheath Outside Diameter
T.D.	Theoretical Density

INTRODUCTION OF CERMET FUEL CONCEPT

This paper is one of three in this proceedings describing a 1999 NERI project designed to develop the potential and demonstrate the feasibility of a (Th,U)O₂ cermet fuel [1,2]. The basic fuel concept and experimental cermet fabrication method will be described below, whereas the fundamental nuclear and thermal modeling [1] and the developmental fuel microsphere fabrication method [2] will be described in the other papers.

fuels research noted that the French atomic energy agency (CEA) has demonstrated a fuel discharge burnup in a cermet test rod of ~136 GWD/MT with minimal fission gas release, good irradiation stability, low stored energy, and outstanding thermal conductivity [4]. Therefore, the primary innovation of this fuel cycle concept is to bring these two promising concepts together in a new way.

BACKGROUND ON THE THORIUM FUEL CYCLE

Various approaches to thorium-based fuel cycles have been advanced since the advent of nuclear power in the mid-1900's [5]. During the Atoms for Peace Program (1955–1975), thorium was predicted to be equal to uranium in its importance to the future of nuclear energy [6]. This prediction was based on optimistic assumptions regarding the growth of the nuclear power industry, but thorium still has considerable potential as a nuclear fuel. Some of the more compelling benefits to the thorium fuel cycle include high actinide burnup, improved proliferation resistance, and improved waste characteristics [6,7].

Thorium is considerably more abundant than uranium. Concentration estimates for the Earth's crust are between 7 and 10 ppm for thorium and ~2 ppm for uranium (compared to ~16 ppm for lead), so there is 3 to 5 times more natural thorium than uranium [8,9]. Thorium is present in monazite, zircon, and other natural ores, and thorium nitrate is a common by product from the recovery of the lanthanide elements (e.g., La, Ce, and Nd). As a result, the commercial thorium supply tends to exceed the market demand for lighting, welding electrodes, and other applications [9]. In addition, the National Defense Stockpile of the United States was authorized in 1994 to reduce its thorium nitrate inventory by ~3 million kilograms (~6.5 million pounds) [9]. Therefore, there is a significant opportunity to transform an existing waste stream into an energy resource if an appropriate fuel cycle can be developed.

Natural thorium is primarily present as the isotope Th-232. Very minor traces of Th-228 (from Th-232 decay), Th-231 and Th-227 (from U-235 decay), and Th-234 (from U-238 decay) are detectable, but they are present on the order of 1 part in $\sim 10^{10}$ [5,8]. Therefore, Th-232 is the only isotope relevant to the startup of a thorium fuel cycle, and thorium "enrichment" is a meaningless concept. Thorium-232 is not fissile, that is, it does not readily fission upon the capture of a thermal neutron in the nucleus. However, it is fertile because a captured neutron initiates a radioactive decay chain that produces the fissile U-233.

The transmutation of Th-232 into U-233 in a thermal reactor is what makes thorium useful as a potential energy supply. Uranium-233 has a thermal fission cross section comparable to that of U-235, and it has a relatively high neutron yield per fission. Therefore, the Th-232/U-233 fuel

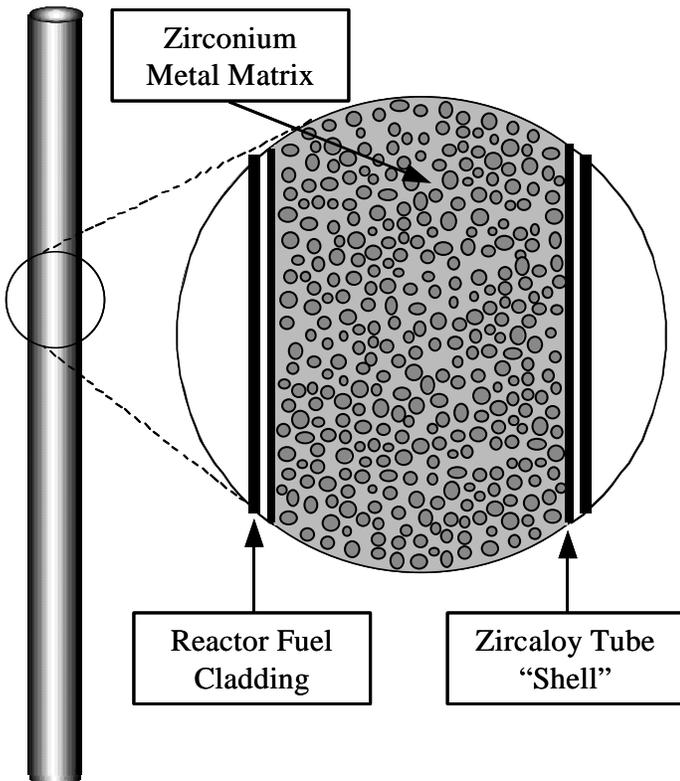


Figure 1. Concept Sketch for (Th,U)O₂ Dispersion Fuel Pin.

