

Technical Project Plan
for
The Enhanced Thermal Conductivity of
Oxide Fuels Through the Addition of
High Thermal Conductivity Fibers and
Microstructural Engineering

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Advanced Fuels Campaign

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**TECHNICAL PROJECT PLAN
FOR
THE ENHANCED THERMAL CONDUCTIVITY OF OXIDE FUELS THROUGH THE
ADDITION OF HIGH THERMAL CONDUCTIVITY FIBERS AND MICROSTRUCTURAL
ENGINEERING**

Prepared for the DOE-NE
Fuel Cycle Research and Development Program
Advanced Fuels Campaign

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EXECUTIVE SUMMARY

The commercial nuclear power industry is investing heavily in advanced fuels that can produce higher power levels with a higher safety margin and be produced at low cost. Although chemically stable and inexpensive to manufacture, the in-core performance of UO_2 fuel is limited by its low thermal conductivity. There will be enormous financial benefits to any utility that can exploit a new type of fuel that is chemically stable, has a high thermal conductivity, and is inexpensive to manufacture. At reactor operating temperatures, UO_2 has a very low thermal conductivity ($<5 \text{ W/m}\cdot\text{K}$), which decreases with temperature and fuel burnup. This low thermal conductivity limits the rate at which energy can be removed from the fuel, thus limiting the total integrated reactor power. If the fuel thermal conductivity could be increased, nuclear reactors would be able to operate at higher powers and larger safety margins thus decreasing the overall cost of electricity by increasing the power output from existing reactors and decreasing the number of new electrical generating plants needed to meet base load demand. The objective of the work defined herein is to produce an advanced nuclear fuel based on the current UO_2 fuel with superior thermal conductivity and structural integrity that is suitable for current and future nuclear reactors, using the existing fuel fabrication infrastructure with minimal modifications.

There are two separate components to the research: (1) Enhanced Thermal Conductivity (ETC) - adding high conductivity fibers to the UO_2 prior to sintering, which act as conduits for moving the heat energy generated within the pellet to the outer surface, (2) Microstructural Engineering (ME) - adding second phase particulates to UO_2 bodies to retard grain growth and to increase thermal conductivity, as well as improve fracture and creep resistance. Different groups will perform the laboratory work for each of these research components with some overlap in personnel. The overlapping areas primarily involve computer simulations and final testing of the fuel in a reactor.

The estimated cost and duration of this project is \$5,000,000 over three years.

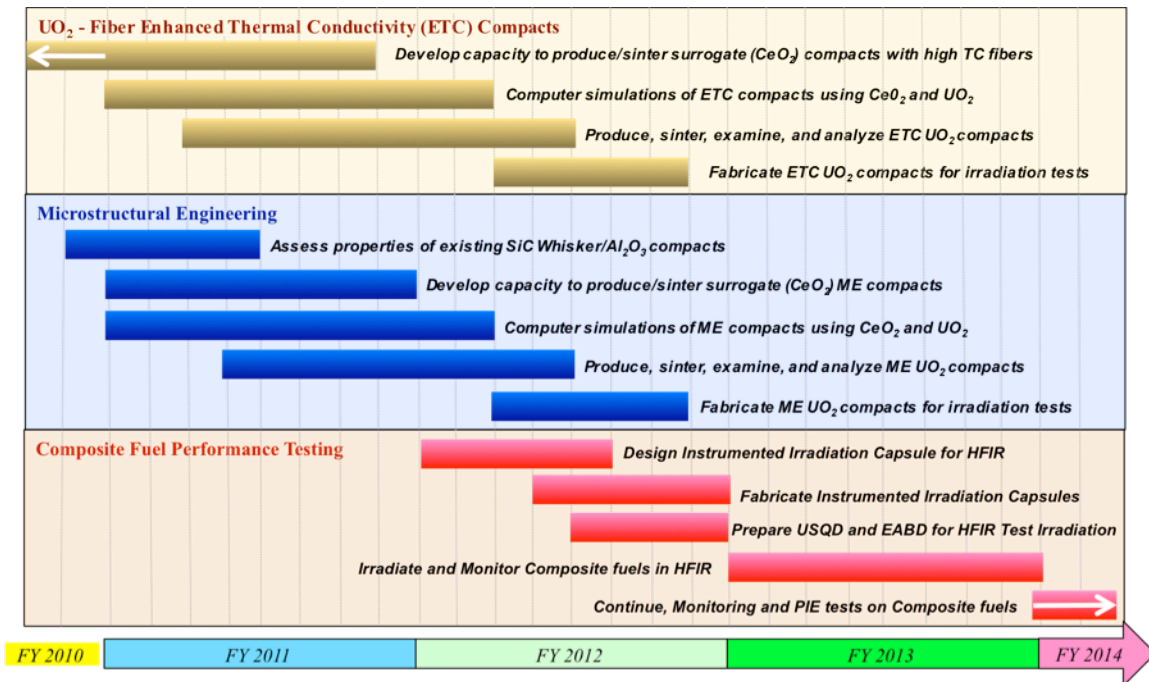


Fig. E-1. Flowchart of Proposed Work Schedule.

