
NUCLEAR ENERGY RESEARCH INITIATIVE

5. Advanced Nuclear Fuels/Fuel Cycles

This element of the program includes 16 NERI research projects to date of which 8 were awarded in FY 1999, 1 in FY 2000, 1 in FY 2001, and 6 in FY 2002. It includes research and development to provide measurable improvements in the understanding and performance of nuclear fuel and fuel cycles with respect to safety, waste production, proliferation-resistance, and economics, in order to enhance the long-term viability of nuclear energy systems. This effort includes enhanced performance of fuels for advanced systems, and development of fuels capable of withstanding the conditions in the supercritical LWR regime and of advanced proliferation-resistant fuels capable of high burn-up such as those needed in support of the Generation IV concepts.

The scope of this long-term R&D encompasses a variety of thermal and fast spectrum power reactor fuel forms, including ceramic, metal, hybrid, (e.g., cermet, cermet), and liquid, as well as such fuel types as oxides, nitrides, carbides, and metallics. Enabling technologies such as advanced cladding, water chemistry, and alternative moderators and coolants are also considered.

The fuel cycle research includes consideration of advanced enrichment technologies for fuel and burnable absorbers and considers the impact of fuel cycle options on the proliferation of nuclear weapons materials. R&D topics also include development of higher density LEU (<20 percent U-235) fuels for research and development reactors.

Currently selected projects include innovative concepts for the following:

- Material preparation and production of nuclear fuels
- Inherently safe fuel designs and core response
- Study of life-limiting phenomena for high burn-up or long life fuels
- High temperature fuel and material performance
- Critical safety data and reactor physics data for advanced fuel compositions and enrichments above five percent
- Innovation in fuel design, composition, or other attributes that maximize energy production, optimize fissile material utilization, or reduce production costs

NUCLEAR ENERGY RESEARCH INITIATIVE

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NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Improved Burnable Poisons for Commercial Nuclear Power Reactors

Primary Investigator: M.L. Grossbeck, Oak Ridge National Laboratory

Project Number: 99-074

Project Start Date: August 1999

Project End Date: June 2003

Research Objectives

Burnable poisons (BPs) are materials that are strong absorbers of neutrons but transmute to weak absorbers upon absorption of a neutron. Such materials are used in nearly all nuclear reactors to level the power distribution and to aid in reactivity control. Burnable poisons will become even more important in Generation IV reactors where a blend of various materials with differing absorption cross sections and, therefore, lifetimes, will be necessary to permit high initial fuel loading, making long-life cores possible.

Burnable poisons used at the present time suffer from two disadvantages. The first is that boron, which is widely used, transmutes to helium, which creates undesirable internal fuel pin pressures. The second is that all other materials do not burn, or transmute, fully, and thus some absorbing material remains at the end of fuel life. This limits the amount of fuel that can be used, resulting in less efficient operation than could otherwise be achieved. Elimination or reduction of these two effects will lead to higher fuel burnup, resulting in lower cost of operation.

For many absorbing elements, such as gadolinium, it is isotopes other than the primary absorber that lead to the undesirable residual reactivity. The objective of this research is to identify single isotopes that can be used as burnable poisons that would not remain in significant quantities at the end of core life. State-of-the-art computer codes are being used to model a pressurized water reactor core with various burnable poison configurations. The second phase of the project is to separate isotopes of candidate elements to determine the enrichment attainable, the annual production, and the cost. The third phase of the project investigates compatibility of the absorber materials with the fuel and identifies potential difficulties in their use.

Research Progress

The progress of each phase of the project will be discussed in turn.

Identification of Candidate Isotopes: The first phase of the project is complete in that isotopes have been identified as potential advanced burnable poisons. The isotopes ^{157}Gd , ^{149}Sm , ^{167}Er , ^{164}Dy , ^{177}Hf , and ^{151}Eu have all been identified and studied as candidate burnable poisons.

For this analysis, a 3,400-MWth pressurized water reactor (PWR) with a 17 x 17 array of fuel rods per assembly was modeled. Cases of BP loading in 8, 16, 64, and 104 fuel rods were studied in four configurations:

- BP homogeneously mixed with fuel
- BP mixed in the outer one third of the fuel pellets
- A thin coating of BP on the outside surface of the fuel pellets
- BP metal alloyed with the cladding

A fuel enrichment of 4.5 percent was used for most cases in an effort to achieve a four-year fuel cycle, although an enrichment of 6 percent was studied in a few cases to increase the length of the fuel cycle.

Three-dimensional neutronics calculations were performed using a sequence of the MCNP4C, Tally, and Origen2 codes in order to generate fine group fluxes and cross sections. The resulting zone-dependent fluxes and cross sections were then used to generate power and burn-up distributions.

A fuel cycle length of four years was selected for study with the residual negative reactivity at the end of the cycle of primary interest. This residual absorber penalty (RAP), expressed in terms of days of operation, was calculated for each isotope and configuration of interest. By comparing the RAP for naturally occurring elements and separated isotopes, promising isotopes were

identified. Figure 1 shows a plot of negative reactivity due to the BP at the beginning of life (BOL) as a function of the RAP for the case of Gd in 16 fuel rods.

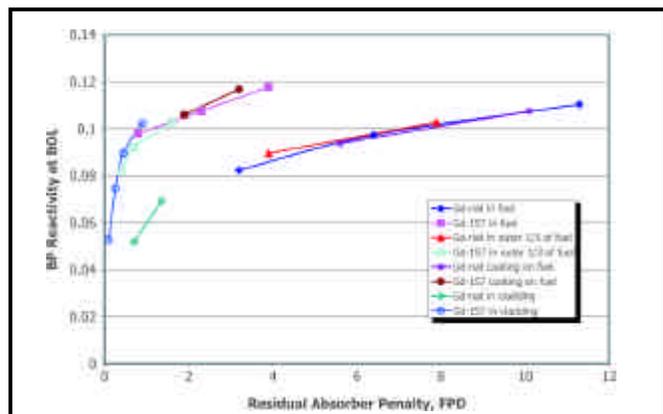


Figure 1. The graph illustrates negative reactivity from burnable poison at beginning of life as a function of the residual absorber penalty in full power days. The case is for Gd in 16 fuel rods in four configurations.

For an initial negative reactivity of $-0.1 \delta k/k_{\text{eff}}$, the difference between the two families of curves indicates a savings of six days of operation by the use of Gd-157 instead of natural Gd. This would achieve a savings of approximately \$6 million. Similar analyses indicate that Sm-149 results in a savings of 44 days and Er-167 in a savings of 36 days.

An interesting result seen from Figure 1 is that the RAP for a given initial negative reactivity is independent of the configuration for a given number of fuel rods. The initial reactivity is, of course, strongly dependent upon the configuration due to self-shielding effects.

The RAP is not the only parameter of interest. Selection of a burnable poison depends upon such properties as the burn-out rate and the initial negative reactivity. A BP that burns out within the first month would be nearly useless, and a very high negative reactivity could result in a positive void coefficient. The burn-out rate is illustrated in Figure 2 in terms of the reactivity remaining at 120 days, one year, and four years. It can be seen that natural Gd is satisfactory, although improved by isotope separation. Sm becomes promising when only Sm-149 is present, and Er-167 also becomes a candidate. Although not shown in Figure 2, Er-167 has improved burnout time dependence and a lower RAP than Gd if it is mixed into only the outer one-third of the fuel.

Separation of Isotopes: The plasma separation process is to be used to demonstrate the separation of candidate

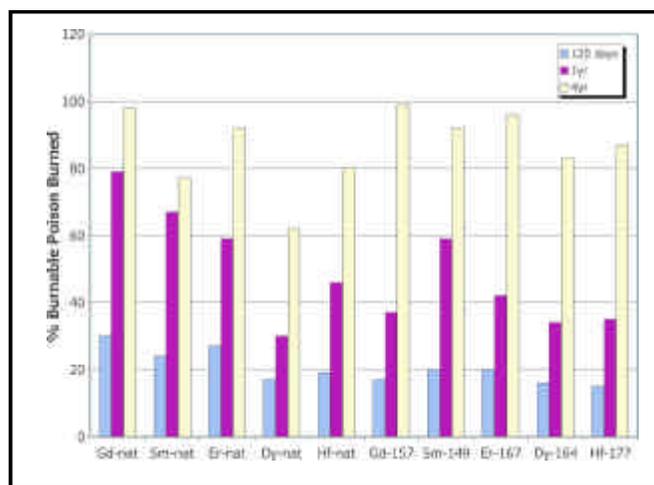


Figure 2. The time dependence of burnable poison depletion is shown, comparing naturally occurring elements and separated isotopes for the case of the BP mixed in the fuel as an oxide in eight fuel rods.

isotopes. This phase of the project was delayed because of unavailability of the separation plant, which is only now in the shake-down phase. However, fabrication of a gadolinium target for the plasma process is nearly complete. Ingots of approximately 20 kg each of Gd and Dy have been melted and cast into a large ingot. The ingot was sliced into 3.2 mm slabs by electro-discharge machining. The slabs were then welded into a single plate 0.61 x 0.58 m. This plate is now being plasma sprayed with copper on one side after which cooling coils will be soldered onto the copper-coated surface. This plate will then serve as the source target for the plasma separation process.

Compatibility of Absorber Materials: Considerable work has already been done by other investigators on the compatibility of rare earth oxides, especially gadolinium oxide, with uranium oxide. This work has identified reduction in thermal conductivity as the primary disadvantage in incorporating burnable poisons with fuel. Separation of isotopes permits lower concentrations of burnable poisons to be used, and thus tends to ameliorate the problem.

As was previously mentioned, this study suggests incorporating burnable poison in cladding as one option. Making a homogeneous alloy is generally easier than blending a ceramic, and incorporating the burnable poison in the fuel cladding, where the thermal flux is higher than in the fuel, permits faster and more complete burn-up. A patent disclosure for this invention has been filed (DOE Docket S-99,217; UT Battelle Docket ID 1044). Several scoping alloys of zirconium and Gd, Dy, Sm, and Er have

been prepared. Bend tests have indicated no serious embrittlement resulting from the introduction of the rare earths. Larger heats of Zircaloy-rare earth alloys have been prepared in anticipation of corrosion testing.

Planned Activities

The actual separation of isotopes is a major part of the project that has been seriously delayed because of unavailability of the plasma separation plant. This was a known uncertainty from the beginning, but the plant is now partially operational. Because of this delay, a no-cost extension of the project through June 2003 has been

approved by the DOE. Preparations of a target proved to be far more difficult than anticipated, but a gadolinium target is now nearing completion. Methods developed for gadolinium are expected to apply to other rare earths, although time and funds may preclude all but perhaps one additional element. A separation run is expected to be made in the next two months.

The Zircaloy-rare earth alloys will be fabricated into the proper specimens for corrosion testing. Preparations are being made for high -pressure water and steam corrosion testing.

NUCLEAR ENERGY RESEARCH INITIATIVE

Fuel for a Once-Through Cycle - (Th,U)O₂ in a Metal Matrix

Primary Investigator: Sean M. McDevitt, Argonne National Laboratory

Collaborators: Purdue University

Project Number: 99-095

Project Start Date: August 1999

Project End Date: December 2002

Research Objectives

This project seeks to combine the advantages to be gained from metal-matrix cermet nuclear fuel with the resource-extension potential of the thorium oxide fuel cycle and the inherent proliferation resistance of mixed oxide ceramics. The approach involves fuel pins containing (Th,U)O₂ microspheres dispersed in a zirconium metal matrix that can achieve high burn-up and be directly disposed in a once-through fuel cycle. The beneficial aspects of the high conductivity fuel may also enable fuel assembly and reactor designs that support advanced boiling or supercritical water concepts. These advantages fit well with the Department of Energy's focus on the development of Generation IV nuclear power systems and proliferation-resistant fuel cycles.

Research Progress

Figure 1 shows a diagram of the proposed cermet nuclear fuel concept. Cermet fuels have demonstrated the ability to enhance fuel performance and reactor safety because their high-conductivity matrix maintains low internal temperatures, which restrains fuel performance limiting phenomena and minimizes stored energy in the fuel pins. The combination of these benefits with the inherent proliferation resistance, high burn-up capability, and favorable neutronic properties of the thorium fuel cycle produces intriguing options for advanced nuclear fuel cycles. The fuel "meat" is composed of a fine dispersion of (Th,U)O₂ microspheres that have a theoretical density between 70 percent and 99 percent and a uranium enrichment below 20 percent U-235. Nominal values for the microsphere diameter, ThO₂-to-UO₂ ratio, fuel-to-matrix ratio, and U-235 enrichment were selected as approximately 50 μm, 50:50, 50:50, and approximately 19.5 percent, respectively, to provide guidance for the calculation and experimental activities carried out within

the project. Important project achievements include (1) simulations of the core design and fuel cycle, (2) creation of a detailed thermal model for cermet fuels, (3) establishment of laboratory-scale fabrication equipment for (Th,U)O₂ microspheres by spray drying and sintering, and (4) establishment of laboratory-scale fabrication equipment for the powder-in-tube drawing of cermet rods.