The principal goal of this project is to demonstrate the feasibility of a metal-matrix dispersion, or cermet, fuel comprising (Th,U)O₂ microspheres in a zirconium matrix that can achieve high burnup and subsequently be directly disposed as nuclear waste. The potential benefits that may be gained with this fuel include high actinide burnup, inherent proliferation resistance, improved irradiation stability due to low internal fuel temperatures, low fuel failure rate, and minimal waste treatment. The cermet fuel concept is shown schematically above in Fig. 1. The fuel "meat" is composed of a fine dispersion of (Th,U)O₂ microspheres that have a theoretical density between 70 and 99% and a uranium enrichment below 20% U-235. Nominal values for the microsphere diameter, ThO₂-to-UO₂ ratio, fuel-to-matrix ratio, and U-235 enrichment were selected as ~50 μm, 50:50, 50:50, and ~19.5%, respectively, to provide guidance for the calculational and experimental activities carried out within the project [3].

As described in the following section, the potential for high actinide burnup is inherent to the thorium fuel cycle because of the buildup of U-233 that follows neutron capture in Th-232. In addition, cermet fuels have demonstrated potential for use in a high-burnup fuel cycle. A recent summary of international

system is capable of sustaining a nuclear chain reaction and driving a reactor system [5]. On the other hand, without a naturally occurring fissile thorium isotope, the initial loading of a thorium-based fuel system requires a fissile driver isotope, or “match” [6] to provide the initial reactivity for the fuel. It is also established that Th-232 is not competitive with U-238 as a fast reactor fuel because its fast fission cross section has a higher energy threshold and a smaller value, its fast neutron capture cross section is greater, and its fast fission neutron yield is lower [5].

Previous reactor designs have proposed and, in some cases, demonstrated fuel design approaches that have included 1) a mixture of fertile thorium oxide (ThO_2) and highly enriched uranium carbide (UC) microspheres in a gas-cooled, graphite-moderated reactor, 2) monolithic $(\text{Th,U})\text{O}_2$ fuel pellets with a U-235 enrichment as high as 95% in water-cooled and moderated reactors, and 3) light water breeding reactors which involved breeding in a thorium blanket, recovering the U-233, and ultimately replacing U-235 with U-233 as the fissile driver [5]. Radkowski and Galperin [7] have proposed the “Radkowski thorium fuel” (RTF) system, which would make use of the “seed-blanket” fuel concept demonstrated in the Shippingport reactor from the mid-1970s up to 1982. For the RTF concept, the seed would be UO_2 enriched to <20%, and the blanket would be ThO_2 with a small amount of UO_2 enriched to <20%. The enriched UO_2 in the RTF blanket prevents the chemical separation of pure U-233, which was a proliferation concern in early thorium fuel cycles, but the blanket is designed to be reprocessed after 10 years [7].

In 1999, the United States Department of Energy NERI program initiated the development of two new thorium fuel types: 1) a monolithic ceramic fuel form that is analogous to existing UO_2 or MOX ceramic fuel pellets [10], and 2) the zirconium matrix cermet fuel form described here [1-3]. Both of these new fuel types move away from breeding blankets and the recycling of U-233 by blending the fissile and fertile materials into a single fuel form that will be irradiated to high burnup in a once-through cycle and disposed without processing.

This blending of the actinide oxides helps to improve the proliferation resistant qualities of this fuel. The use of mixed oxides prohibits the direct chemical separation of pure U-233 or Pu-239 without subsequent isotope separation. Recall that the U-233 and Pu-239 isotopes are generated by the conversion of Th-232 and U-238, respectively. The direct chemical separation of pure U-233 is not possible here because it is intimately mixed with U-235 and U-238 from the UO_2 feed. The direct chemical separation of Pu-239, or even low-grade Pu-239, is complicated by a significant quantity of Pu-238 that is bred into the fuel at higher burnups.