1. INTRODUCTION

All current power reactors use UO_2 as their fuel because it is relatively chemically inert and inexpensive to manufacture. The primary problem with UO_2 fuel is that it has a very low thermal conductivity (~ 2.5 $\text{W/m}\cdot\text{K}$ at 1500 K for fresh fuel) that decreases as the fuel is burned,¹ see Figure 1. This limits the rate at which heat energy can be removed from the fuel and thus limits the rate of power generation within the fuel. NRC regulations specify the fuel temperatures must not exceed melting temperatures even under accident conditions. The higher the thermal conductivity, the lower the temperature difference between the fuel centerline temperature and fuel edge temperature thus decreasing the amount of stored heat energy in the fuel and increasing the safety margin. Figure 2 shows the radial temperature profile of a fuel rod as a function of fuel thermal conductivity. As shown, simply doubling the fuel thermal conductivity decreases the centerline temperature by over 400 K whereas increasing it by a factor of 5 drops the centerline temperature only another 300 K for a total difference of 700 K. If the fuel thermal conductivity could be increased, the heat energy could more quickly be extracted from the fuel rod, resulting in cooler, more stable fuel, less fission gas release, and possibly higher total reactor power levels. Accident scenarios (such as Loss of Coolant Accidents) are analyzed assuming certain levels of stored energy in the fuel; since the fuel would be operating at much lower temperatures, the current restrictions imposed by these analyses would be extremely conservative and could be eased.

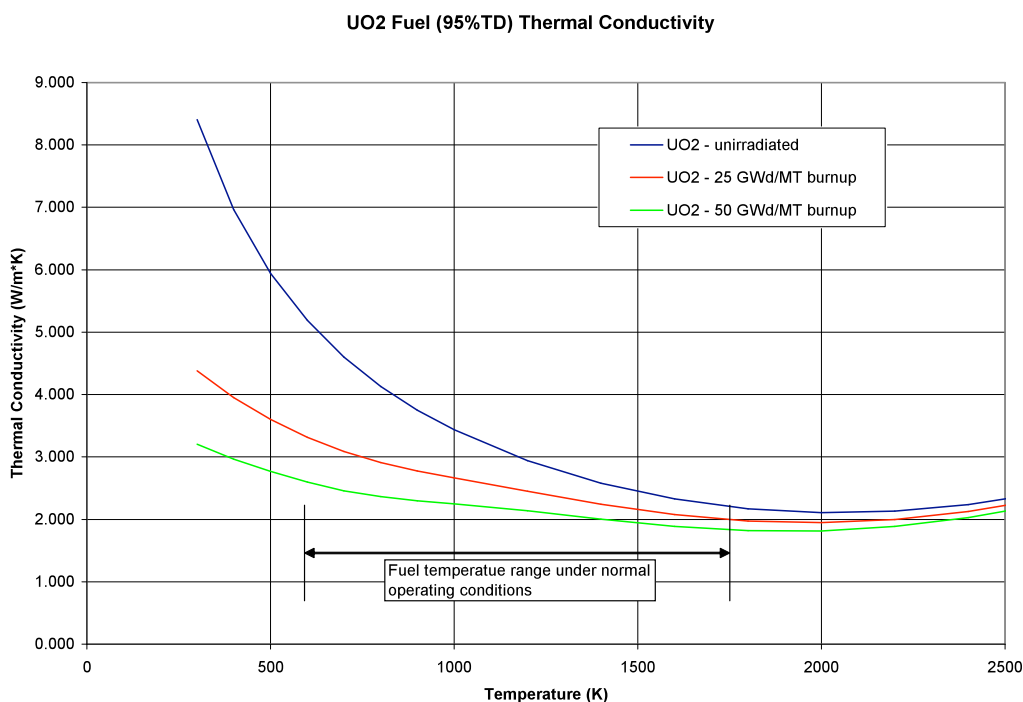


Fig. 1. UO_2 fuel thermal conductivity as a function of temperature (K) for UO_2 fuel at different burnups of gigawatt-days per metric ton of uranium (GWd/Mt).

Historically,^{2,3} several advanced nuclear fuels having higher thermal conductivities (k) have been extensively studied. Different chemical compositions of uranium having higher material thermal conductivities were examined such as: uranium metal, uranium carbide (UC), and uranium nitride (UN). Although uranium metal has been successfully used in fast reactors using liquid sodium as a coolant, its suitability for use in water reactors is problematic since uranium metal reacts with water. UC and UN also have very high thermal conductivities (>20 $\text{W/m}\cdot\text{K}$) but are more expensive to manufacture and are

chemically more reactive than UO_2 with water at operating temperatures. Even given these limitations, U-metal, UC and UN are being examined as possible replacements for UO_2 as the primary fuel in water-cooled reactors because of their high thermal conductivities and mechanical stability as the fuel is burned. A different approach to creating a high thermal conductivity fuel is to start with UO_2 and add a second high thermal conductivity material. This second material that is added to the UO_2 must be compatible, so it does not react with the UO_2 or water, and have a very low neutron absorption cross-section. The composition, orientation, and amount of secondary material are all crucial elements in optimizing the thermal conductivity and structural integrity of the fuel. The objective is to improve the physical properties of the fuel using the minimum amount of secondary material.