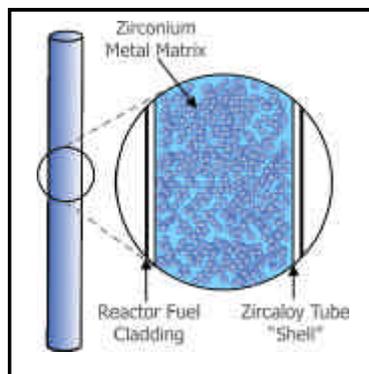


Figure 1. The schematic is a conceptual sketch for the (Th,U)O₂ Dispersion Fuel Pin.

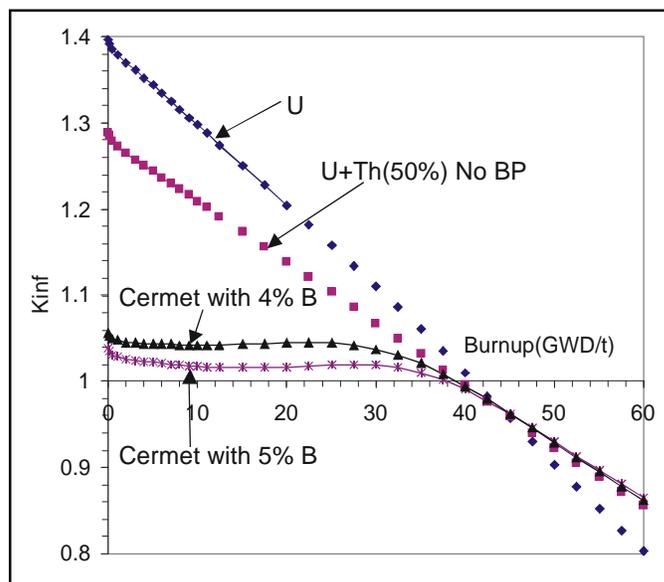


Figure 2. The graph illustrates inverse multiplication ($1/M$) plot for the first Burn-up Credit critical assembly. The final fuel rod configuration for this core is shown in the photographic insert.

Neutronics calculations were performed using the commercial lattice physics code HELIOS from Studsvik/Scandpower. Benchmarking calculations with Monte Carlo burn-up codes were used to demonstrate that HELIOS was effective for modeling thorium fuel lattices. The initial baseline simulations of the (Th,U)O₂ cermet used variations of the nominal fuel description (above) to generate comparisons with typical reactor fuels and to perform parametric studies. An increased-diameter ("fat") fuel pin in a tight lattice boiling water reactor (BWR) had a discharge burnup of approximately 80 MWd/kgHM using a U-235 enrichment of about 10 percent in a pin cell calculation. A tight-pitch hexagonal lattice was then benchmarked and studied for a BWR core. Because of the substantial boiling in a BWR core, there is a natural hardening of the neutron spectrum with a corresponding increase in the fuel conversion ratio. Using the nominal (Th,U)O₂ cermet, the moderator-to-fuel area was reduced in the simulated core and the conversion ratio increased from approximately 0.6 to more than 0.9, which resulted in a substantial increase in the fuel burnup over the initial result.

One notable calculation that was made is illustrated in Figure 2. In this simulation using a standard 8x8 BWR lattice, burnable absorbers are designed into the fuel matrix (as either B₄C or ZrB₂) with a cermet composition of 40 vol. percent Zr and 60 vol. percent heavy metal. Two types of heavy metal loadings were examined, one with only uranium and the other with mixed 50 percent thorium and 50 percent uranium (10 percent U-235 enrichment). The net fissile pin enrichment was approximately 5 percent in both cases. Two results are shown with four of the special burnable absorber rods present in the 8x8 assembly; these rods were doped with boron in the zirconium metal matrix at concentrations of 4 and 5 atom percent. The use of 4 rods at 5 percent boron very nearly controls all the excess reactivity in the assembly with no significant penalty to the achievable burn-up. This is an important result which may enable autonomous control in future reactor designs.

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 contributes to severe accident mitigation and a low fuel failure rate. A detailed thermal model has been developed based on the effective conductivity across an ideal interface to simulate the behavior of cermet fuel.

Spray drying is a physical process for granulating fine powders that is widely used in the chemical, pharmaceutical, ceramic, and food industries. The spray



Figure 3. Components of the Spray Drying Process are pictured: (a) Photograph of the spray drying system in its enclosure with a HEPA filtration system, and (b) Electron micrograph of spray-dried (U,Th)O₂ microspheres sintered at 1,650°C for 10h in flowing Ar-5 percent H₂.

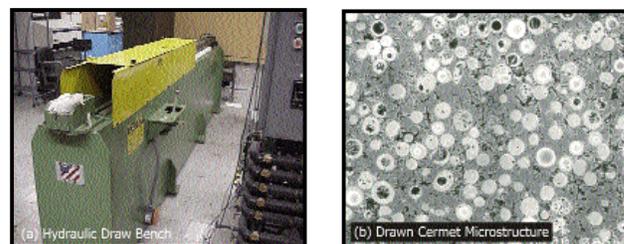


Figure 4. Components of the Powder-in-Tube Fabrication Process are shown: (a) Photograph of the hydraulic draw bench and (b) Electron micrograph of tungsten carbide microspheres in a zirconium matrix fabricated by powder-in tube drawing.

drying system (Figure 3a) is a commercial laboratory-scale spray dryer made by Niro Inc., and it consists of a funnel-shaped chamber approximately 1m in diameter with an insulated stainless steel double wall. Parametric studies were carried out to formulate stable, dense, and homogeneous aqueous slurries of urania and thoria powders for the production of microspheres. The parameters studied include (a) particle size distribution after ball-milling; (b) slurry viscosity; (c) zeta potential; (d) slurry flowability, stability and cleanability; (e) microsphere green strength; and (f) effects of organic dispersants. In an initial spray drying run, a slurry of (U,Th)O₂ was prepared by wet-milling fine urania and thoria powders and 0.5 volume percent triethanolamine for 24 hours (hydrochloric acid was used to adjust the pH of the slurry to 3 and the volume of the slurry was about 50 ml). A representative picture of the final sintered microspheres is shown in Figure 3b.

The powder-in-tube fabrication method is being developed as a simple low-temperature alternative to elevated temperature methods. In this process, oxide microspheres (50 to 1,000 μm diameter) and zirconium metal powders (~ 44 μm nominal diameter) are dry-mixed and loaded into stainless steel or Zircaloy drawing tubes and vibratory-packed. The powder-containing tube is drawn through a die to reduce the diameter and compact the powder into a dense matrix. The metals are annealed between 500°C and 1,000°C to remove strain hardening and strengthen interfacial bonding. Multiple cycles are used with sequentially decreasing die sizes to achieve complete densification. A hydraulic draw bench, manufactured by Fenn, has been installed in a radioactive materials lab (Figure 4a) alongside a materials research furnace. Initial fabrication demonstrations were completed using non-radioactive surrogate materials and a representative cermet cross-section is shown in Figure 4b.

Planned Activities

This project is nearly complete. Remaining efforts will include completing the neutronic modeling activities and fabricating a representative cermet fuel pin using spray dried (Th,U)O₂ microspheres. The modeling work is focussed on applying the unique advantages of this novel fuel form toward high-conversion reactor concepts including BWR, pressurized water reactor, reduced moderator water reactor, and supercritical light water reactor systems. The (Th,U)O₂ microspheres have been manufactured and the final product is scheduled to be drawn in November 2002.

NUCLEAR ENERGY RESEARCH INITIATIVE

Fundamental Mechanisms of Corrosion of Advanced Light Water Reactor Fuel Cladding Alloys at High Burn-Up

Primary Investigator: Randy G. Lott, Westinghouse Electric Company LLC

Project Number: 99-128

Collaborators: Pennsylvania State University; Argonne National Laboratory (West); Idaho National Engineering and Environmental Laboratory

Project Start Date: August 1999

Project End Date: January 2003

Research Objectives

The corrosion behavior of nuclear fuel cladding is a key factor limiting the performance of nuclear fuel elements. Improved cladding alloys, which resist corrosion and radiation damage, will facilitate higher burn-up core designs. The objective of this study is to understand the mechanisms by which alloy composition, heat treatment, and microstructure affect corrosion rate. This knowledge will be used to predict the behavior of existing alloys outside the current experience base (for example at high burn-up) and predict the effects of changes in operational conditions on zirconium alloy behavior.

Zirconium alloys corrode by the formation of a highly adherent protective oxide layer. The working hypothesis of this project is that alloy composition, microstructure, and heat treatment affect corrosion rates through their effect on the protective oxide structure and ion transport properties. Therefore, particular emphasis has been placed on detailed characterizations of the oxides formed on a series of experimental alloys. The experimental task in this project is to identify these differences and understand how they affect corrosion behavior. To do this, several microstructural examination techniques are being employed, including transmission electron microscopy (TEM), electrochemical impedance spectroscopy (EIS), and a selection of fluorescence and diffraction techniques using synchrotron radiation at the Advanced Photon Source (APS).

Detailed characterizations of oxides are only useful if the observations can be linked to the corrosion behavior of the alloy. That link requires a model of the corrosion mechanism. The modeling effort is designed to organize the data from the characterization studies in a self-consistent manner and link those observations to the corrosion behavior. The ultimate objective of this study is

a linkage between the characterization and theoretical modeling efforts that will produce improved alloy specifications.

Research Progress

Collaboration among the various members of the research team has led to the development of innovative descriptions of the corrosion process. The most promising of these ideas have been built into the corrosion model. A relevant set of alloys has been selected that bracket the expected range of corrosion behavior and that will be used both for the experimental characterization and for the modeling effort. The testing has been expanded to include oxide produced in steam and Li environments.

A systematic series of experiments have been performed at APS and the TEM characterization of oxides is continuing. Both microfluorescence compositional information and diffraction have been obtained using a unique x-ray microprobe facility, as shown schematically in Figure 1.

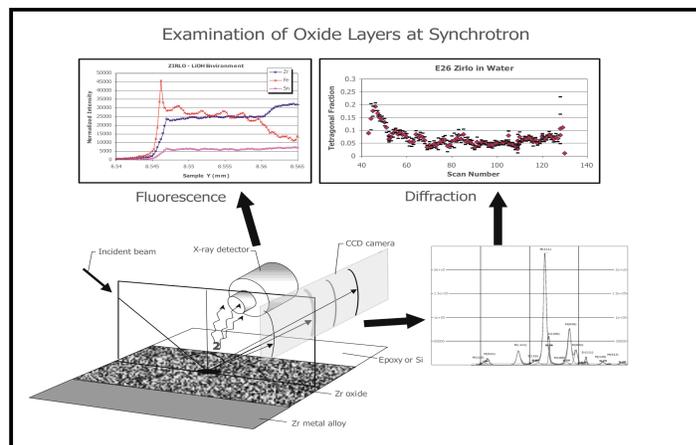


Figure 1. Oxide layers are examined in cross section at APS, and both microfluorescence and microdiffraction information can be obtained, with a resolution below 0.3 micron. Examples of real data obtained using this technique are shown in the graphs above.

Various oxides formed in various alloys and under three different environments have been characterized with respect to their tetragonal-to-monoclinic ratio and alloying element distribution in the oxide layer using synchrotron radiation. Cross-sectional TEM samples have been examined and more detailed work will be performed that will serve as complementary information to the synchrotron work.

The initial certification and experiments with EIS have been completed and data has been collected. The EIS results are consistent with the existence of a porous outer oxide and a dense, insulating inner oxide. The results have been analyzed in an attempt to understand the electronic differences in the two layers. The noted variations did not appear to be significant. Newly developed optical techniques have proven to be more efficient at identifying the layered structures of the oxides. Therefore, the EIS studies have been scaled back in favor of more detailed TEM and APS evaluations.

The autoclave design for the electrochemistry and radiolysis studies was completed. However, in accordance with the modified program plan, no further development of this facility will be undertaken in Year 3 of the Program.

The model engine, which connects the various model components and tracks the evolution of both the continuous and porous oxides, has been developed. This engine also includes a hydrogen accumulation mechanism. The engine has been developed as a Visual Basic Application, which writes the output directly into an Excel spreadsheet.

A schematic of the modeling process is indicated in Figure 2. Oxide growth is simulated using suitably small time steps. The environmental factors controlling the growth rate and critical thickness are calculated at each increment. These factors are then used as inputs to the growth rate and critical thickness modules. The

continuous oxide growth rate and the critical thickness are fed into the model engine. The basic model engine is used to determine the oxide thickness as a function of time. The engine incrementally increases the continuous oxide thickness and then checks to see if the thickness exceeds the critical value. When the continuous oxide measurement exceeds the critical value, the entire thickness is added to the porous oxide thickness and the continuous oxide thickness is returned to zero. The model is structured to assure consistency in the inputs to all modules. A single set of external inputs, describing the alloy and the reactor operating conditions, is used for all modules.