CERMET FUEL BEHAVIOR ESTIMATES

Previous irradiation experience with stainless steel matrix cermet fuel has been reported for uranium dioxide, UO_2 , and mixed plutonium-uranium dioxide, $(\text{Pu,U})\text{O}_2$ [11,12]. The referenced fuels were irradiated in the Advanced Test Reactor in rod and plate geometries. Irradiation-induced swelling and matrix rupture were serious problems when the starting fuel particles were dense (~95% T.D.). However, low-density (70 to 80% T.D.) fuel particles were found to mitigate swelling because of their ability to accommodate accumulation of solid and gaseous fission products [12]. Similar, but only remotely related, data are available from recent ceramic-matrix dispersion fuel irradiations in France [13,14]. The thermal conductivity and physical behavior of the inert ceramic matrix are critical to performance. In addition, the ceramic dispersion fuel particle size was observed to have a marked effect on matrix durability (e.g., microsphere fuel with diameters below ~100 μm generated major cracks in an MgAl_2O_4 spinel matrix, but macrosphere fuel on the order of 100 to 300 μm had minimal effect on the matrix [13]). It is expected that the zirconium matrix being considered here will behave comparably to stainless steel with a relatively low centerline temperature and durable physical properties such that fuel particle size variations may be considered without the fuel performance impact seen in ceramic matrix fuels.

For a once-through fuel cycle, the cermet fuel is designed to be disposed of after irradiation without processing. If the zirconium matrix is fabricated with a high theoretical density (i.e., >95% T.D.), it will combine with the Zircaloy shell and cladding shown in Fig. 1 to form an effective waste containment system. Since this is a thorium-based cermet, an additional waste disposal benefit arises because ThO_2 and $(\text{Th,U})\text{O}_2$ are known to be more stable than UO_2 in oxidizing environments such as groundwater or hot air. This occurs because Th does not have higher valence states available for further oxidation of the fuel. (On the other hand, fuel recycling would enable even greater resource utilization with higher actinide consumption and lower high-level waste volume. However, consideration of fuel recycling options are beyond the scope of this present project.)

The high thermal conductivity of the zirconium matrix greatly enhances heat removal; thus, the centerline fuel temperature will be significantly lower than that of a monolithic ceramic fuel pin. This point is important because the lower overall fuel temperature reduces the performance-limiting impact of fission product migration, fuel swelling, and other in-reactor phenomena. In addition, the high-conductivity matrix results in a low stored energy content due to the low internal fuel temperatures, which will contribute to severe accident mitigation and a low fuel failure rate.

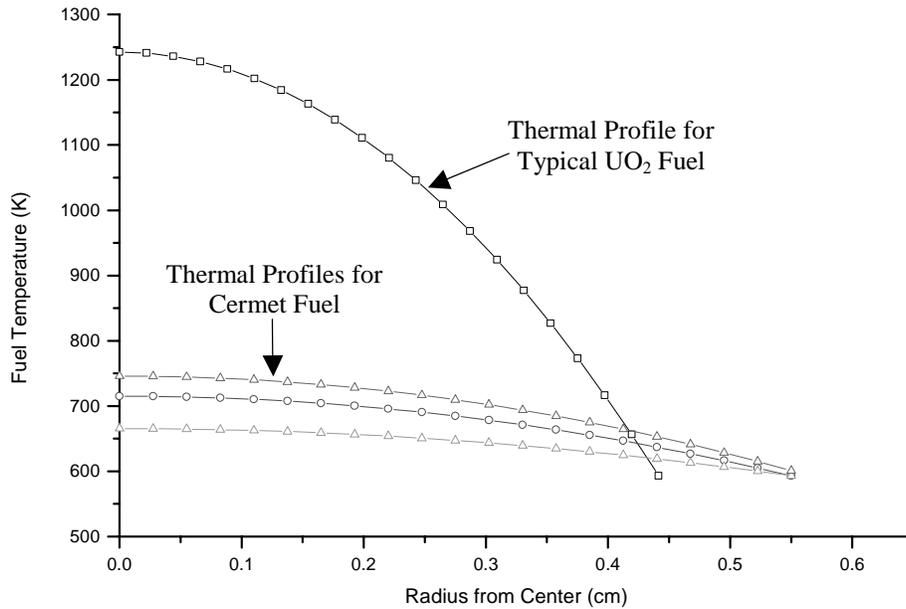


Figure 2. Calculated Radial Temperature Distributions for Cermet Fuel and Ceramic PWR Fuel using the Effective Conductivity Model from Downar, *et al.* [1] (Top: 19.5% Enriched UO_2 in Zr; Middle: Nominal $(\text{Th,U})\text{O}_2$ in Zr; Bottom: 15% Enriched UO_2 in Zr)