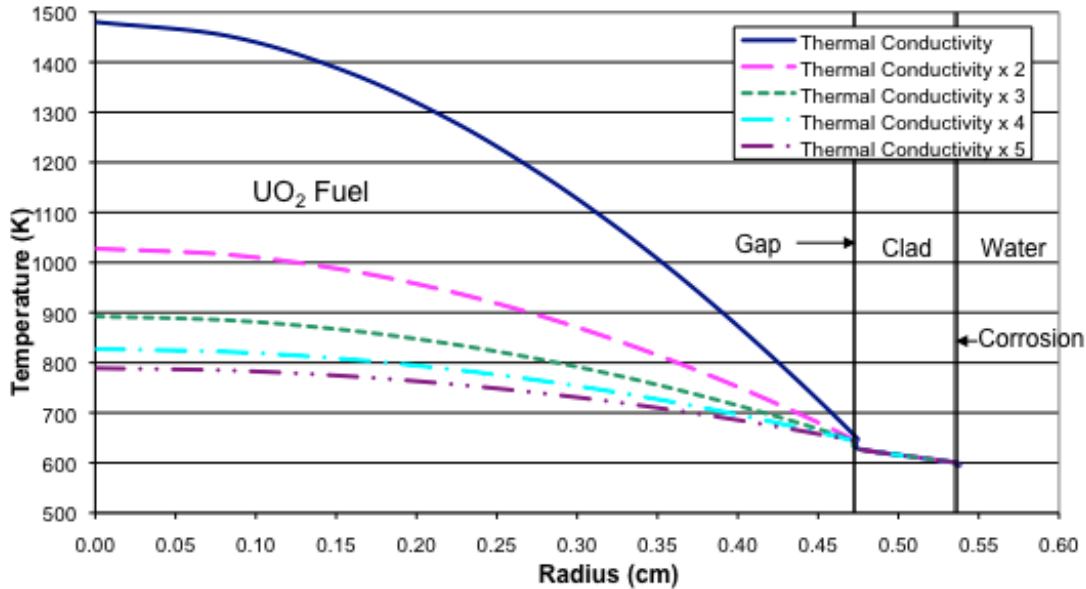


Fig. 2. Radial temperature gradient across a UO_2 fuel pin as a function of thermal conductivity.

1.1 HIGH THERMAL CONDUCTIVITY FIBER BACKGROUND

One method used to increase the thermal conductivity of UO_2 fuel currently being explored at ORNL is the addition of long thin fibers of highly ordered graphite encased within a thin SiC outer layer. Theoretically, graphite or other types of fibers in UO_2 fuel pellets should act as heat conduits, which more efficiently transfer the energy generated deep inside the pellet to the outer edge. Both carbon and silicon have very low absorption cross-sections and have been used in nuclear reactors in the form of coatings on particulate gas-cooled reactor fuel. Amorphous graphite has a thermal conductivity of about $10 \text{ W/m}\cdot\text{K}$. However, the theoretical thermal conductivity of highly ordered graphite along crystallographic basal planes is more than $2000 \text{ W/m}\cdot\text{K}$ at room temperature, and $\sim 10 \text{ W/m}\cdot\text{K}$ perpendicular to the basal planes.⁴ At reactor operating temperatures the planar dimension thermal conductivity drops to $\sim 200 \text{ W/m}\cdot\text{K}$ but the transverse dimension thermal conductivity stays fairly constant. SiC is much more chemically stable than graphite with respect to UO_2 but has a lower thermal conductivity and is more susceptible to radiation damage. SiC coated graphite fiber is a reasonable compromise between high thermal conductivity and chemical compatibility with UO_2 .

There are concerns associated with compressing and sintering UO_2 pellets containing graphite fibers at high temperatures. As the pellet is compressed and sintered the graphite fibers may shatter and disperse or interact with the UO_2 forming CO_2 and UC thus destroying the heat transfer pathway. Ideally, to optimize

thermal conductivity, the fibers need to be intact, should extend from the pellet centerline to the outer edge, have a regular symmetric spacing and be oriented radially perpendicular to the Z-axis. Each of these characteristics will affect the bulk thermal conductivity of the pellet. The problem lies in not knowing the exact form and orientation of the fibers after compression and sintering. Mixing will randomly orient the fibers while uni-axial compression should orient the fibers in a direction more perpendicular to the Z-axis, the axis of compression. In an open system or when combined with UO_x where $x > 2$ at high temperatures, graphite will combine with the oxygen in UO_2 forming CO or CO_2 . Currently, UO_2 pellets are sintered in a reducing (argon/4–6% H_2) atmosphere at temperatures $>1800K$ yielding a pellet having on average 1.98 oxygen atoms per uranium atom. However, within the pellet the oxygen potential will remain high enough that during the reduction to $UO_{1.98}$ the excess oxygen will oxidize the graphite. The interaction between the UO_2 and graphite should be all but eliminated by the thin layer of SiC on the outer surface of the graphite fiber under processing and operating conditions.

Preliminary computational studies have shown that a substantial increase in the bulk thermal conductivity of low thermally conductive material is possible through the addition of long thin fibers of high thermal conductivity material. Simplified cases consisting of long thin fibers having a length/diameter ratio of 100 were computationally analyzed for different volume percentage of high thermally conducting fibers in a low thermally conducting material. For this study the low thermally conducting material has properties similar to UO_2 with its thermal conductivity equal to 3 W/m·K. The high thermally conducting material has properties similar to idealized highly ordered graphite with its thermal conductivity ranging from 50 to 500 W/m·K along the axis and 10 W/m·K radially. An additional calculation using Tungsten wire having an isotropic thermal conductivity of 173 W/m·K was also done. Figure 3 below shows how the bulk material thermal conductivity increases with increased fiber volume percent in an idealized system with the fibers perpendicular to the heat transfer surfaces. As shown in Figure 3, a four volume percent addition of highly conducting fibers increases the bulk thermal conductivity of the material by about a factor of 7 for 500 W/m·K fibers; which has the equivalent thermal conductivity of UC or UN fuel without their inherent problems and at a substantially lower manufacturing cost. A 2-4 volume percent addition of SiC coated graphite fibers on the order of 1 to 5 mm in length and 10 μm in diameter should provide the desired substantial bulk thermal conductivity increase.