The primary task in designing an advanced alloy is understanding how each of the potential alloy constituents affect the corrosion behavior. Both the composition and the heat treatment of the material determine the corrosion behavior of the alloy. Therefore, it is important to understand how the microstructure of the alloy controls oxide formation. Factors controlling corrosion have been identified as the fraction of the alloying element precipitated, and the precipitate size. Lessons learned from the corrosion model have provided insight into these processes. As more is learned about the behavior of each alloying element, it should become possible to use the model as a tool for designing new alloys.

Planned Activities

During the final quarter of Budget Year 2, preparatory work has been undertaken for several new initiatives. A change in the workscope was negotiated to allow TEM examination of the irradiated specimens at Argonne National Laboratory. The APS runs have produced a large volume of diffraction data that is still being analyzed. Revisions to the model that will more accurately describe the oxidation of second phase particles within the growing oxide film are being developed.

In the final Budget Year of the project, it is expected that the combined knowledge derived from the various parts of the project will allow the design of new alloys for extended burn-up applications. Basing the design of the new alloys on mechanistic knowledge will be a big step forward to ensure reliable fuel operation at high burn-up rates.

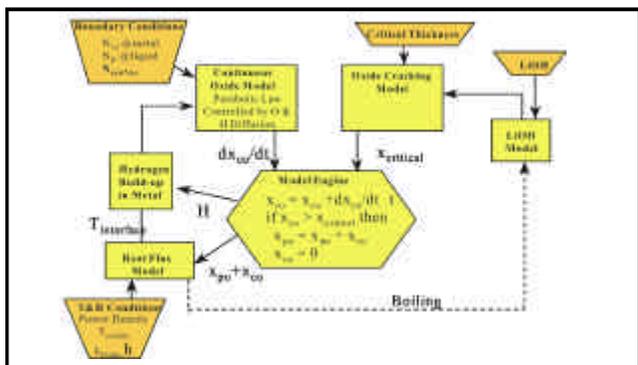


Figure 2. The schematic shows the application of the model to calculating oxide thickness.

NUCLEAR ENERGY RESEARCH INITIATIVE

Advanced Proliferation-Resistant, Lower-Cost, Uranium-Thorium Dioxide Fuels for Light Water Reactors

Primary Investigator: Philip E. MacDonald, Idaho National Engineering and Environmental Laboratory (INEEL)

Project Number: 99-153

Project Start Date: August 1999

Project End Date: August 2003

Collaborators: Argonne National Laboratory; University of Florida; Framatome ANP; Korea Atomic Energy Research Institute; Massachusetts Institute of Technology (MIT); Purdue University; Westinghouse Electric Corporation

Research Objectives

The overall objective of this NERI project is to evaluate the efficacy of high burn-up mixed thorium-uranium dioxide ($\text{ThO}_2\text{-UO}_2$) fuels for light water reactors (LWRs). A mixed thorium-uranium fuel that can be operated to a relatively high burn-up level in current and future LWRs may have the potential to improve fuel cycle economics (allow higher sustainable plant capacity factors), improve fuel performance, increase proliferation resistance, and be a more stable and insoluble waste product than UO_2 .

The project has been organized into four tasks:

- (1) Task 1 will consist of fuel cycle neutronics and economics analysis to determine the economic viability of a $\text{ThO}_2\text{-UO}_2$ fuel cycle in PWRs.
- (2) Task 2 will determine whether or not $\text{ThO}_2\text{-UO}_2$ fuel can be manufactured economically.
- (3) Task 3 will evaluate the behavior of $\text{ThO}_2\text{-UO}_2$ fuel during normal, off-normal, and accident conditions and compare the results with the results of previous UO_2 fuel evaluations and U.S. Nuclear Regulatory Commission (NRC) licensing standards.
- (4) Task 4 will determine the long-term stability of $\text{ThO}_2\text{-UO}_2$ waste.

Research Progress

Progress made on each of the tasks will be described in turn in this section.

Task 1 - Reactor Core Analysis and Fuel Cycle Design: Due to the relatively poor economic performance of the homogeneously mixed uranium-thorium fuel, the focus of the physics work under Task 1 during Year 2 and the first part of Year 3 was primarily on the performance and economics of using micro-heterogeneous fuel forms, where some small distance physically separates the uranium and thorium. When compared to the equivalent homogeneous case (i.e., the same uranium-thorium weight percentages), an increase in burn-up is observed, which improves the economics of using thorium-based fuels. However, the economic improvement due to the use of any of the various micro-heterogeneous fuel forms is not sufficient to compensate for the costs of the increased Separative Work Units (SWUs) required for thorium oxide fuels. Therefore, the work at Framatome, INEEL, and MIT during most of Year 3 has focused on use of thorium to burn unwanted reactor or weapons grade plutonium.

Framatome ANP has completed the design of a weapons grade plutonium-thorium oxide-fueled core. Three different fuel pin loadings of plutonium are used to minimize the power peaking in the corner pins and in the pins near the water holes. The design meets current thermal-hydraulic and safety criteria. A lifetime depletion calculation was completed and the plutonium-thorium fuel design was found to be as reactive as plutonium-uranium cores. However, in the case of the plutonium-thorium core, less than 10 percent of the original plutonium inventory remains at 50,000 MWd/MtHM. The analysis of selected reactor transients demonstrated that it is feasible to license and operate safely a reactor fueled with plutonium-thorium blended fuel. In most cases analyzed,

the thorium mixture had less severe consequences than those for a core comprised of Low Enriched Uranium (LEU) fuel. In the analyzed cases where the consequences were more severe, they were still within acceptable limits.

The INEEL has completed analyses of a plutonium-thorium oxide-fueled core with every other fuel rod LEU UO_2 and plutonium-thorium oxide. These preliminary analyses have shown that the combination of thorium, recovered uranium, and reactor grade plutonium in an oxide fuel for light water reactors can be an effective way to consume reactor grade plutonium and to transmute the remaining plutonium isotopes into a mixture that is more proliferation-resistant, while still allowing the reactor reactivity control systems to remain similar to that for UO_2 cores.

MIT researchers benchmarked the analysis tools CASMO4 and MCODE against the IAEA standard problem for analysis of plutonia-thoria lattices. They then used those codes to perform a comprehensive study of the reactor grade plutonium destruction capabilities of homogeneously mixed PuO_2 - ThO_2 fuels in LWRs as a function of H/HM ratios. For the un-denatured cases, up to 1,000 kg of plutonium can potentially be destroyed per GWe-yr. The residual plutonium fraction (relative to the initially loaded Pu) in discharged fuel can be minimized by increasing the H/HM ratio and can potentially be as low as 25 percent. However, denaturing of mixed plutonium-thorium oxide fuel impairs the plutonium destruction effectiveness by about 20 percent. This penalty can also be minimized by increasing the H/HM ratio. The results of the reactivity coefficient evaluations indicate that mixed plutonium-thorium oxide fuel can be used for plutonium disposition in conventional PWRs with some changes in reactor reactivity control systems. The Doppler coefficient, boron worth, and β_{eff} of the plutonium-thorium oxide fuel are of about the same order as plutonium-uranium oxide fuel values.

Task 2 - Fuel Manufacturing Costs: This task was organized into three major activities: an engineering study of the feasibility of producing the thorium-uranium fuel in current nuclear fuel production facilities, an effort to estimate the cost of fabricating ThO_2 - UO_2 oxide fuel, and a developmental effort to make fuel pellets with appropriate densities and to use this material to determine fundamental heat transfer properties to use in the modeling efforts. The Westinghouse Electric Company completed the first two tasks at the end of Year 2 and the results were reported in the 2nd Annual Progress Report. Purdue University, with help from Westinghouse, is

continuing to evaluate the fabrication issues associated with co-precipitation of the powder and with pressing, sintering, and grinding ThO_2 - UO_2 fuel pellets. Purdue University is also tasked with making various material measurements to support the modeling efforts.

Task 3 - Fuel Performance: This task is providing tools to evaluate the thermal, mechanical, and chemical aspects of the behavior of ThO_2 - UO_2 fuel rods during normal, off-normal, and design basis accident conditions. During Year 3, MIT has been developing a version of the transient code FRAP-T6 for analyses of ThO_2 - UO_2 fuel behavior during a Reactivity Initiated Accident (RIA) event. Modifications to FRAP-T6 included the addition of thoria fuel properties (heat capacity, thermal expansion, thermal conductivity); a low temperature burst stress model; and the gaseous swelling contribution to the cladding strain. The available high burnup UO_2 fuel tests have been analyzed and the modified FRAP-T6 seems to reasonably predict the residual cladding strains in the tests. Analyses of ThO_2 - UO_2 fuel in a typical LWR indicate that it will tend to perform better than UO_2 fuel under RIA event conditions due to its lower thermal expansion and flatter power distribution in the fuel pellet (less power and less fission gas in the rim region).

During Year 2, MIT researchers calculated that the most promising micro-heterogeneous thoria-uranium arrangement with respect to achievable burnup is the axial micro-heterogeneous design with UO_2 and thorium section lengths of 4 and 8 cm, respectively. This design increases the fuel discharge burnup by a significant amount over the UO_2 base case, about 13 percent to 15 percent. In addition, this design offers the benefit of a substantial reduction of poison to compensate for the reactivity excess at beginning-of-life. Although this design manifests appreciable neutronic advantages, the absence of fissile material in the ThO_2 section at beginning-of-life results in large local power peaking. The most effective way to reduce the local peaking is to add uranium with fissile U-235 into the ThO_2 section. However, because homogeneous mixing of uranium in the thorium slug significantly impairs the reactivity-limited burnup performance, a modified axial and radial micro-heterogeneous fuel pin design (DuUAX4) was developed by introducing a 25 vol percent central void in the UO_2 driver zone and moving the extra UO_2 into the blanket zone as an inner ring with ThO_2 as an outer ring.

During Year 3, calculations were performed at the INEEL to compare the temperature behavior of DuUAX4

and conventional 100 percent UO_2 fuel rods during a large break LOCA. The calculations were performed with the SCDAP/RELAP MOD3.3 code extended for the analysis of ThO_2 - UO_2 fuel rods and extended for the modeling of axial heat conduction. A solution scheme for temperature involving either a fixed or moving fine mesh was added to the SCDAP/RELAP5 computer code. The fine mesh allows for mesh sizes in the axial direction as small as 1 mm (order of the thickness of the cladding). This small mesh size provides for an accurate calculation of cladding temperatures in the vicinity of the quench front or at the axial interface of a seed and blanket region in a fuel rod. It was found that the maximum cladding temperature of the DuUAX4 fuel during a LOCA is not significantly greater than that in conventional 100 percent UO_2 fuel.

Task 4 - Long Term Stability of ThO_2 - UO_2 Waste: This research is focused on measuring uranium dissolution from (U,Th) O_2 solid solutions as a function of the uranium content to determine the degree to which the mixed oxide is superior to UO_2 as a waste form. Dissolution studies on irradiated and unirradiated (U,Th) O_2 pellets and pellet fragments are underway. Irradiated fuels under investigation range in composition from 2 to 5.2 percent UO_2 . This work is being conducted at Argonne National Laboratory East (ANL E). The experiments on the unirradiated fuels at the University of Florida involve compositions of from 5 to 50 percent UO_2 . Dissolution behavior is being studied in J13 well water at both 90°C and room temperature. The results of this study will include (1) comparison of the dissolution behavior of irradiated and unirradiated fuel (nominal composition of 5 percent UO_2) to determine the effect of fuel burnup on dissolution and (2) the effect of solid solution composition on the dissolution behavior of the unirradiated fuel. Studies of the dry oxidation behavior of (U,Th) O_2 are also continuing. The experiments involve gravimetric analysis of sample powders heated in an oxidizing or reducing environment. The results will show the oxidation behavior of (U,Th) O_2 as a function of the solid composition, and give an indication of the effect thorium has on the oxidation of uranium.

Task 5 - Korean Work: The Koreans have been working on four tasks: core design analyses, fuel pellet manufacturing technologies, fuel rod performance analysis, and xenon diffusivity measurements. In the area of core design

analyses, the Koreans completed their analysis of the mixed core concept of duplex (Th,U) O_2 and UO_2 fuels. The uranium ore and SWU costs per MWD for the mixed core of thorium-uranium oxide with uranium oxide fuel are improved over the costs for a homogeneous thoria-uranium core. However, even with a long fuel cycle scheme, the thorium-based mixed core concept does not show a superior potential in fuel economics to an all-uranium oxide core. To further improve their fuel pellet manufacturing technologies, three kinds of pellets (ThO_2 , ThO_2 -35% UO_2 , ThO_2 -65% UO_2) have been fabricated by powder processing. However, good (Th,U) O_2 pellet homogeneity could not be achieved using dry milling and a wet mill process was developed. The resulting (Th,U) O_2 pellets had a density greater than 95 percent TD and good homogeneity.