A detailed thermal model has been developed to simulate the behavior of this cermet fuel. The model is based on the effective conductivity across an ideal interface, accounting for the Kapitza contact resistance; a detailed description of this model is provided by Downar *et al.* [1] Figure 2 shows calculated radial temperature profiles for typical UO_2 fuel compared with some cermet fuel examples. The cermet centerline temperatures are notably low, on the order of $\sim 100^\circ\text{C}$ greater than the cladding surface. In addition to the beneficial effects, this expected outstanding thermal behavior will have on the cermet fuel performance, we have also shown that it has a positive influence on the neutronics fuel cycle.

Our primary neutronics calculations are being performed using the commercial lattice physics code HELIOS from Studsvik/Scandpower [15] and more detail is again provided by Downar *et al.*[1] Benchmarking calculations with Monte Carlo burnup codes were used to demonstrate that HELIOS was effective for modeling thorium fuel lattices [16]. Fuel pin/assembly designs have been established and core depletion, fuel cycle depletion, core design, and economic calculations are being made.

The initial baseline simulations, made with HELIOS, used the nominal fuel description (i.e., $\text{ThO}_2:\text{UO}_2 = 1:1$, fuel:matrix = 1:1, and enrichment = 19.5% U-235) to generate

comparisons with typical PWR fuels. Figure 3 shows data from one early set of calculations where a burnup of ~ 80 MWd/kgHM was found to be readily achieved in an unmodified LWR reactor fuel assembly. (A positive by product of high burnup is a reduction in the overall fuel cycle cost. Even though the direct fuel fabrication costs may initially be higher for thorium fuel, especially at first, spreading the cost over a longer time should mitigate the increase in fuel costs.)

Following these initial simulations, the fuel conversion ratio was improved by hardening the neutron spectrum. Specifically, a tight-pitch hexagonal lattice was benchmarked and studied within the context of a boiling water reactor (BWR) core. Because a BWR is operated at a lower pressure than a pressurized water reactor, there is substantial boiling in the core, and therefore a natural hardening of the neutron spectrum occurs with a corresponding increase in the fuel conversion ratio. Using our nominal $(\text{Th,U})\text{O}_2$ cermet in a typical BWR, the moderator-to-fuel area was reduced and the conversion ratio increased from ~ 0.6 to more than 0.9, which resulted in a substantial increase in the fuel burnup. (A minimum moderator-to-fuel ratio was found at which a negative void coefficient could be insured.) Work in this area is now focussed on applying these unique advantages toward high-conversion reactor concepts, including BWR, PWR, Reduced Moderator Water Reactor, and Supercritical Light Water Reactor systems [1].

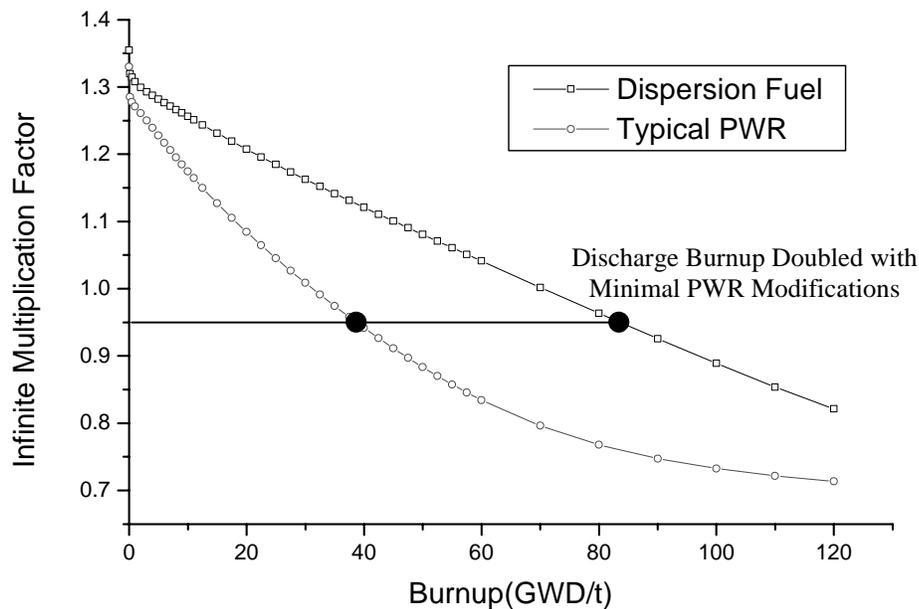


Figure 3. Multiplication Factor vs. Burnup of a Cermet Pin Cell and a Typical PWR Pin Cell Calculated using HELIOS [1,3].