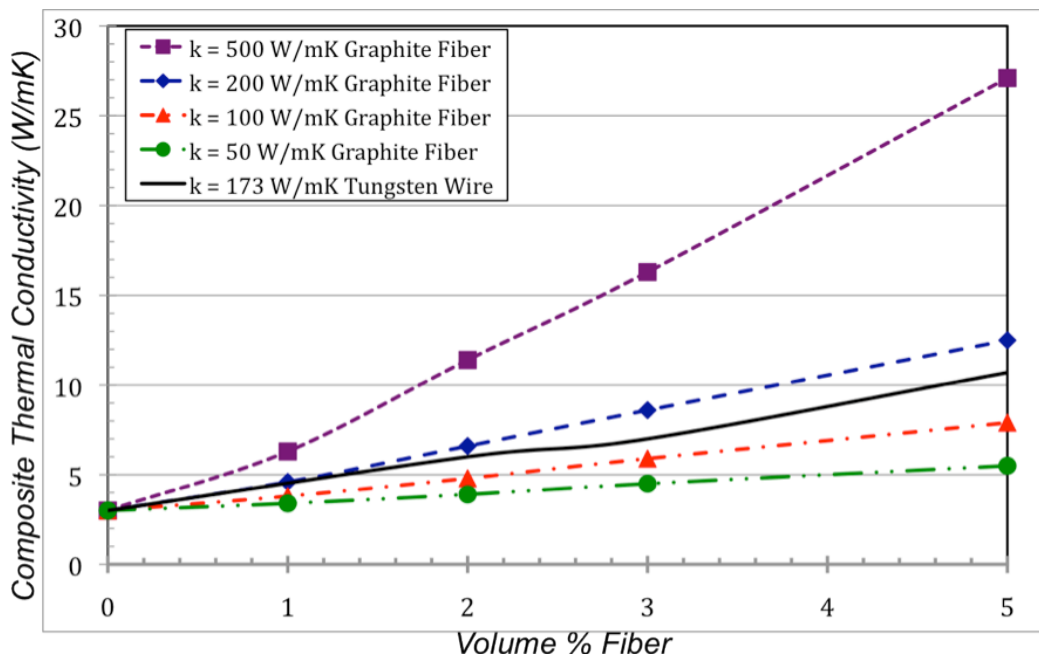


Fig. 3. Increase in thermal conductivity as a function of graphite fiber volume percent.

1.2 MICROSTRUCTURAL ENGINEERING BACKGROUND

The objective of this work is to improve the thermal conductivity and other material properties of UO_2 fuel through the introduction of second phase particles to polycrystalline UO_2 . Fission gases (Xe, Kr) produced during fission have low solubility in UO_2 . After a relatively short irradiation period a large number of fission-gas filled bubbles are generated within the fuel grain. The gas atoms in the UO_2 migrate to the grain boundaries unless trapped by bubbles. At temperatures where ample grain growth occurs, processes like grain boundary movement can sweep up fission gas atoms more rapidly than they could have arrived at the boundary by diffusion. In addition to providing a heat conduction path the secondary particles can also pin the grain boundaries preventing excessive UO_2 grain growth, which stabilizes the material properties and minimizes fission gas release. As the content of the second phase particles increases, a rapid increase in the bulk thermal conductivity occurs at the percolation limit; when the secondary material forms an interconnected path through the material instead of small isolated patches. The resultant increase in thermal conductivity will reduce the thermal gradient in the fuel further minimizing excessive grain growth and reducing fission gas release.

Several different approaches have been taken to increase the overall bulk thermal conductivity of UO_2 fuel. One approach involves adding a high conductivity metal to the fuel that provides a continuous heat conduction path; this type of composite material is more commonly known as cermet (ceramic metal). Conventional cermet contains at least 30% metal in order to provide a continuous metal phase throughout the fuel.⁵ A fabrication process creating a UO_2 -W composite was developed which significantly reduces the amount of metal required. A UO_2 -W composite with only 6 vol% W was manufactured with an 80% increase in the thermal diffusivity compared to a standard UO_2 pellet.⁶ A major problem with most cermet materials is that the neutronic properties of the metal is not appropriate for use in a nuclear reactor. The metal has a high neutron absorption cross-section thus competing with the uranium, and activates, thus increasing the radioactivity of the spent fuel.

The bulk electrical and thermal conductivity properties of oxide can be improved by the addition of secondary phase particles. Mebane and Gerhardt⁷ have shown that the addition of about 10 vol.% of a second phase particle increased the electrical conductivity by two orders of magnitude once the percolation limit was reached. Likewise, the thermal conductivity of aluminum oxide ceramics increases with the addition of second phase particles having high thermal conductivity.⁸

Tulenko at the University of Florida has been working on adding SiC secondary particles to UO_2 to increase the bulk thermal conductivity.⁹ Silicon Carbide (SiC) is a potentially good material because of its high thermal conductivity, low neutron absorption cross-section, and relative stability in a radiation field. Several methods of mixing UO_2 and SiC were investigated. Directly mixing SiC Whiskers and UO_2 in a liquid medium was unsuccessful because the SiC, being less dense, tended to stay on top of the UO_2 . It was necessary to grind the dry UO_2 and SiC together to homogenize the mixture. An attempt was also made to directly coat the UO_2 particles with SiC but this proved unsuccessful due to the polymer precursor being oxidized by the UO_{2+x} .

The physical properties of both SiC and Alumina (Al_2O_3) whiskers make them reasonable candidates as second phase particles added to UO_2 . Both materials have melting temperatures in excess of 2000°C , low neutron absorption cross-sections, and are relatively stable when exposed to radiation. Alumina has a relatively stable thermal conductivity of $30 \text{ W/m}\cdot\text{K}$ over temperature while the thermal conductivity of high purity SiC peaks at over $400 \text{ W/m}\cdot\text{K}$ but decreases with temperature as shown in Figure 4. SiC whiskers have a much lower thermal conductivity, $\sim 50 \text{ W/m}\cdot\text{K}$, but dramatically decreases above about 1000°C .