In the area of fuel rod performance, the INFRA-Th computer code was developed by adding the thermal conductivity, radial power, and burn-up distribution, and thermal expansion models for ThO_2 - UO_2 fuel to the UO_2 performance analysis code, INFRA. Also, a ThO_2 - UO_2 irradiation test, called IFA-652.1, was started in June 2000 in the Halden Reactor. The test rod is instrumented with a thermo-couple and pressure transducer. The fuel centerline temperature and rod internal pressure data from IFA-652.1 has been compared with predictions from the INFRA-Th computer code. Four xenon diffusivity measurement experiments have been completed during Year 3 using natural UO_2 . The diffusion coefficients measured in this work for xenon in UO_2 are consistent with the published literature. Work with ThO_2 - UO_2 fuel will start shortly.

Planned Activities

All of the planned work is complete in Tasks 1, 2, and 3 of this NERI project except for the thoria-uranium fuel property measurements that were to have been done by Purdue University. It should be noted that significantly more work was done in Task 1 by Framatome ANP and MIT than was originally proposed and planned. The waste studies in Task 4 at the University of Florida and ANL are on schedule for completion by the end of Year 3. The work in Korea is not directly funded by the Department of Energy; however, all the work originally agreed to has been carried out as planned and is essentially complete.

NUCLEAR ENERGY RESEARCH INITIATIVE

A Proliferation-Resistant Hexagonal Tight Lattice BWR Fuel Core Design for Increased Burn-up and Reduced Fuel Storage Requirements

Primary Investigator: Hiroshi Takahashi,
Brookhaven National Laboratory

Project Number: 99-164

Collaborators: Purdue University; Korean Atomic Energy Research Institute (KAERI); University of Mining & Metallurgy (Poland)

Project Start Date: August 1999

Project End Date: September 2002

Research Objectives

The major objective of this project is to advance the well-developed, water-cooled reactor technology in order to make efficient use of the abundant thorium resource available in the earth's crust. Considerable effort has been invested in development of the sodium-cooled fast reactor to breed fissionable ^{239}Pu from natural uranium. Much less effort has been expended into development of alternative technologies to safely and efficiently make use of the thorium resource. This project will investigate the feasibility of a plutonium-thorium (Pu-Th) fuel cycle for a new type of high conversion reactor cooled by boiling water (HCBWR). The technology will be developed to burn existing stocks of plutonium, while converting the fertile thorium to fissile ^{233}U . The high conversion rate will take place in a fast neutron spectrum through the design feature that minimizes the volume of water in very tight fuel assembly lattices. A segmented core design will be used, consisting of multiple radial and axial zones, and the design will burn the plutonium and will convert thorium into the more proliferation-resistant ^{233}U . High fuel burn up will be possible as a result of the continuous generation and fission of ^{233}U as the plutonium is consumed. Inherent safety will be designed into the reactor through the use of thorium as fertile material. This will introduce a negative void coefficient for those accident sequences which result in off-normal coolant boiling.

Thus, the major technical objective of the proposed project is to develop a reactor design which will accomplish the following:

- Minimize the potential for proliferation of weapons grade fissionable materials
- Maximize the inherent safety features of the reactor

- Maximize the achievable fuel burn up and plant capacity factor
- Minimize the cost of electricity generation

The research and development program proposed here will satisfy the major objectives of the NERI program:

- (1) Advance the state of nuclear technology through introduction of an alternative, proliferation resistant, new type of fast reactor
- (2) Advance the state of nuclear technology through extending the well-developed boiling water technologies
- (3) Maintain a nuclear science and engineering capability through use of major nuclear industry reactor design and safety analysis tools during the design and safety analysis of the project
- (4) Improve the safety performance of fast reactors through use of potentially safer water, which is not susceptible to combustion in air as is sodium
- (5) Improve the safety of fast reactors as a result of using thorium fertile material to provide a negative void reactivity coefficient.

Research Progress

Research has focused on neutronics design analysis of a HCBWR with Pu-Th fuel.

One of the primary innovative design features of the core proposed here is the use of thorium as fertile material. In addition to the advantageous nonproliferation and waste characteristics of thorium fuel cycles, the use of thorium is particularly important in a tight-pitch, high-conversion lattice in order to ensure a negative void

coefficient throughout the operating life of the reactor.

The principal design objective of a high-conversion light water reactor (LWR) is to substantially increase the conversion ratio (fissile atoms produced per fissile atoms consumed) of the reactor without compromising the safety performance of the plant. Since existing LWRs have a relatively low conversion ratio, they require relatively frequent refueling, which limits the economic efficiency of the plant. Also, the high volume of spent fuel can pose a burden for waste storage and the accumulation of plutonium in the uranium fuel cycle can become a materials-proliferation issue. The development of fast breeder reactors (FBRs) as an alternative technology to alleviate some of these concerns has been delayed for various reasons. An intermediate solution has been to examine tight pitch LWRs that can provide significant improvements in the fuel cycle performance of the existing LWRs by taking advantage of the increased conversion ratios from the harder neutron spectrum in the tight pitch lattice, as well as by taking advantage of the waste and nonproliferation benefits of the thorium fuel cycle.

Several HCBWR designs have been proposed by researchers in Japan and elsewhere during the past several years. One of the more promising high-conversion reactor (HCR) designs is the reduced moderation water reactor (RMWR) proposed by The Japan Atomic Energy Research Institute (JAERI). Their design was based on a uranium fuel cycle and showed significant improvements in the fuel cycle performance compared to conventional BWRs. However, one of the drawbacks of their design was the potential for a positive void coefficient. In order to ensure a negative void coefficient, the JAERI researchers designed a "flat core" and introduced void tube assemblies in order to enhance neutron leakage in the event of core voiding. The use of thorium in the Purdue/BNL HCBWR design proposed here obviates the need for void tubes and makes it possible to increase the core height and improve neutron economy without the risk of a positive void coefficient. The principal reason for the improvement in the void coefficient is that Th-232 has a smaller fast fission cross section and resonance integral than U-238. In the design proposed here, it is possible to eliminate the void tubes in the RMWR design and replace the axial blanket with active fuel to increase the core height and further improve neutron economy.

The core analyses in the work here were performed with the Purdue Fuel Management Code System which is based on the Studsvik/Scandpower lattice physics code HELIOS, and the U.S. NRC core neutronics simulator,

PARCS, which is coupled to the thermal-hydraulics code RELAP5. All these codes have been well-assessed and benchmarked for analysis of LWR systems.

The HCBWR developed here is characterized by a very tight lattice with a relatively small water volume fraction in the core, which therefore operates with a fast reactor neutron spectrum, and a considerably improved neutron economy compared to the current generation of LWRs. A tight lattice BWR core has a very narrow flow channels with a hydraulic diameter less than half of the regular BWR core. The tight lattice core presented a special challenge to core cooling, because of reduced water inventory and high friction in the core. The primary safety concern when reducing the moderator to fuel ratio and when using a tightly packed lattice arrangement is to maintain adequate cooling of the core during both normal operation and accident scenarios.

In the HCBWR design, the core has been placed in a vessel with a large chimney section, and the vessel is connected to the isolation cooling system (ICS). The vessel is placed in containment with a gravity driven cooling system (GDCS) and a passive containment cooling system (PCCS) in a configuration similar to General Electric's Simplified Boiling Water Reactor (SBWR). The safety systems are similar to SBWR; ICS and PCCS are scaled with power. An internal recirculation pump was placed in the downcomer to augment the buoyancy head provided by the chimney. The buoyancy provided by the chimney alone could not generate sufficient recirculation in the vessel since the tight lattice configuration resulted in much larger friction in the core than the SBWR.

The modified RELAP5 was used to simulate and analyze two of the most limiting events for a tight pitch lattice core: the Station Blackout and the Main Steam Line Break events. The constitutive relationships for RELAP5 were compared with the correlations and the data available for narrow channels, and heat transfer package was modified for narrow channel application. The results of the analyses indicate that the HCBWR system will be safely shutdown for these transients.

Planned Activities

Evaluation of safety analyses will be continued with newly obtained core data using the RELAP5 code.

The economic and non-proliferation studies will be continued. The potential for transmutation of minor actinides using the hard neutron energy spectrum in HCBWR will be studied, as well as the deep-underground

concept, which provides a safer operation than the surface reactor.

A final evaluation report is being prepared with an overview of the HCBWR system.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of a Stabilized Light Water Reactor (LWR) Fuel Matrix for Extended Burn-up

Primary Investigator: Brady D. Hanson, Pacific Northwest National Laboratory (PNNL)

Collaborators: University of California, Berkeley

Project Number: 99-197

Project Start Date: August 1999

Project End Date: May 2003

Research Objectives

The main objective of this project is to develop an advanced fuel matrix based on the currently licensed UO_2 structure capable of achieving extended burn-up while improving safety margins and reliability for present operations. Burn-up is currently limited by fission gas release and the associated increase in fuel rod internal pressure, fuel swelling, and cladding degradation. Once fuels exceed a threshold burn-up, a "rim" or high burn-up structure (HBS) forms. The HBS is characterized by the development of a subgrain microstructure having high porosity and low thermal conductivity. It is believed that the lower thermal conductivity results in larger temperature gradients and contributes to subsequent fission gas release. Fuel designs that decrease the centerline temperature while limiting the HBS restructuring, thereby decreasing the fission gas release, should be able to achieve higher burn-up and even allow higher operating power for increased efficiency.

Research at PNNL has demonstrated that the soluble fission products and actinides present in the matrix of irradiated (spent) fuels stabilize the fuel matrix with respect to oxidation to U_3O_8 . The higher the soluble dopant concentration, the more resistant the fuel has been to restructuring of the matrix from the cubic phase of UO_2 to the orthorhombic U_3O_8 phase. It is hypothesized that such restructuring of the uranium planes within the matrix due to oxidation is similar to the microstructural changes that occur during HBS formation. In this project, the attempt is to utilize the changes in fuel chemistry that result from doping the fuel to design a fuel that minimizes HBS formation and is more resistant to corrosion if it is ever exposed to water or air. The use of dopants that can act as getters of free oxygen and fission products (e.g, Cs, I, Tc) to minimize fuel-side corrosion of the cladding or release from the fuel matrix is also being studied.

Other fuel designs, such as radial variations in enrichment, are being examined to further minimize HBS formation. The growth of large grains, which should reduce both HBS formation and fission gas release, using a steam oxidation process, is also being studied. A combination of experimental studies and theoretical modeling is being used to determine the optimal design.

Research Progress

Work on Task 1, Understand and Model HBS Formation, involves increasing the understanding of HBS formation in order to better design a fuel to delay or prevent its onset. A comprehensive literature review has been performed. While it is clear that no one fully understands how the HBS forms, it is becoming apparent that the accumulation of radiation damage is the main driving force. High burn-up fuels (>60 MWd/kg) have been examined at PNNL using Atomic Force Microscopy (AFM). These examinations have shown the subgrain microstructure similar to the HBS, even in the central regions of the fuel pellet where the local burn-up is below the threshold reported for bulk formation of the HBS. The temperatures near the fuel centerline are also high enough that radiation damage is readily annealed. However, the areas adjacent to pores seem to act as pinning sites for radiation damage and result in very localized restructuring. The large grain size for the advanced fuel being designed for this project should help delay HBS formation.

Most studies of HBS have utilized Electron Microprobe Analysis (EMPA) to determine concentrations of Xe, U, or Pu as a function of radial position in the fuel. In order to more fully understand the phenomena associated with HBS formation, this project has sponsored the continuing development of the Resonance Absorption Burn-up (RABURN) model at UC-Berkeley. The objective is to develop a simplified code that can, in a computationally

efficient and user friendly way, provide a good approximation for calculating the radial-dependence of the fission rate and actinide production in reactor fuels. The radial dependence of soluble dopants in the UO_2 matrix, local temperature, and extent of radiation damage are being calculated to provide the basis for the HBS and matrix stabilization models.