CERMET FABRICATION METHOD

A powder-in-tube drawing technique with interim heat treatment is being developed to fabricate fuel pins comprising (Th,U)O₂ microspheres dispersed in a zirconium matrix and contained within a thin alloy sheath. The following requirements were established as guides in this pursuit: 1) the metal matrix must have a high density; 2) the metal matrix must effectively encapsulate the oxide fuel particles to isolate fission products during burnup; 3) the fabrication method developed must be amenable to fabricate full-scale fuel rods; and 4) the fabrication method must have the potential for low-cost implementation. The process involves mixing metal and ceramic powders, filling the alloy tube sheath, drawing the rod to compact the metal matrix, annealing the drawn rod, and repeating the draw-anneal sequence until the desired compaction is achieved. Processing variables and equipment specifications are being established for powder mixing of metals and microspheres, fuel pin drawing and annealing, and microsphere particle coating. In the initial drawing experiments, simulated fuel pins were fabricated with the assistance of ARi Industries (Addison, IL).

Initial fabrication demonstrations were completed using no radioactive surrogate materials to establish mechanical processing parameters and to evaluate the capabilities of this method for metal matrix compaction. Figure 4 illustrates this method by showing the simple powder filling method and a

schematic for multiple-sample drawing experiments. For lab-scale experiments, the oxide and metal powders are dry-loaded into alloy drawing tubes and vibratory-packed to obtain a green density between 40 and 50% theoretical density (T.D.). The powder-containing tube is then drawn through a reducing die to reduce the pin's outer diameter and thus compact the powders within. The internal microstructure is stabilized through a post-drawing anneal to remove strain hardening, facilitate sintering and interfacial bonding, and promote additional densification. Multiple drawing/heat-treatment cycles may be used with sequentially decreasing die sizes to achieve complete densification.

Powder Mixing Methods

Two methods are being examined for combining the oxide ceramic microspheres with the zirconium metal precursors: 1) simple dry mixing and 2) wet vibratory milling. Both mixing methods appear effective and are simple and scalable.

For simple dry mixing, particle size distributions of these materials must be adjusted to prevent segregation. As a starting point for this parameter, we are keeping the particle size distributions as similar as possible. For handling purposes, only powder materials with a mean particle diameter greater than ~40 μm are being used. Using relatively coarse precursors enables the flow behavior necessary to achieve sufficient dry mixing. This unit operation is simple, fast, and does not involve any intermediate steps.