The objective would be to mix UO_2 with second phase particles. The effects of the shape, size and volume content of the second phase particles will be assessed. As noted above, second phases in the form of whiskers having relatively large aspect ratios offer the benefit of a high thermal conductivity along the particle axis. It has been shown that during uniaxial pressing the whiskers rotate the long axis toward the plane normal to the uniaxial pressing axis. This should assist in increasing the radial thermal conductivity of fuel pellets. There are added benefits to fuel pellets aside from an increased thermal conductivity. The whiskers impart to the material increased fracture strength and toughness as well as resistance to thermal shock and creep resistance.^{11, 12}

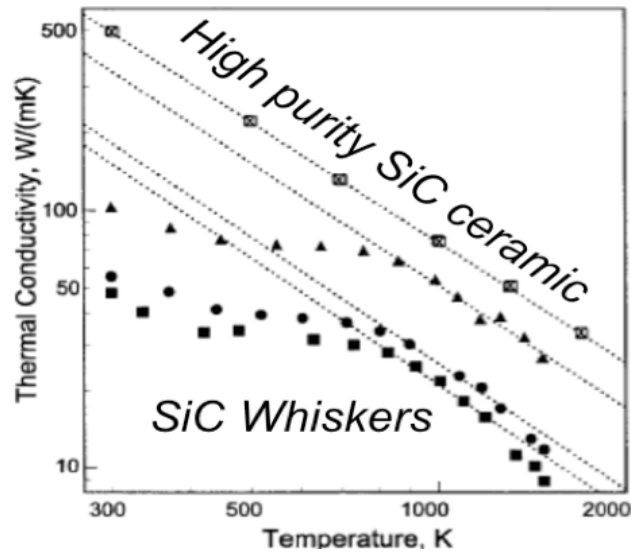


Fig. 4. SiC Thermal Conductivity as function of Temperature.¹⁰

2. PROJECT ELEMENTS

The overall objective of this project is to improve the thermal conductivity and other physical properties of UO_2 fuel through the addition of a minimum amount of secondary material. Two separate approaches are being investigated: adding long high thermal conductivity fibers to UO_2 prior to pressing and sintering and adding second phase particles/whiskers to the UO_2 prior to pressing and sintering. Both approaches will be designed to fit into standard fuel fabrication techniques with minimal changes.

2.1 TASK 1 - HIGH THERMAL CONDUCTIVITY FIBER TASKS

This line of research investigates improving the thermal conductivity of UO_2 fuel through the addition of high thermal conductivity fibers. In order to simplify the initial phase of the research and development, CeO_2 will be used as a surrogate material for UO_2 . The overall objective of the initial phase is to determine the change in bulk thermal conductivity of a compact as a function of CeO_2 grain size, fiber volume percent, fiber length, fiber material, and temperature. The fibers being examined will be composed of two different materials: SiC coated highly ordered graphitic fibers (referred to as graphite fibers) and tungsten fibers. Both fibers have diameters of $\sim 10 \mu\text{m}$ and lengths from 1 – 5 mm resulting in an aspect ratio of from 100 to 500.

Graphite fibers and tungsten fibers are both being examined because of their vastly different physical properties. The graphite fibers have the best neutronic characteristics of all known high thermal conductivity fibers. They are however brittle, the thermal conductivity changes by an order of magnitude between room temperature and reactor operating temperature, and the physical characteristics of SiC and possibly graphite degrades over time in a neutron radiation field. Tungsten fibers are tough, flexible, do not react with UO_2 and have a stable thermal conductivity of $\sim 180 \text{ W/m}\cdot\text{K}$ over the operating range of a power reactor. However tungsten has unfavorable neutronic properties due to its large neutron absorption cross sections and large resonances.

The approach to protecting the graphitic fibers from oxidation during sintering and irradiation involves having a thin SiC layer on the outer surface of the fibers. A successful approach recently demonstrated at ORNL siliconizes the graphite fiber outer surface. A gas mixture of Ar-4%SiH₄ flows through a bed of fibers in a reaction chamber at elevated temperature. Elemental silicon deposits on the fibers, with some interaction/carbide formation. In order to have sufficient reaction to form a SiC layer on the surface of the fibers a post-deposition heat treatment is performed under inert gas. Electron microscopy of the product fibers, see Fig. 5, indicate sufficient SiC surface material on the fibers that is adherent and coherent, yet a large majority of the diameter remains solely carbon as desired.

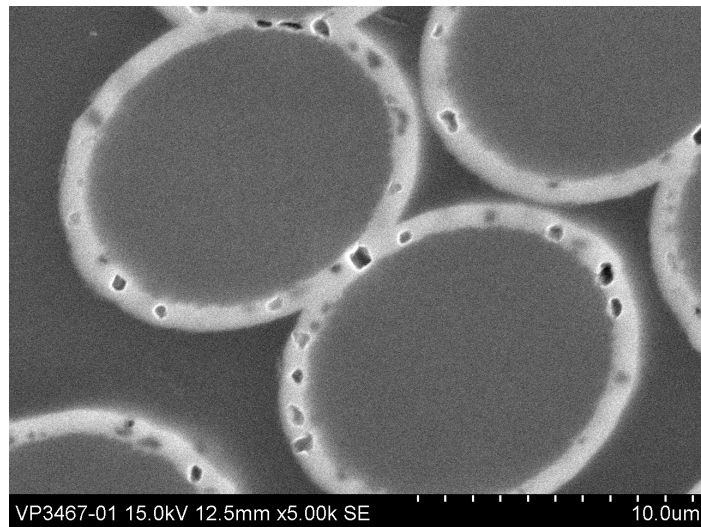


Fig. 5. Micrograph of SiC coated graphite fibers.