Development has continued of the one-dimensional code, RABURN. In particular, it has been possible to verify most of the fundamental assumptions in the code. These assumptions are that (1) the only relevant portions of the neutron flux incident at the fuel surface correspond to those associated with the quasi-thermal and the epithermal fluxes; (2) the ratio of the integrated thermal flux to the magnitude of the epithermal flux, assumed to be of the form ϕ_0/E , is about 0.25 to 0.35; (3) because of the $1/E$ variation of the epithermal flux, only the lowest-energy strong resonances need be considered explicitly; (4) because of their large mean free paths in the fuel, a neutron incident on the surface of the fuel would interact once or not at all in passing through a dimension of the fuel diameter; and, (5) the main resonances that should be considered are those in (even, even) actinides at low energies. These assumptions have all been verified by performing pin-cell calculations with the Monte Carlo code MCNP and a well-characterized spent fuel (ATM-103). RABURN has been modified to account for temperature- and burn-up-dependent thermal conductivities to calculate radial-dependent temperature distributions. The code is able to reproduce the radial-averaged concentrations of ^{235}U , ^{236}U , ^{238}U , ^{237}Np , ^{239}Pu , and ^{240}Pu to within ± 5 to 10 percent. However, the code still predicts Pu concentrations in the rim region will be higher by a factor of 2.7 compared to that measured. The results have also been more insensitive to parameter changes than would have been predicted. The reason for these discrepancies appears to be that the neutron flux depression at energies in the immediate vicinity of resonances in the fuel was accounted for, but the corresponding depression in the water immediately adjacent to the fuel was not.

Task 2, Develop Matrix Stabilization Model, has focused on determining the lattice parameter of both spent fuels and doped fuels produced by this project. While the team has a general understanding of how dopants stabilize the matrix with respect to oxidation, a model is being developed that accounts for these changes as a function of lattice parameter and the ionic radii and oxidation state of the dopants. Powder X-ray Diffractometry (XRD) has been used to determine the

phases and lattice parameters of the fuels tested. However, the sensitivity and precision using standard quantitative XRD were not sufficient for the modeling efforts. A whole pattern fitting/Reitveld refinement program has been procured and initial results indicate much less uncertainty in the re-analyzed data.

Task 3, Design and Test Advanced Fuel Matrix, is focused on the dopant concept. A thorough literature review was performed on the effect of dopants on the UO_2 matrix. Additionally, a lab-scale fuel fabrication laboratory was established at PNNL and UO_2 and doped- UO_2 pellets were produced. Initial efforts have concentrated on producing homogeneously doped pellets so the results from thermogravimetric analysis (TGA) studies to examine oxidation response will be representative.

UO_2 powder has been procured from Framatome ANP. The UO_2 powder and the desired dopant are wet-milled for 24 hours to assure intimate mixing. Excess water is then removed by vacuum drying. Pellets are formed by first granulating and then pressing the mixed powder. The pellets are sintered in a 3 percent H_2 atmosphere at $1,650^\circ\text{C}$ for 24 hours. The pellets are weighed and measured using laser dimensional analysis, and the density is determined using gas pycnometry. The pellet densities have typically been approximately equal to 95 percent of the theoretical density. The fuel pellet is then subsectioned and examined using XRD to determine lattice parameter and to verify that a solid solution was obtained. Other sections of the pellet are examined using Scanning Electron Microscopy (SEM) with an EDAX Light Element Detector and backscattered imaging to look for areas of heterogeneous dopant/fuel mixtures. It appears that homogeneous solid solution pellets can be made successfully with dopant concentrations up to at least 10 wt percent. Examples of the XRD fits to pellets with 8 wt percent Gd_2O_3 and 10 weight (wt) percent ZrO_2 , respectively, are shown in Figure 1.

Planned Activities

The following activities are scheduled for the final year of this project:

- (1) RABURN will be modified to a true cylindrical geometry and any necessary approximations to the neutron flux depression in the water will be incorporated. RABURN will then be run to determine local dopant concentrations and temperatures for comparison with AFM data to

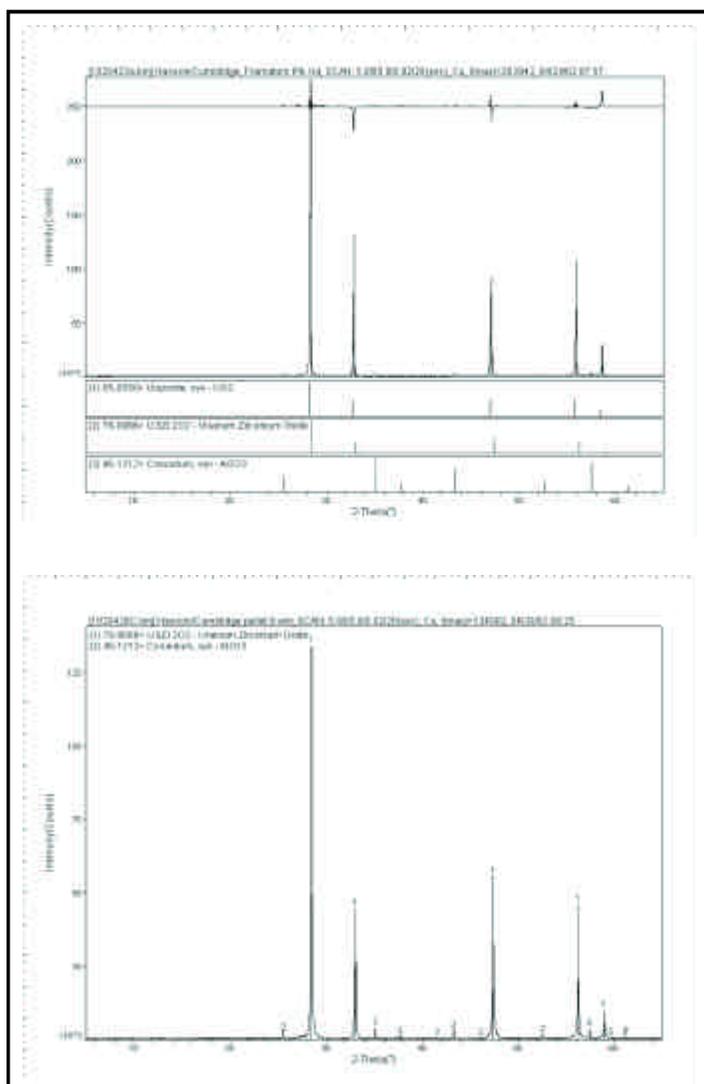


Figure 1. The schematics show XRD fits to 8 wt percent Gd_2O_3 (top) and 10 wt percent ZrO_2 (bottom) doped UO_2 pellets fabricated at PNNL. The Gd pellet is fit using Reitveld refinements of the theoretical lattice parameter with excellent results. The Zr pellet is fit exactly with an established card from the international database. There are no indications of peak broadening or separate phases. Corundum was added as an internal standard for XRD only.

allow complete development of the HBS formation model.

- (2) TGA will be performed on commercial pellets with various Gd-doping levels obtained from Framatome ANP. All pellets produced at PNNL will then be analyzed with TGA to determine the oxidation response. This data will be coupled with the lattice parameters to accurately develop the matrix stabilization model as a function of dopant, ionic radius, and oxidation state so the optimum fuel can be designed.
- (3) Additional fuels will be produced and tested for density, oxidation response, and lattice parameter. The thermal conductivities will be measured and the neutronic effects modeled.
- (4) Fuels with large grains, both with and without dopants, will be made using high-temperature steam oxidation.
- (5) It is hoped that a few candidate fuels will be trace-irradiated in a university research reactor so the fission gas release characteristics can be measured.

NUCLEAR ENERGY RESEARCH INITIATIVE

Continuous Fiber Ceramic Composite (CFCC) Cladding for Commercial Water Reactor Fuel

Primary Investigator: Herbert Feinroth, Gamma Engineering Corporation

Project Number: 99-224

Collaborators: Ceramic Composites Inc.; Massachusetts Institute of Technology (MIT); McDermott Technology, Inc.; Swales Aerospace Corporation

Project Start Date: August 1999

Project End Date: December 2000

SBIR Project Start Date: August 2001

Research Objectives

Existing commercial water-cooled nuclear reactors use zircaloy as the reactor structural material for fuel cladding. Zircaloy loses most of its strength over 1,000°F and reacts exothermically with water during low probability events such as loss-of-coolant accidents (LOCAs). As a result, all existing water-cooled reactor systems require complex and redundant safety systems, including rapid acting backup emergency power supplies, to guard against the energy release and hydrogen generation that could occur during a LOCA. The objective of this project is to develop ceramic composite fuel cladding material that behaves at least as well as zircaloy during normal operation, but retains its strength and avoids severe consequences during core overheating accidents such as LOCAs. Consequently, the safety systems of plants that use such cladding could be simplified and made more reliable, leading to more economic plant designs, simplified regulatory requirements, and higher levels of public acceptance.

conducted in which representative samples were quenched in water from temperatures ranging from 1,000°F to 2,500°F. The test specimens demonstrated no visible structural damage in the 1,000°F and 1,800°F tests, and the coated specimens showed minor damage, but no major failure, from the 2,500°F tests.

The maximum density achievable on the CFCC specimens, which were fabricated using sol-gel impregnation technology, was about 80 percent of theoretical density. Consequently, these initial specimens were permeable to fission gases, and not acceptable for reactor application.

Research Progress

In the NERI phase of this project, completed in April 2001, alumina-yttria-based, continuous fiber ceramic composite (CFCC) tubes of about the same diameter as light water reactor fuel cladding, were fabricated and tested in simulated reactor coolant conditions. A 50-day (1,200-hour), in-reactor irradiation and corrosion test in the MIT Research Reactor was conducted at typical pressurized water reactor coolant temperature and chemistry conditions. Results showed that this alumina-based CFCC cladding has good corrosion/erosion resistance under realistic reactor conditions.

A follow-on research project was approved by DOE as part of the DOE Small Business Research Initiative, designed to solve the permeability problem. This project, completed in February 2002, developed a hybrid ceramic composite, with an inner layer of high density monolithic silicon carbide, and an outer layer of CFCC impregnated via a Chemical Vapor Infiltration (CVI) process. Silicon carbide was used, rather than oxide based ceramics, because of its excellent high-temperature and thermal-conductivity properties, and because of its ready availability. Several processes were used in fabricating the hybrid structure, including different means of braiding and wrapping the fibers around the monolith in order to achieve good bonding and high strength. Photographs of the resulting product are shown in Figure 1.

All hybrid material specimens were tested and proven to be gas-impermeable up to 125 psi at room temperature. The fabricated specimens showed high matrix material density, significant mechanical strength, and a graceful failure mode when stressed to material fracture (i.e., showing a metal-like stress strain behavior over time).

Accident tests for simulated loss of coolant were

Planned Activities

The NERI and SBIR 1 research projects have been completed. It is believed that this hybrid ceramic composite cladding has significant potential for Generation IV reactor concepts that require cladding and structural materials to perform at higher temperatures than can be achieved by using traditional metallic cladding.

NUCLEAR ENERGY RESEARCH INITIATIVE

An Innovative Ceramic Corrosion Protection System for Zircaloy Cladding

Primary Investigator: Ronald H. Baney, University of Florida

Project Number: 99-229

Project Start Date: July 1999

Project End Date: January 2003

Research Objective

The operational lifetime of light water reactor (LWR) fuel is limited by thermal, chemical, and mechanical constraints associated with the operating conditions of the fuel rod assemblies. A primary limiting factor is the waterside corrosion of the Zircaloy cladding that encases the uranium oxide pellets. Oxidation of the cladding in the high-temperature, high-pressure aqueous environment results in the continuous formation of a zirconium oxide layer that eventually begins to spall, degrading the clad integrity. Protective coatings applied to the Zircaloy cladding can drastically reduce this corrosion and degradation. The objective of this NERI project is to develop ceramic corrosion protection systems for Zircaloy clad for use in LWRs that will allow significantly higher burn-ups resulting in major benefits in plant safety and plant economics. Additionally, the coatings will serve to protect the cladding from fretting failure, a very positive secondary effect.

The major technical challenge for coating a metal with a ceramic protection system is to develop a bond between the low thermal expansion ceramic coating and the more ductile, high thermal expansion metal substrate that will enable the coating to maintain a protective layer even in the subcritical water conditions of a pressurized water reactor (PWR). The stability, interface, and properties of several ceramic coatings on Zircaloy have been examined to select the best combination(s) of coating and processing. Strategies being followed to address this challenge include (1) investigating a range of coating materials including SiC, Al₂O₃, and diamond-like carbon (DLC); (2) studying the effect of processing types including chemical vapor deposition (CVD), sol/gel processing, and sputtering, and processing parameters such as substrate temperature; (3) investigating the use of a compliant interlayer to relieve expansion mismatch stresses; and (4) researching the effects of surface topography.

The thickness and composition of the ceramic passivating layers were determined by Auger electron spectroscopy (AES) and X-ray photoemission spectroscopy (XPS). Continuity of the ceramic layers was determined by scanning electron microscopy (SEM). Ceramic coatings were screened for hydrothermal stability by placing samples in an autoclave at 650°F and 3,000 psi for 24 hours and examining the ceramic coating for continuity by AES and SEM. Electrochemical corrosion resistance was examined by the potentiodynamic polarization (DC method) and electrochemical impedance testing (AC method). Adhesion of the passivating ceramic layers was examined by scratch testing. The scratch-test method consists of the generation of scratches with a spherical stylus (generally Rockwell C diamond, tip radius 200 μm) that is drawn at a constant speed across the coating under either constant or progressive loading.