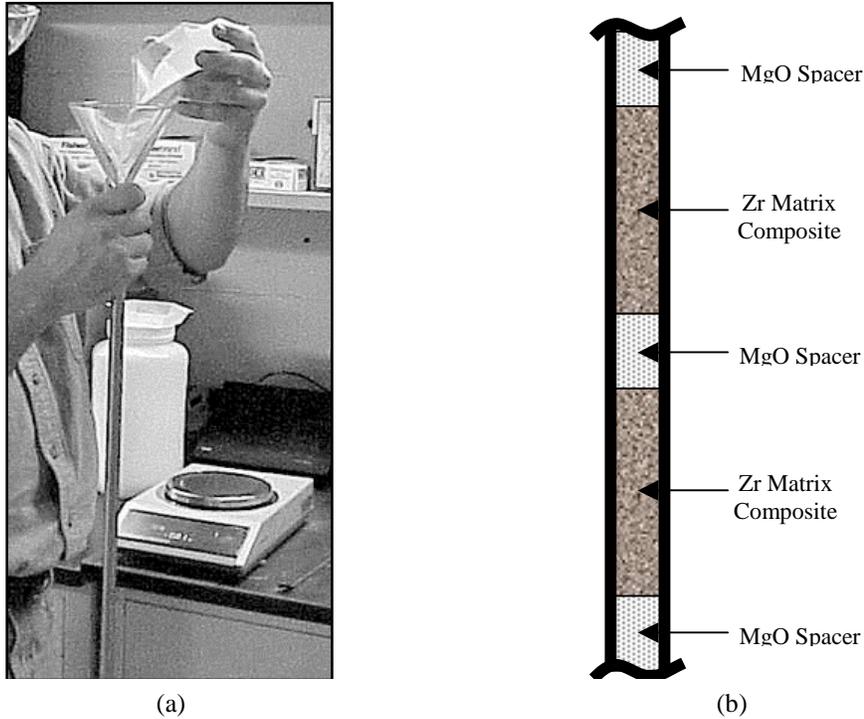


Figure 4. Details of the Powder-in-Tube Fabrication Method Showing (a) the Simple Powder Filling Method for Lab-Scale Experiments and (b) the Multiple-Sample Packing Scheme.

For wet vibratory milling, relatively coarse materials are again being used to enable easy handling. Unlike the dry method, additional processing steps include the addition of a fluid (e.g., water) and post-mill removal of this fluid. This makes the processing time longer and increases opportunity for impurity contamination in the metal matrix. On the other hand, the primary advantage of wet milling is the introduction of much more mechanical energy into the powder mixture. This can enhance particle coating, as discussed below.

Coating Methods for Oxide Microspheres

There are thermal-property and thermodynamic motivations for coating the microspheres. Thermally, the individual oxide fuel particles will require intimate and complete contact with the metallic matrix to minimize overheating the oxide particles and to achieve the maximum thermal benefit of the metal matrix. Thermodynamically, the potential exists for uranium dioxide-zirconium metal chemical interactions that may affect fuel performance.

The desired metallic coating is being designed to ensure that each granule or bead of fuel is isolated within the zirconium matrix to minimize fuel operating temperature. The initial intent is to evaluate a simple Zr coating that may be

oxidized to provide continuous surface encapsulation. Mechanical, liquid, and vapor deposition are being evaluated.

Figure 5 shows a representative mechanical coating of zirconium on zirconium oxide achieved using the wet vibratory milling method. This operation involves combining +325 mesh (~44 μm) zirconium metal and ZrO₂ beads in water in a high-

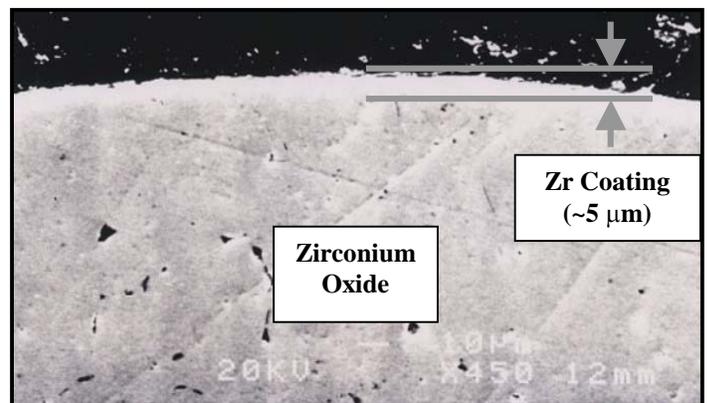


Figure 5. (Secondary Electron Image (450X) of Thin Zirconium Metal Coating on a ZrO₂ Particle after Mechanical Milling for 16 Hours.

density polyethylene bottle that is shaken vigorously to provide aggressive mixing/milling. Zirconium metal is relatively soft compared to the hard oxide beads. The resulting coating of Zr on ZrO_2 was very dense and had a uniform thickness of $\sim 5 \mu\text{m}$ after 16 hours of vibratory milling. (The $(\text{Th,U})O_2$ microspheres being fabricated at Purdue University [2] should be of comparable hardness to the ZrO_2 beads used in this test.)

Simulated Fuel Pin Fabrication

Initial cermet test rods were drawn using Type 316 stainless steel and Zircaloy-4 sheaths. (The sheath material used in the final fuel form will be Zircaloy or a comparable nuclear-grade cladding material.) The experiments using stainless steel (OD = 1.275 cm, ID = 0.959 cm, initial wall thickness = 0.158 cm) used ~ 5 feet long tubes, and multiple samples were generated within a single draw-tube by separating regions of cermet powder with granulated magnesium oxide (Fig. 4b). In all cases, the filled sheath was vibratory packed to achieve a green powder density between 40 and 50% T.D.