Different compact fabrication techniques are being developed that take advantage of the different fiber characteristics. The flexibility of the tungsten fibers means they will not shatter when exposed to excessive forces as happens with graphite fibers. However, the tungsten fibers tend to entangle into a single mass when mixed with CeO_2 particles in a blender. Due to the tendency of the tungsten to entangle and the brittle nature of the graphite fibers, a mixing and processing method that imparts minimal shear forces on them is required. If the mixing is too aggressive the graphite fibers will break into smaller pieces and expose carbon surfaces. Dispersants are used in liquid suspensions of oxide and fibers to form a homogeneous mixture; which are then dried. The dried powder/fiber mixture is then isostatically pressed to form a green pellet of at least 50% theoretical density. The compact is then sintered in a controlled atmosphere to produce a pellet with the desired density, generally $\geq 95\%$ theoretical density (TD).

The thermal conductivity of each compact will be determined using Laser Flash Thermal Diffusivity over the range of temperatures from 20 – 1000°C. The technique works by applying a short laser pulse at one

side of a compact and measuring the time it takes for the heat to travel through the compact to the other side. Using the time needed to reach 50% of maximum temperature and the sample thickness a diffusivity coefficient can be calculated. Using the diffusivity coefficient, the material density and heat capacity the bulk thermal conductivity can be determined. Even in the absence of known heat capacity, comparative thermal diffusivities will provide the necessary guidance.

Concurrent with the manufacturing and testing of the CeO₂-fiber compacts, computer simulations will be conducted with the intent to calculate the bulk thermal conductivity using various amounts and sizes of graphite and tungsten fibers in compacts. ORNL researchers have developed the capability, based on finite element analysis (FEA), to computationally evaluate the bulk material properties, such as thermal conductivity, of composites composed of multiple materials having different properties.¹³ These computer simulations will be used to assist in the selection of fiber dimensions and amounts to be mixed with the CeO₂ powder to create the compacts. Optical micrographs of the compacts will be created to physically examine the final structure of the fibers in the compacts. These will be used as input to the computer codes to benchmark the analytical techniques and help determine other fiber dimensions and concentrations to be used in compacts.

When sufficient progress has been made using CeO₂, work will begin using UO₂. The types and volume percents of fibers should be very similar to those determined using CeO₂. Likewise, the mixing and sintering techniques developed should also be directly applicable. The primary work will involve adjusting the chemistry parameters such as binding and dispersant agents and concentrations. After UO₂/fiber compacts have been manufactured, detailed chemical and physical analyses of the compacts will be performed. Laser Flash Thermal Diffusivity measurements of the compacts ranging from 20°C to 1000°C will be performed. Mechanical properties of the compacts such as toughness and strength will be assessed. Scanning Electron Microscopy (SEM) will be used to ensure the fibers are properly distributed in the compacts. Other physical properties of the compacts will be fully assessed and compared to standard UO₂ fuel prior to the next step.

Once the physical characteristics of the compacts are fully understood and found to be acceptable, test rods will be manufactured and placed in the High Flux Isotope Reactor (HFIR) for full power irradiation testing. ORNL has developed a test assembly for HFIR that approximates a standard PWR's power and neutron flux profile.¹⁴ Temperature and pressure sensors in the fuel pin will continuously monitor the pellet internal centerline temperatures and gas pressure as the fuel pin is irradiated. At regular intervals individual fuel rods will be removed and post-irradiation examination (PIE) will be performed. The biggest unknowns are the effects of the radiation on the pellet's physical properties as the fuel burns. Questions that will be addressed in the irradiation/power testing phase include but are not limited to: will the damage from the radiation and neutron absorption change the thermal conductivity, how is fission gas release affected, what is the effect on pellet clad interaction, and how is pellet swelling affected.

The end result of this research is to develop a UO₂ fuel pellet that has superior physical properties to standard UO₂ pellets and can retain those properties for much higher burnups and power densities. For this to work the high thermal conductivity fiber needs to retain its properties for the length of time the fuel is in the reactor and be neutronically benign. Below is a summary of the necessary procedures to complete this research and bring it to a level where it can be used in industry.

1. A technique for adding a SiC outer layer to graphite fibers has already been developed at ORNL involving passing Ar-4%SiH₄ (silane) gas over graphite fibers at elevated temperature
 - a) Manufacture additional SiC coated graphite fibers using the techniques developed
2. ORNL has obtained several spools of 99.9% pure 10 μm diameter tungsten wire
 - a) Cut the tungsten wire into lengths ranging from 1 to 5 mm
3. Create CeO₂/fiber (SiC coated graphite or tungsten) compacts using ceramic processing techniques
 - a) Examine different binding agents and concentrations
 - b) Examine different dispersant agents and concentrations
 - c) Vary fiber volume percent and lengths
4. Sinter the compacts at 1700°C to 95%TD
 - a) SEM and optical micrographs
5. Measure thermal diffusivity/conductivity (*k*) of the compacts using laser flash thermal diffusivity:
 - a) Perform measurement at room temperature
 - b) Perform measurement at elevated temperatures to up to 1000°C
6. Perform computer simulations of thermal conductivity (*k*) at various fiber content
7. Repeat steps 3, 4 and 5 using UO₂ in place of CeO₂
8. Manufacture fuel pellets and an instrumented test assembly for HFIR
9. Place test assembly in HFIR for radiation/power tests
10. Periodically remove fuel rods for PIE

2.2 TASK 2 - MICROSTRUCTURAL ENGINEERING TASKS

This line of research investigates improving the thermal conductivity and other physical properties of UO₂ fuel through the addition of second phase particulates. Evaluations will include the restraint of grain growth and the immobilization of solid fission product inclusions through pinning of grain boundaries by second phase particles. In order to simplify the initial phase of the research and development, CeO₂ will be used as a surrogate material for UO₂. The overall objective of the initial phase is to determine the change in bulk thermal conductivity and other physical properties of a compact as a function of CeO₂ grain size; second phase particle type, grain size, and volume fraction; and percolation limit.