Research Progress

Results are presented for each of the three years of the project.

Summary of Results from the First Year

- A study of coefficient of thermal expansion (CTE) data extracted from the literature suggested that Al₂O₃ and SiC would be good candidates to match the CTE of zirconium.
- Thermal conductivity considerations indicated that SiC would be a good choice, although Al₂O₃ films with less than 50 μm thickness were shown to have only very minor effects upon thermal conductivity and center line fuel temperatures.
- A neutronic analysis indicated that thin ceramic coatings of less than 50 microns would have a negligible effect upon reactivity.
- SiC and carbon were deposited by a plasma assisted chemical vapor deposition (PACVD) process onto

Zircaloy-4 coupons and characterized by Auger electron spectroscopy (AES). The SiC coatings prepared from silacyclobutane (ScB) as the precursor did not have continuity under the initial processing conditions studied. Processing studies were continued in the second year.

- Diamond-like carbon (DLC) coatings were deposited onto Zircaloy-4 coupons by a PACVD process and characterized by AES. The promising results of the first year prompted continuation into the second year.
- Al₂O₃ coatings were deposited by an Ultramet proprietary ultraviolet chemical vapor deposition (UVCVD) process onto Zircaloy-4 coupons that had their surfaces modified by either polishing, laser roughening, or chemical roughening. Evaluation of these samples by AES indicated some chloride impurities in the coatings, suggesting a probable point of failure, and so this effort was terminated.
- Al₂O₃ coatings were deposited on Zircaloy-4 coupons by sol-gel processing from aluminum alkoxides and characterized by AES. Ultimately, work conducted during the second year showed that no sol/gel coatings would withstand even 24 hours in an autoclave at 650°F/2,500 psi and so this approach was also abandoned.
- A 6"x4" sheet of Zircaloy-4 was coated with Al₂O₃ by a thermal barrier coat (TBC) sputtering process in Praxair's Appleton, Wisconsin, laboratory. The coating was too thick (i.e., 2 mm) and no other coating processor could be identified that could apply thin Al₂O₃ by this process, so this approach was terminated.
- Nearly stoichiometric SiC coatings were deposited on Zircaloy-4 coupons by a laser ablation deposition (LAD) process and characterized by AES, X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS). Preliminary economic analysis suggested that this process was prohibitively expensive and the work was not continued in subsequent years.

Summary of Results from the Second Year

- SiC films of approximately 700 nm thickness were deposited by the PACVD process, using ScB and H₂ as process gases. XRD, AES, and energy dispersive X-ray (EDX) spectroscopy data showed the film compositions were nearly stoichiometric SiC and uniform, containing

little oxygen content. Characterization indicated that film morphologies were influenced by process variables such as substrate temperature during deposition, the ScB/H₂ ratio, as well as the surface condition of the substrate. Film thickness, surface morphology, and EDX data indicated good reproducibility in film deposition. Though the results looked promising, Florida's PACVD system proved to be very unstable and reproducibility became very problematic. For this reason, PACVD work was continued at MER Corporation in Tucson, Arizona.

- Sputter-deposited alumina films exhibited very uniform and smooth surface morphologies with sharp interfaces between film and substrate. Problems with poor film adhesion during autoclave testing were partially solved by annealing the films at 500°C in argon.
- Zircaloy-4 substrates were surface alloyed by first sputtering on a thin layer of aluminum metal. These samples were then oxidized at 500°C in air for 2 hours and exhibited promising compositional gradient structures. Reducing the aluminum film thickness allowed the oxidation to reach the substrate. The result is that there is no intermediate region of unoxidized metal and the film adhesion properties are improved.
- DLC coatings were deposited onto Zircaloy-4 substrates having various surface finishes by Los Alamos National Laboratory using a plasma source ion implantation (PSII) process. They all exhibited high corrosion resistance and corrosion potential in electrochemical tests.
- Electrochemical corrosion studies showed that surface finish influenced the corrosion behavior of coated Zircaloy-4 substrates. The DLC coated sample with a 600 grit surface finish contained defects that act as initiation sites for breakdown. The DLC coated sample with a 0.3 μm surface finish provides good corrosion protection during short-term autoclave exposure; however, at longer exposure times all of the DLC coatings failed.
- The sputtered alumina samples completely failed during the electrochemical impedance corrosion testing.
- The PACVD-deposited SiC coatings provided excellent corrosion protection before autoclave exposure. Silicon carbide films with 5 μm thickness showed significant cracking, which decreased the

effectiveness of these films in protecting the underlying substrate. By comparison, 1 μm thick films exhibited a much larger degree of corrosion protection.

Summary of Third Year Efforts to Date

- Aluminum oxide coatings were prepared by controlled oxidation of aluminum metal films on Zircaloy substrates. Analysis of the oxidized coatings showed that gradient compositions were obtained, with Al, Zr, and O content varying through the coating thickness. X-ray diffraction analysis also showed that a variety of intermetallic and oxide phases (such as Al_3Zr , Al_2Zr_3 , Al_2O_3 , ZrO_2 , and Zr_3O) were formed in the coatings during processing. None of the alumina coatings tested to date have survived dynamic autoclave exposure (350°C, 3,000 psi) intact. Well-faceted particulates were observed on the sample surfaces after autoclave treatment, suggesting the hydrothermal re-crystallization to AlOOH .
- A detailed study on the effect of Zircaloy substrate surface treatment on the adhesion of silicon carbide coatings was performed. Substrates were mechanically, chemically, and thermally treated prior to coating deposition to obtain a variety of different surface conditions. Silicon carbide coatings were then deposited on the treated substrates using a PACVD process at MER Corporation (see Figure 1). The adhesion of the resulting coatings was assessed using scratch tests. The results indicated that the surface roughness affects film adhesion, with intermediate surface finishes (substrate polished with 240 grit paper) resulting in higher adhesion than samples with finer (600 grit) or coarser (grit blasted)

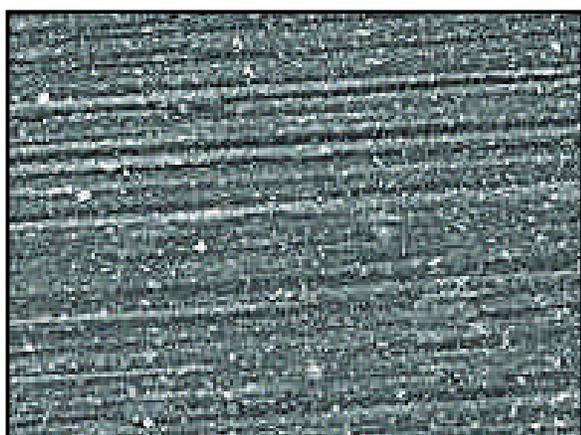


Figure 1. SEM image of the surface of a SiC film (1 mm nominal thickness) deposited on a Zircaloy-4 substrate by PACVD. The coating was deposited at MER Corporation.

treatments. None of the silicon carbide coatings tested to date have survived autoclave exposure (350°C, 3,000 psi).

- DLC coatings (deposited in the last quarter) have not survived autoclave testing (see later explanation), although electrochemical corrosion tests indicate some degree of protection compared to the bare Zircaloy substrates.
- MER Corporation has carried out a detailed economic analysis of the process for coating 1.5 million full-length Zircaloy-4 clad tubes per year with silicon carbide by their PACVD process. Their analysis shows that coating clad tubes with silicon carbide would add an additional cost of \$42.00 per tube.
- No processing conditions were found where the ceramic coatings were completely stable in an autoclave, which simulated PWR conditions. A detailed failure analysis was undertaken to determine the mode of failure for each of the ceramic coating types, DLC, SiC and Al_2O_3 .

Even though very pure single-crystal Al_2O_3 showed no observable changes when exposed to the autoclave conditions, alumina coatings produced by surface alloying with sputtered aluminum followed by oxidation or produced by sputtering from a pure Al_2O_3 target showed by SEM and glancing angle XRD that significant amounts of boehmite, AlOOH , were produced by hydrothermal growth under these same autoclave conditions (see Figure 2).

The results suggest that the failure of all of the alumina-based coatings under autoclave conditions may have resulted from formation of a less thermodynamically stable form of alumina such as an

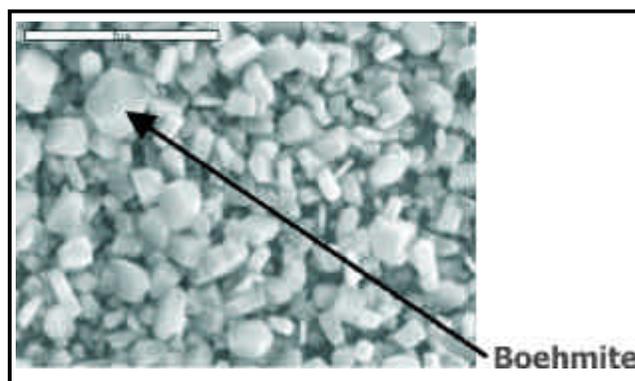


Figure 2. The figure is a SEM micrographs of a 300 nm-thick oxidized aluminum film after autoclave exposure for 24 hours. The aluminum film was deposited on Zircaloy and oxidized at 650°C for 30 minutes followed by 500°C for 2 hours.

amorphous phase or an oxygen deficient phase during the coating process, which would react with water to form the boehmite.

The DLC coatings generally failed after the autoclave exposure by lifting from the surface in large pieces. These pieces would then curl (see figure 3). These results suggest a weak substrate/coating interface but a tough film. The curling after removal from the substrate suggests that the films had residual stresses. The origin of the failure could be a pin hole where the underlying Zircaloy-4 began to oxidize putting stresses onto the film. The DLC appears to be stable, but the bond between DLC and Zircaloy appears to be the problem, with the DLC spalling off.

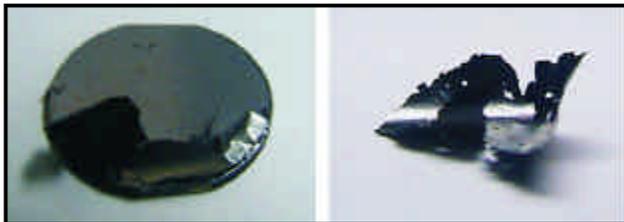


Figure 3. The photographs illustrate the DLC coating after autoclave exposure.

The silicon carbide coatings failed after the autoclave exposure (Figure 4) by first forming blisters generally associated with the the substrate topography caused by the surface finish process before coating. The origin of the blisters is probably pin holes in the coating caused by surface roughness and shadowing effects during the CVD coating process giving rise to holes or thin spots. Oxidation of the underlying zirconium alloy would result in stresses and blistering of the coating.

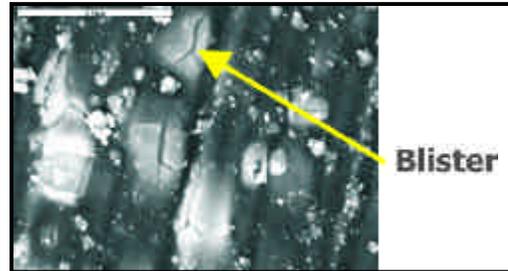


Figure 4. The figure illustrates SEM images of 1 micron SiC film on Zr-4 substrate (600 grit polished), after autoclave test.

Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

Optimization of Heterogeneous Utilization of Thorium in PWRs to Enhance Proliferation Resistance and Reduce Waste

Primary Investigator: Michael Todosow, Brookhaven National Laboratory (BNL)

Project Number: 00-014

Project Start Date: September 2000

Project End Date: September 2003

Collaborators: Massachusetts Institute of Technology; Ben-Gurion University of the Negev, Israel; the Russian Research Center-Kurchatov Institute (RRC-KI); Commissariat a l'Energie Atomique (CEA), France (inactive); Korea Atomic Energy Institute (KAERI); Kyung Hee University, Korea; Korea Advanced Institute of Science and Technology (KAIST)

Research Objectives

The objective of this work is to examine heterogeneous core design options for the implementation of the Th-U233 fuel cycle in pressurized water reactors (PWRs) and to identify the core design and fuel management strategies which will maximize the benefits from inclusion of thorium in the fuel. The assessment concentrates on key measures of performance in several important areas including proliferation characteristics of the spent fuel, reliability, safety, cost, environmental impact, and licensing issues. The focus is on once-through fuel cycles that do not involve reprocessing of the spent fuel. A 193 assembly Westinghouse reactor utilizing 17x17 fuel is taken as the model core.