The powder mixtures studied thus far have comprised variable volume fractions of zirconium metal ($\sim 44 \mu\text{m}$ nominal particle size) combined with either nonspherical magnesium titanate (~ 44 , ~ 125 , and $\sim 250 \mu\text{m}$ nominal particle size $2\text{MgO}\cdot 3\text{TiO}_2$) or coarse zirconium oxide beads ($\sim 1 \text{mm}$ nominal particle size ZrO_2). Multiple sample compositions and drawing schedules were examined to study the effects of zirconium content, oxide particle size, mixing method, and compaction. Samples were fabricated with zirconium levels ranging from 5 to 67 volume percent (10 to 80 wt%) to examine the metal matrix densification efficiency and determine processing limits. The three sizes of magnesium titanate were tested to explore particle size effects, and it was observed that even the finest powders readily flow into the tubes during the loading step.

The drawn tubes were sectioned and characterized. Electrical resistance measurements between the center of a section and the sheath wall showed that an “electrically” continuous matrix is established at zirconium fractions of ~ 25 volume percent ($\sim 40 \text{ wt\% Zr}$) and higher. While this connectivity demonstrates that the drawing operation successfully bonds the zirconium metal matrix throughout the composite and to the tube wall, it does not imply that the oxide particles are well encapsulated. Metallurgical characterization of the magnesium titanate-containing experiments revealed that oxide agglomeration prevented effective encapsulation. However, the precursor powder mixing methods were found to be successful at dispersing the powders uniformly. It was also evident that the zirconium metal powders were effectively fused to form a dense, continuous matrix. The use of coated $(\text{Th,U})O_2$ microspheres (see previous section) is expected to alleviate this agglomeration issue.

To quantify the physical changes occurring in the sheath during drawing, stainless steel tubes were processed to various



Figure 6. Drawn Powder-in-Tube Test Specimens with Different Reductions in Cross-Sectional Area.

levels of area reduction. Multiple draw-anneal sequences were made through successively smaller diameter dies and the final rod cross-sectional areas were reduced by ~ 28 , ~ 49 , and ~ 66 percent; selected rod segments are shown in Figure 6. These tubes all contained a 1-to-1 mixture by volume of zirconium metal powder and zirconium oxide spheres packed to identical green density. The tubes were evaluated to determine the final elongation and the ID, OD, and sheath wall thickness of the compacted tube. The axial elongation and wall thickness reduction are compared with the cross-sectional area reduction in Figure 7. As expected, there is a notable correlation between the area reduction, which is controlled by the drawing die size, and the elongation and the reduction in wall thickness. Further study is required to generate a statistically sound correlation to guide fuel pin design.

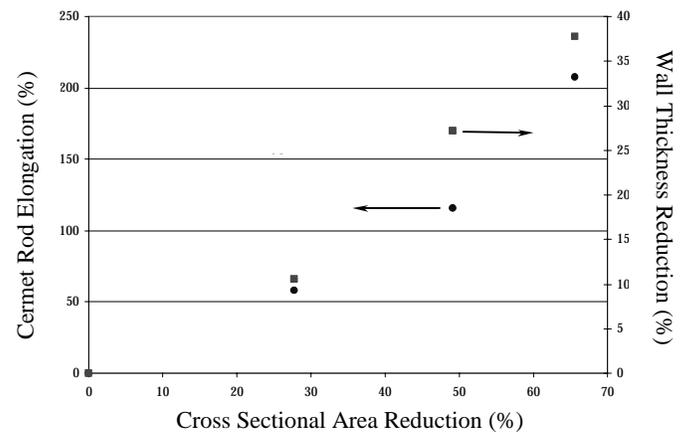


Figure 7. Comparison of Drawing Reductions for Cermet Test Specimens in a Stainless Steel Sheath.