Initial studies employing CeO₂ as the matrix (also fluorite structure with melting point 2400°C somewhat lower than UO₂ 2865°C) will examine the effects of second phase particle size and volume fraction on the microstructural stability. With the knowledge gained in these studies, similar studies will combine microstructural stability with increased thermal conductivity with the addition of a high thermal conductivity second phase (e.g., BeO 280-300 W/ m·K or SiC particles (~400 W/ m·K) or whiskers (~50 W/ m·K). The percolation limit (i.e., the amount of second phase needed to increase thermal conductivity) will be established as a function of volume content and size of the second phase material through modeling and experimental studies. Laser Flash Thermal Diffusivity measurements of the compacts ranging from 20°C temperature to 1000°C will be performed.

Computer simulations will be conducted with the intent to calculate the bulk thermal conductivity, fracture strength, toughness, and creep resistance using various amounts and types of second phase particles. ORNL has developed the capability, based on finite element analysis (FEA), to computationally evaluate the bulk material properties, such as thermal conductivity, of composites composed of multiple materials having different properties.¹³ These computer simulations will be used to assist in the selection of second phase particle type and amounts to be mixed with the CeO₂ powder to create the compacts. Optical micrographs of the compacts will be created to physically examine the final structure of the compacts. These will be used as input to the computer codes to benchmark the analytical techniques and help determine the composition of future compacts.

A critical element in the effectiveness of high thermal conductivity additions (fibers, whiskers or particles) is that of the interfacial thermal resistance, which dictates whether or not one achieves any improvement in the thermal conductivity in the resultant composite. The interfacial resistance can result from several factors, e.g., non-intimate contact between the matrix and the second phase (fiber, whisker or particle), or accumulation of segregating species (impurities, additives) at the interfaces. In essence, the interfacial thermal resistance can negate any contribution of the high conductivity phase. For instance in Aluminum Nitride (AlN), sintering additives and the grain size can alter the thermal resistance of grain boundaries and hence, the overall thermal conductivity of the ceramic; high thermal conductivity is achieved by eliminating segregation at grain boundaries.^{15,16} Recent results with SiC whisker reinforced alumina composites reveal that a combination of MgO and Y₂O₃ sintering additives produce temperature-dependent thermal conductivities that are essentially the same as that of a dense alumina ceramic. Both MgO and Y₂O₃ have extremely limited solubility in alumina; thus, they typically will tend to reside at grain or whisker interfaces. In the absence of additives, the conductivity increases with content of SiC whiskers.¹⁷ The results of second phase particle characteristics can also have an affect.^{10,18} Differences in thermal expansion coefficients between matrix and second phase additions can alter the contact between them. The expansion coefficient of UO₂ is about twice as large as that of SiC. The interface characteristics (e.g., continuity, chemistry and thermal resistance) will be characterized using various techniques (e.g., Scanning Transmission and Transmission Electron Microscopy combined with chemical analysis to detect interfacial segregation and/or interfacial films, and Scanning Probe Microscopy thermal characterization¹⁹ to measure thermal transport.

When sufficient progress has been made using CeO₂, work will begin using UO₂. The types and volume fractions of second phase particles should be very similar to those determined using CeO₂. Likewise, the mixing and sintering techniques developed should also be directly applicable. After compacts have been manufactured detailed chemical and physical analyses of the compacts will be performed. Flash Laser Diffusivity measurements of the compacts ranging from 20°C to 1000°C will be performed. Mechanical properties of the compacts such as toughness and strength will be assessed to ensure the second phase particle/UO₂ matrix is stable. SEM will be used to examine the distribution of second phase particles in the compacts. The physical properties of the compacts will be fully assessed and compared to standard UO₂ fuel prior to the next step.

Once the physical characteristics of the compacts are fully understood and found to be acceptable, fully instrumented test rods will be manufactured and placed in HFIR for full power irradiation testing. ORNL has developed a test assembly for HFIR that approximates a standard PWR's power and neutron flux profile. Temperature and pressure sensors in the fuel will continuously monitor the pellet internal centerline temperatures and gas pressure as the fuel pins are irradiated. At regular intervals individual fuel rods will be removed and PIE will be performed. The biggest unknowns are the effects of the irradiation on the pellet's physical properties as the fuel burns. Questions that will be addressed in the irradiation testing phase include but are not limited to: will the damage from the radiation and neutron absorption change the thermal conductivity of the compacts, how is fission gas release affected, what is the effect on pellet clad interaction, and how is pellet swelling affected.

The end result of this research is to develop a UO₂ fuel pellet that has superior physical properties to standard UO₂ pellets and can retain those properties for much higher burnups and power densities. For this to work the compacts need to retain their properties for the length of time the fuel is in the reactor and be neutronicly benign. Below is a summary of the necessary procedures to complete this research and bring it to a level where it can be used in industry.