Design optimizations involve heterogeneous core options that aggregate the thorium in subassembly units or whole typical PWR assembly units. The assessment will include comparison to the case of all-uranium fuel (the current fuel cycle and its future extrapolations), as well as the case of Th-U fuel mixtures within individual fuel pins (in both homogeneous and micro-heterogeneous embodiments). Optimization of the homogeneous fuel cycles is being performed under separate projects.

Two heterogeneous thorium implementation options are being explored, and expanded on, in the course of this NERI investigation: 1) the Seed-Blanket Unit (SBU)/Radkowsky Thorium Fuel (RTF) concept, which employs a seed-blanket unit that is a one-for-one replacement for a conventional PWR fuel assembly; and 2) the whole assembly seed and blanket (WASB), where the

seed and blanket units each occupy one full-size PWR assembly and the assemblies are arranged in the core in a modified checkerboard array (see Figure 1). The studies for both approaches seek to (1) identify the core design and fuel management strategies that will maximize the benefits from inclusion of thorium in the fuel, and (2) extend the analyses to validate the results over a range of possible operating conditions.

Research Progress

Designs for both the SBU and WASB approaches have been developed that significantly improve the intrinsic proliferation resistance of the fuel while achieving 18-

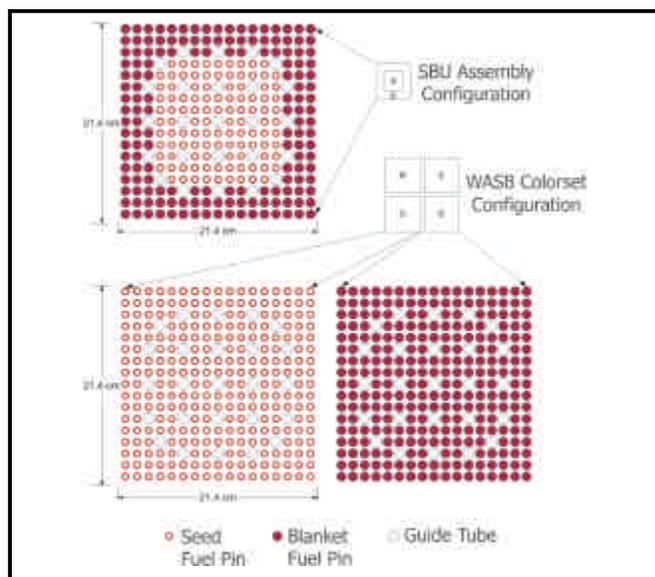


Figure 1. The graphic illustrates various SBU and WASB fuel assembly designs.

month cycles. The total production of plutonium is reduced by a factor of approximately 3-5 relative to a present commercial PWR operating on the uranium cycle. Furthermore, the plutonium that is produced is of inferior quality for potential utilization in weapons: it has a higher heat generation from increased Pu-238, a higher level of radioactivity, and a stronger neutron source from increased Pu-238, Pu-240, and Pu-240. Significantly, both approaches utilize assembly designs based on a Westinghouse 17x17 assembly where the sole modification is in the details of the fuel rods. Therefore, in principle, they are retrofittable into existing PWRs.

In addition, the details of a formalism to evaluate the fuel cycle costs of both SBU and WASB fuel cycles in comparison to a standard PWR cycle have been agreed upon, including the exact formulas and the input data required. The approach includes consideration of the different time periods that seed and blanket components spend in-core, using the levelized fuel cost over a single blanket cycle (typically six to nine operating cycles) as the primary measure of economic value. It also produces comparisons of fuel utilization and material flows. Specific achievements for each approach are summarized below.

The heterogeneous thorium implementation scheme based on the seed-blanket unit is being examined by BNL (with support from Ben Gurion University) under the present NERI project. The project is closely related to an ongoing project supported by the DOE Initiatives for Proliferation Prevention (IPP) program and private funding, which is focused on RD&D activities in Russia and the West. The initial reference point for the optimization studies is based on the results for the PWR from that IPP study.

Progress over the past two years includes the following:

- Conducted independent neutronic and thermal-hydraulic assessment of the reference SBU PWR design. The neutronic evaluations included the MCNP4C and RECOL Monte Carlo codes with explicit geometry modeling and continuous nuclear data, compared to results from the deterministic code BOXER with multi-group cross sections. The results provided initial confirmation of some key aspects of the design.
- Performed simple validation of nuclear data and codes for burnup reactivity and isotopics by analyzing several benchmarks, including the Thorium-Uranium PWR rod-cell

benchmark. Results with the BNL methodologies DRAGON and MONTEBURNS (MCNP Monte Carlo + ORIGEN) were in good agreement with results from other codes.

- Defined a reference "improved" SBU design based on sensitivity studies that included varying seed fuel and burnable poison compositions, and rod parameters, within the constraints of a Westinghouse 17x17 assembly design (rod pitch, guide-tube locations). The design assumes annular uranium dioxide pellets with a central burnable poison annulus for the seed, and solid thorium/uranium dioxide pellets for the blanket. The resistance of the grid spacers and the rod outer diameters are profiled to enhance coolant flow into the high-power seed region.
- Adopted a reference mechanical design for the SBU that permits inserting and removing seed rods into an SBU. In the selected approach, rods will be removed and inserted rod-by-rod, using a machine based on one designed to replace failed fuel rods. The virtue of this approach is that the basic mechanical design of the assembly (e.g., grid-spacers, upper and lower end-fittings) can be identical to that currently employed.
- Constructed a detailed COBRA-EN model for the SBU, and performed initial calculations of departure from nucleate boiling ratio (DNBR), and its variation with power. The resultant minimum DNBR based on the well-known W-3 correlation is 1.28 at nominal power based on conservative estimates for pin, assembly, and axial peaking factors, and nominal uniform grid resistances. Therefore, further optimization of power distributions and thermal-hydraulic characteristics is required in order to achieve an acceptable design.

The WASB design assumes that each type of fuel occupies a whole PWR assembly. The assessment of this design is largely performed at MIT.

Progress over the past two years includes the following:

- Designed WASB seed assemblies and blanket assemblies, which, in principle, are backfittable into existing PWRs. They employ oxide fuel in both seed and blanket assemblies (annular uranium dioxide pellets with erbia burnable poison in the

central region of the seed, and solid thorium/uranium dioxide pellets in the blanket). For the model Westinghouse PWR, the same 17x17 rod array is utilized as in commercial reactors, but with smaller diameter fuel rods in the seed, and larger diameter rods plus flow-resistance grids in the blanket assemblies, in order to enhance flow in the higher-power seed assemblies and reduce it in the lower-power blanket assemblies.

- Attained acceptable DNB margins under overpower conditions typical of anticipated operational transients through use of these assembly design features, together with the nuclear design of the cores. Evaluations were conducted with a full-core model using EPRI's VIPRE thermal margin code with conservative axial power peaking and thermal-hydraulic inputs.
- Investigated numerous core designs, but the most interesting appears to be one utilizing 84 seed assemblies and 109 blanket assemblies. Its reactivity coefficients are similar to those for commercial PWR cores. Typical three-batch fuel management is employed for the seeds, so that groups of 28 are loaded in sequential cycles and spend three cycles in the core, achieving a discharge burn-up of about 140 MWD/kgU. The 109 blanket assemblies are loaded and discharged together, and spend six to nine cycles in core, achieving a discharge burn-up of about 90 MWD/kgHM.
- Conducted preliminary analyses of fuel rod mechanical behavior at these high burn-ups utilizing the NRC's computer code for this purpose, and identified high fission gas release in the seed and oxide growth on the surface of blanket rods as challenges to the mechanical design of the fuel rods. Subsequent analyses have shown that the use of a larger fuel rod plenum combined with optimization of the burnable poison could probably reduce the fission gas issue in the seed to manageable levels, and that advanced clad types now coming into use, M5 for example, could eliminate the oxide corrosion problem.

- Developed reactor physics methods suitable for "production" analyses of WASB type seed-blanket cores. This will be particularly valuable for future work, since it allows analyses to proceed rapidly using convenient computer codes well-known in the industry. Because the applicability of these codes to WASB type fuel had not been verified, their use has been validated by comparison with Monte Carlo analyses using MCNP and MOCUP. Additionally, the NRC's FRAPCON-3 code for fuel rod mechanical analysis was modified to include both the thermal properties of thorium-uranium oxides, and a calculation of the rim effect in the presence of both U238 and U233.

Planned Activities

Planned work for the final year of the project will include the following:

- Modify the blanket neutronic design so that the combined total of U233 and U235 does not exceed the proliferation limit proposed by Forsberg at ORNL. Although it is anticipated that this will not be difficult, it will require re-analysis of the core designs to assure that they remain acceptable.
- Evaluate reactivity parameters related to control rods - e.g., total bank worth, stuck rod worth.
- Perform an initial assessment of the safety characteristics of both designs.

NUCLEAR ENERGY RESEARCH INITIATIVE

High Performance Fuel Design for Next Generation PWRs (Annular Fuel Project)

Primary Investigator: Mujid S. Kazimi,
Massachusetts Institute of Technology

Project Number: 01-005

Collaborators: Gamma Engineering Corporation;
Westinghouse Electric Corporation; Duke Engineering
& Services (now Framatome); Atomic Energy of
Canada Limited

Project Start Date: August 2001

Project End Date: September 2004

Research Objectives

The overall objective of this NERI project is to examine the potential for a high performance advanced fuel for pressurized water reactors (PWRs), which would accommodate a substantial increase of core power density while simultaneously providing larger thermal margins than current PWRs. This advanced fuel will have an annular geometry that allows internal and external coolant flow and heat removal. Details of the tasks follow.

- (1) Identify the most promising fuel assembly arrangement of internally and externally cooled annular fuel for PWRs to achieve a significant increase of power density (by at least 30 percent), based, to a large extent, on the extensive PWR fuel database to minimize research and development (R&D) expenses and deployment risks.
- (2) Optimize the fuel for superior thermal hydraulic and safety performance. Examine the optimum flow distribution, core pressure drop, maximum departure from nucleate boiling ratio (DNBR), and the resistance against parallel channel instabilities. Perform safety analyses, such as loss of coolant accident (LOCA) analyses, to confirm safety benefits expected for the new fuel.
- (3) Evaluate the neutronic fuel design with respect to achievement of high reactivity-limited burnup and reasonably long refueling cycle to attain good economic, waste, and proliferation-resistance features. Confirm that reactivity feedbacks and reactivity control aspects are acceptable.
- (4) Select fabrication processes to produce annular fuel elements with the required product characteristics, including fissile loading and high

integrity cladding, which are capable of eventual scale-up into a low-cost, efficient production process for economic and reliable fuel element performance.

- (5) Evaluate the performance of UO₂ fuel forms obtained by production technologies different from current U.S. practices (e.g., vibropacked fuel), and operating under new conditions (especially low peak fuel temperature) on fission gas release, and fuel dimensional properties during burnup. Models will be developed and used for the fuel performance assessment as well as scoping irradiation tests performed at the MIT reactor.
- (6) Optimize the core and plant design to minimize the electricity cost in cases of using the annular fuel for uprating current Generation II PWRs or in new advanced PWRs.

Research Progress

The progress will be summarized according to tasks.

Task 1. Thermal Hydraulic and Mechanical Design and Safety Analysis: A computer code for an isolated channel thermal hydraulic analysis of Internally and eXternally cooled Annular Fuel (IXAF) has been developed and used for the thermal-hydraulic optimization of fuel design. Also, the well-established VIPRE-01 model of the hot fuel assembly having 13x13 annular fuel rods was developed and comparisons made with the isolated channel model. Good Minimum Departure from Nucleate Boiling Ratio (MDNBR) agreement was shown between the two models. To identify the optimum dimensions of annular fuel in a square lattice, various array sizes (11x11 to 15x15) that fit in the fixed dimensions of a fuel assembly were explored using the isolated channel model.

The most promising option, based on thermal hydraulic considerations, was found to be a 13x13 array with inner and outer cladding diameters of 8.6 and 15.4 mm, respectively (see Figure 1). This geometry offers the highest Departure from Nucleate Boiling Ratio (DNBR) margin, a low peak fuel temperature and tolerable pressure drop. The 13x13 design, designated as PQN-02, was found to have a peak fuel temperature (at 150 percent power) that is about 1,300°C lower than the reference solid fuel. Therefore, a large power density increase and simultaneous increase of margin for cladding temperature during LOCA is feasible. The major limiting thermal hydraulic parameter appears to be the axial pressure drop across the core, because higher coolant flow rates would be required to maintain a core outlet temperature comparable to current designs and prevent saturated boiling. Additionally, vibration-related issues may limit the core power density increase, since they impose limits on coolant velocity. Therefore, only a 50 percent power density increase was adopted as the target power level. This is a significant power uprate, raising the extracted power from the same core size to support increasing the plant output from the current 1,150 MWe to 1,750 MWe.