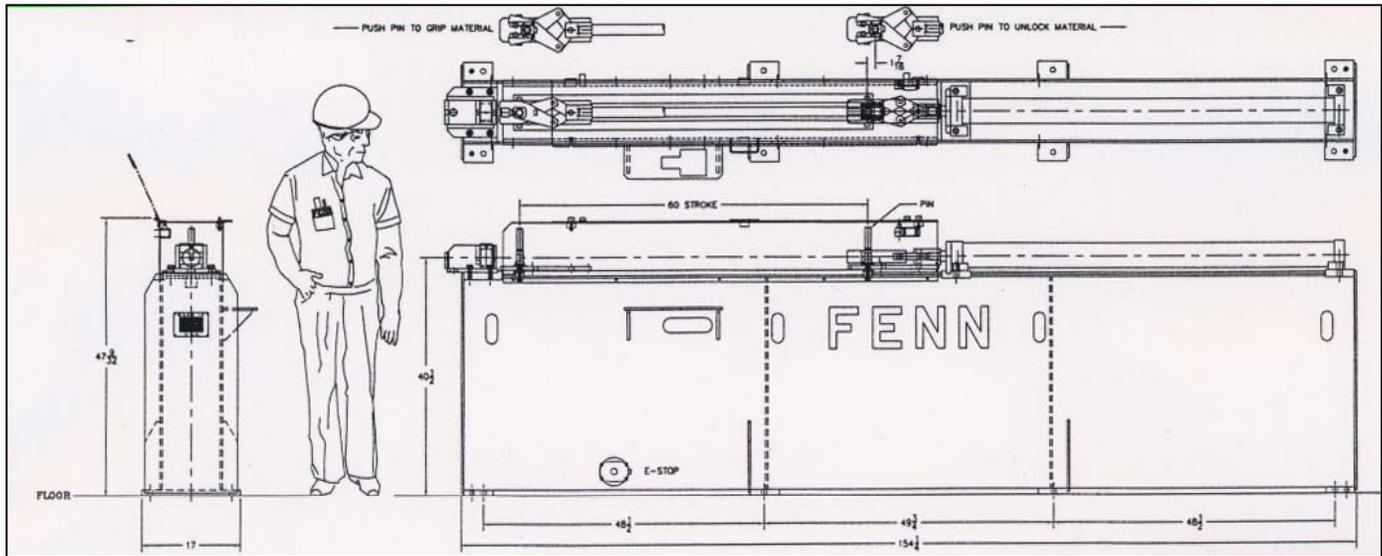


Figure 8. Schematic Diagram of the Laboratory-Scale Hydraulic Drawbench for Fabrication of Simulated (Th,U)O₂ Fuel Pins.

Specification of Lab-Scale Processing Equipment

Two pieces of processing equipment are presently being installed to enable the fabrication of cermet rods containing (Th,U)O₂ microspheres. The first is a high-temperature controlled-atmosphere materials research furnace to be used for the inter-drawing heat treatments. The furnace has a tungsten-element hot zone that is ~5-in diameter and ~12-in tall to accommodate multiple short tubes in a single anneal.

A hydraulic drawbench is also being installed. The bench is capable of draw rods up to ~4-ft in length. A schematic of the drawbench received from the manufacture is shown in Figure 8, and a photograph of the uninstalled bench is shown in Figure 9. Installation of both the drawbench and the high-temperature annealing furnace will facilitate processing radioactive specimens in the final year of this program.

SUMMARY

The zirconium matrix (Th,U)O₂ cermet nuclear fuel has been demonstrated to have significant potential for high burnup and superior fuel performance through modeling simulations. Initial lab-scale experiments are being used to discern the requisite processing parameters and behavior limitations that must be addressed to enable the ultimate implementation of this promising fuel form. The initial results from the powder-in-tube drawing and microsphere coating methods described above indicate that the fabrication of this fuel may be achievable using relatively low-cost methodology.

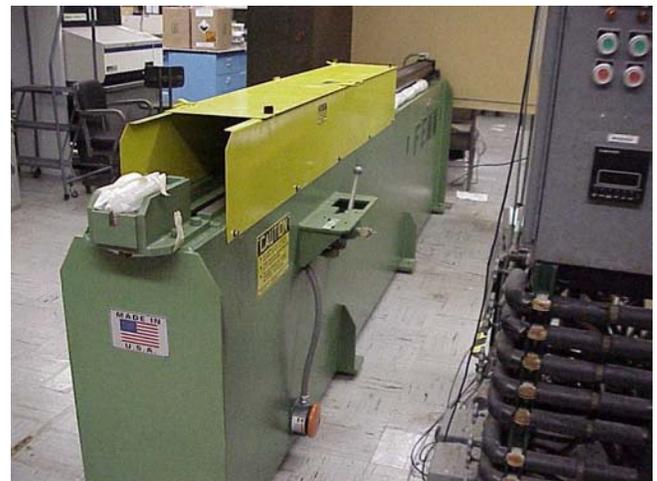


Figure 9. Photograph of the Laboratory-Scale Hydraulic Drawbench for Fabrication of (Th,U)O₂ Fuel Pins.

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