1. Assess properties of existing $\text{UO}_2/\text{Al}_2\text{O}_3$ compacts existing at ORNL
 - a) Thermal Conductivity
 - b) Fracture Strength and Toughness
 - c) Thermal Shock and Cyclic Fatigue
 - d) Creep Resistance
2. Create CeO_2 / second phase particle compacts
 - a) Al_2O_3 , SiC, BeO at different concentrations
 - b) Examine physical properties (SEM, ceramotography, optical micrographs, etc.)
 - c) Establish percolation limits
3. Measure thermal diffusivity/conductivity (k) of the compacts using laser flash diffusivity:
 - a) Perform measurement at room temperature
 - b) Perform measurement at elevated temperatures to at least 1000°C
4. Perform computer simulations of thermal conductivity (k), fracture strength and toughness for selected second phase particle compacts
5. Repeat steps 2 and 3 using UO_2 in place of CeO_2
6. Manufacture fuel pellets and an instrumented test assembly for HFIR
7. Place test assembly in HFIR for irradiation tests simulating as close as possible LWR burnup, fluence, and neutron energy spectrum
8. Periodically remove fuel rods for PIE

3. PROJECT SCHEDULE AND COST

The budget proposed extends over three fiscal years and sums to a total of just over \$5,000,000. There are two distinct approaches to the development of the new fuel: compacts composed of UO_2 and high thermal conductivity fibers (ETC) and compacts composed of UO_2 and second phase particles (ME). The cost of each line of research is independent up to the manufacture of the pellets that will be used as test fuel. At this point the total cost of writing the safety analysis report; manufacturing the fuel/test rodlets; inserting, monitoring, retrieving and examining the fuel in the test assemblies is dependent on the number of test articles. Table 1 provides an overview of the project cost; Figure 6 provides the high level schedule. Assuming acceptable fuel compacts can be manufactured and placed in HFIR as part of an instrumented test assembly, further funding would be required for extended monitoring and PIE to further contribute to fuel qualification efforts.

Table 1. Overview of Cost and Work Schedule

2011		2012		2013	
Task	Cost (\$1000)	Task	Cost (\$1,000)	Task	Cost (\$1,000)
Project Management, Quality Assurance, and Travel	158	Project Management, Quality Assurance, and Travel	158	Project Management, Quality Assurance, and Travel	158
ETC - Develop CeO ₂ Compacts	500	ETC - Fuel Pellet Production	400	Irradiate Rodlets in HFIR	100
ETC - Compute Simulations	100	ME - Fuel Pellet Production	400	Monitor Test Irradiation in HFIR	300
ETC - Develop UO ₂ Compacts	300	Design Instrumented Irradiation Facility	750		
ME - Assess existing SiC-Al ₂ O ₃ compacts	200	Fabricate Rodlets and HFIR Instrumented Irradiation Facility	750		
ME - Create CeO ₂ /2 nd phase particle compacts	500	Prepare USQD & EABD for HFIR Test Irradiation	200		
ME - Compute Simulations	100				
ME - Develop UO ₂ Compacts	300				
Total	\$2,158		\$2,658		\$558

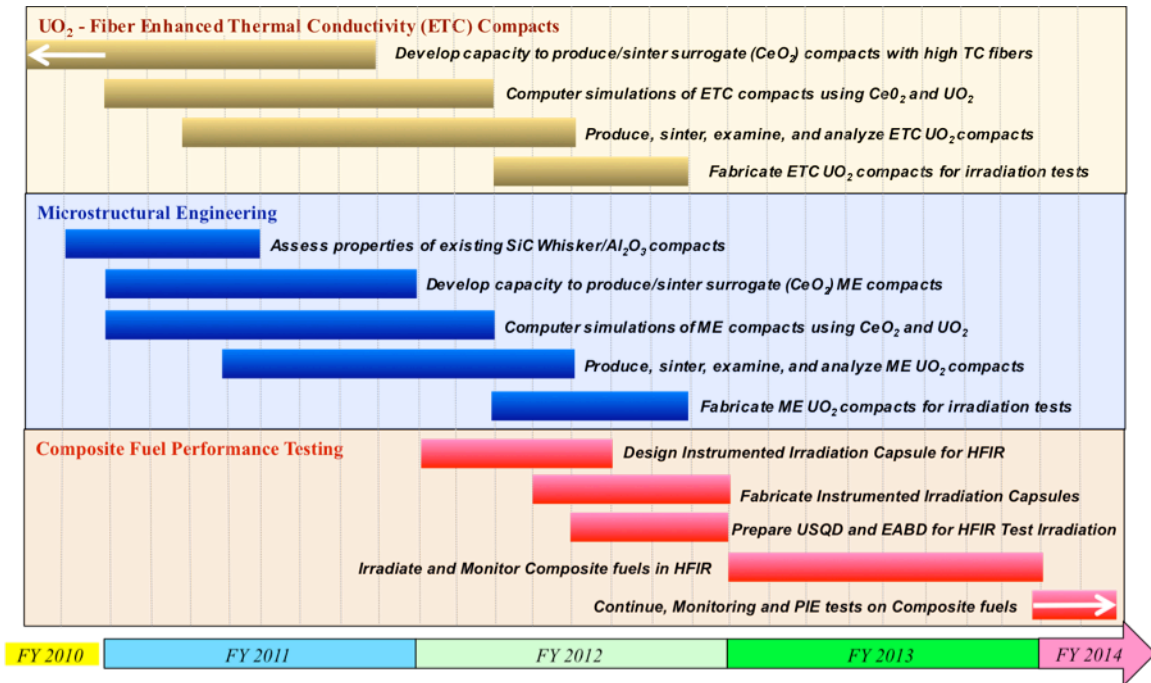


Fig. 6. High level work Schedule.

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