In the safety analysis arena, Duke Engineering & Services (DE&S) finished the development of a suitable RELAP5/MOD3.2 base model for a sample PWR. The model simulates a standard Westinghouse 4-loop design, rated at a power level of 3,411 MWth with solid fuel rods and will be used to provide a benchmark calculation. In addition, an evaluation of LOCA performance of the annular fuel in a Standard Korean Nuclear Power Plant (KSNPP) using RELAP5/MOD3.2 was performed at MIT. The peak cladding temperature was found to be ~300°C smaller in comparison with reference KSNPP fuel and 600°C lower in temperature than the Nuclear Regulatory Commission's (NRC's) 1,200°C limit, allowing significant power uprate.

Task 2. Reactor Core Physics and Fuel Management:

Initial activities were focused on benchmarking and selection of analysis tools. The MCNP based burnup code MCODE (a recently developed code at MIT) was compared against CASMO4 for a typical unit cell with a UO₂ solid pin. A significant discrepancy between CASMO4 and MCODE (Monte Carlo-Origen Depletion) confirmed the expectation that CASMO4 would not be suitable for direct application to IXAF since it employs, as a default, an exponential radial distribution of U-238 resonance integral within a fuel pin. Various options for modifying CASMO-4

input have been investigated to achieve better agreement with MCODE.

The development of a modified CASMO-4 will enable whole core fuel management studies with a standard diffusion code. Using MCNP and MCODE for a unit cell or fuel assembly level, with the same enrichment and core power density, the PQN-02 13x13 design was found to

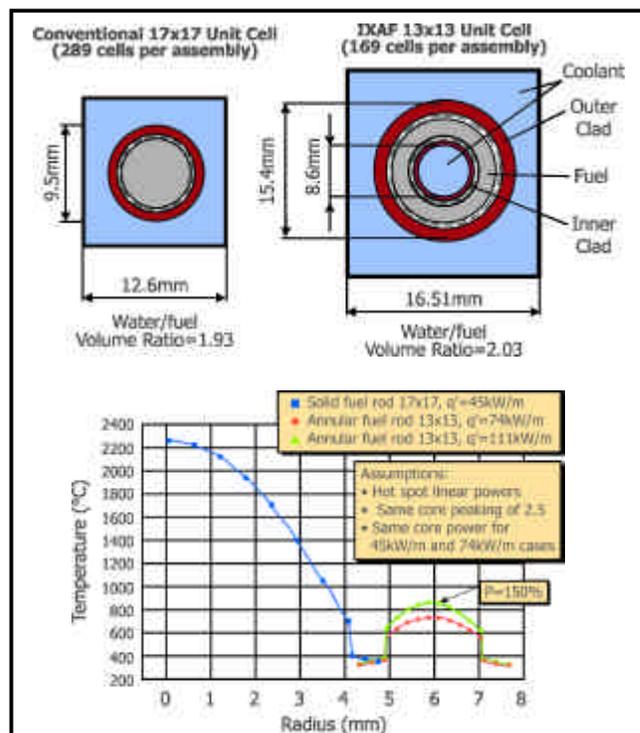


Figure 1. The graphic compares the performance of solid and annular fuel.

achieve the same burnup as the solid pin in spite of larger parasitic losses due to increased cladding volume. This is because of a slightly higher moderator to fuel ratio, higher conversion ratio, and a smaller reactivity penalty from fuel temperature coefficient due to the substantially lower fuel average temperature of the annular fuel. However, less total energy (i.e., fewer effective power days) is achievable because of the smaller heavy metal inventory of the PQN-02 design. Vibro Packed (VIPAC) fuel generally has smaller smear density than the pellet fuel resulting in smaller reactivity-limited burnup than for the pellet fuel. The addition of uranium getter powder, based on Russian manufacturing and irradiation experience, allows the achievement of higher smeared density than in pellet fuel resulting in correspondingly longer cycle. However, the United States does not have experience with nor data for this fuel and an R&D program if required.

Task 3. Fuel Fabrication Studies: Analyses of key design and performance requirements of optimized annular fuel elements were conducted at Gamma Engineering, with

particular focus on those requirements that affect the selection of annular fuel fabrication technology. Potential fabrication routes and processing technologies of annular fuel elements were identified as the following:

- (1) Sintered ring pellets fabrication route with traditional punch and die technology for fabricating green pellets before sintering processing;
- (2) Sintered ring pellets fabrication route with slurry extrusion technology for fabricating green pellets before sintering processing;
- (3) Sintered ring pellets fabrication route with tape casting technology for fabricating segments of green ring pellets before sintering processing;
- (4) VIPAC fuel element fabrication route with different particle size components of crushed high density sintered UO_2 fuel material; and
- (5) VIPAC fuel element fabrication route with different particle size components of spherical high density UO_2 fuel material formed by special sol gel processes.

A subcontract to Atomic Energy of Canada, Ltd (AECL) for the fabrication of VIPAC annular fuel test specimens for irradiation at MIT reactor has been signed and became effective in April of 2002. The delivery of finished products to MIT for in-reactor testing is scheduled in November 2002.

Task 4. Economic Analyses and Optimization: For manufacturing cost evaluations, the baseline constraints and assumptions adopted were those that reflect the current permitting and operational constraints at the Westinghouse Nuclear Fuels plant in Columbia, South Carolina. Changes for all manufacturing procedures associated with annular fuel have been identified and the cost difference is under evaluation. A preliminary assessment indicates small increases in manufacturing costs are expected, about 0.5mills/KWhe.

For the fuel cycle and capital cost assessment, an approach was developed for the evaluation of the costs and benefits of the high-power enabled by the annular fuel and preliminary analyses were performed. The early results indicate that the benefits of increasing the power density in the vessel of new PWRs easily exceed the incremental costs associated with fuel manufacturing.

Task 5. Fuel Performance Evaluation: The activities focused on preparing the design of the fuel irradiation experiment and on fuel modeling. The design of the test

specimens have been proposed and analyzed to ensure that temperature profiles, representative of those in the power reactor, can be achieved under sufficient cooling at the MIT Research Reactor (MITR). Application for license amendment has been submitted to the NRC. NRC's questions have been answered and minor revisions are currently being completed. The Safety Evaluation Report for approval of the planned experiment by the MITR-II Reactor Safeguards Committee is nearing completion and the draft is currently being reviewed by the MITR-II Operations staff.

For the fuel performance modeling, the FRAPCON-3 code is being modified for IXAF. The structure of the original FRAPCON-3 has been redesigned and the proper code initialization has been established for the annular fuel. Calculations of inner channel coolant conditions, cladding corrosion, crud uptake, hydrogen concentration, film temperature drop, and temperature drops in crud, oxide layer, and across inner cladding have been performed. The gas production and burnup subroutines have also been modified for annular conditions. A new fuel cladding mechanical interaction model, based on the existing FRACAS-I model, has been formulated and tested for annular fuel.

Planned Activities

Future activities will be focused primarily on the following efforts:

Task 1. The development of a VIPRE whole core model to obtain more accurate DNBR analysis, and study of the impact of geometrical uncertainties and partial blockage of the inner channel on thermal hydraulic performance. Additionally, assessment of mechanical design issues, in particular flow-induced vibrations and stability against liftoff.

Task 2. Calculation of reactivity feedbacks and boron and control rod worth, design of control strategy and whole core model for fuel management, and further optimization to achieve 12-month cycle length at 150 percent power.

Task 3. The manufacturing of annular fuel elements, using natural uranium or surrogate as the fuel body, using one, or possibly two of the processes examined in year 1. Varying of fabrication processing parameters to determine those parameters that offer the best promise of meeting the fuel product and performance requirements.

Task 4. The quantitative determination of marginal fabrication cost of annular fuel for sintered pellets and vibropack fuel, and evaluation of the economic benefit to PWRs from capital and fuel cycle costs using annular fuel at up to 150 percent power density.

Task 5. The completion of the FRAPCON-3 computer code for analysis of annular fuel performance for both pellet and VIPAC fuels, and of the design work for irradiation of VIPAC test specimens to be loaded in the MIT reactor in January 2003.

NUCLEAR ENERGY RESEARCH INITIATIVE

An Innovative Transport Theory Method for Efficient Design, Analysis, and Monitoring of Generation IV Reactor Cores

Primary Investigator: Farzad Rahnema, Georgia Tech Research Corporation

Project Number: 02-081

Collaborators: Pennsylvania State University; Idaho National Engineering & Environmental Laboratory

Project Start Date: September 2002

Project End Date: September 2005

The current generation of core neutronics methods are based on nodal diffusion theory and utilize homogenized cross sections and other physics data generated by single assembly, infinite medium transport theory calculations. This reactor-analysis methodology was developed and refined for the currently operating class (Generation II) of light water reactors (LWRs). Until about a decade ago, the reload cores of these reactors were designed with relatively homogeneous distributions of fuel, moderator, and absorber materials. For these systems, core-level diffusion theory is a good approximation, and the computational de-coupling of fuel assemblies for generating physics data is acceptable.

The current trend in LWR cores, however, is toward higher degrees of heterogeneity. In order to lengthen operating cycles, recent cores have been designed with higher amounts of total fissile mass, which has necessitated the addition of burnable absorbers to hold down the reactivity at the beginning of core life. Increased fuel utilization has been achieved by varying the fuel enrichment within assemblies and optimizing the arrangement of assemblies with significantly different fissile and fission product compositions. It is reasonable to expect these or similar design features to be present in the Generation IV light water reactors due to the unchanging desire to increase plant availability and reduce cost.

The trend toward compositional heterogeneity in LWRs and the desire for smaller, modular reactors in the Generation IV class will lead to cores with higher neutron flux gradients, resulting from increased core surface area to volume ratios in the latter case. In these systems, the neutron leakage between adjacent assemblies is significant and cannot be neglected. Generating physics data using single assembly infinite medium transport calculations, as is done with current core neutronics methods, may lead to

substantial errors in the homogenized cross sections and discontinuity factors. Without accurate data, the simplified nodal core model will produce inaccurate results. This is the consequence of the computational de-coupling of a highly coupled system.

Non-LWR Generation IV reactor designs are likely to be so different from current LWRs that they will necessitate a different (and probably smaller) set of assumptions on which to base core neutronics models. For example, in the pebble-bed modular reactor (PBMR), a high degree of uncertainty exists in the distribution of fissile mass among localized core regions due to the movement of pebbles with different degrees of burn-up as well as the presence of pebbles that contain only graphite. Further uncertainties resulting from the use of computational methods based on approximations to transport theory with limited ranges of validity will only exacerbate this problem. In addition, accurate calculations in localized portions of the PBMR cores must be performed in three spatial dimensions due to the complex geometry of packed arrangements of spherical pebbles. This aspect of PBMR cores creates problems for the current methods based on two-dimensional transport calculations applied to LWRs in which the variation of core properties in the vertical (third) dimension is relatively weak.

It is clear that the next generation of reactor analysis methods will be based largely on transport theory (both at the assembly and core levels) and involve fewer approximations regarding the nature of the core system than current methods. Diffusion theory and the multitude of methods based on transport corrections to diffusion theory will not be sufficient to support the optimum design, operation, and monitoring of the next two generations of reactor systems for the reasons delineated above. A computationally efficient core-wide transport theory method would provide a highly accurate and

flexible design tool (i.e., applicable to a much broader class of systems). In addition, from an engineering standpoint, it would support the pursuit of maximal increases in fuel utilization and plant availability and decreases in operating margins, the probability of fuel damage, and spent fuel inventory. These are many of the advantages sought in the Generation IV class—all of which lead to reduced overall costs.

The currently available transport theory methods have had limited success when applied to core-level calculations, and nearly all require the homogenization of assembly-level physics data. It is proposed that a next-generation, high-order variational coarse-mesh transport method be developed that does not require any homogenization or the use of discontinuity factors. The method is developed by deriving equations from a variational principle that admits discontinuous trial functions. Surface Green's functions are used for the spatial basis within each coarse-mesh and include all of the local transport characteristics as opposed to polynomial or other simple basis sets. Preliminary work in one-dimensional slab geometry with discrete ordinates and multigroup cross sections has been completed and

demonstrates the feasibility and promise of the approach. The fine-mesh results are reproduced exactly by the coarse-mesh method in the test problems. Integral to the proposed method, and therefore requiring no additional development, is the procedure for flux reconstruction at the detail of the fine-mesh calculations. The speed of the core calculation in higher dimensional geometries is expected to be close to that of current methods so that it can be used for design and core monitoring calculations.

The objective of the project is to develop the transport theory method and implement it for advanced and Generation IV LWRs and for the PBMR. The work will be accomplished through the collaborative effort of three organizations: Georgia Institute of Technology, Idaho National Engineering and Environmental Laboratory (INEEL), and Oak Ridge National Laboratory (ORNL). Georgia Tech will lead the project, develop the transport method, and implement the method for LWR calculations; INEEL will provide expertise in the area of PBMR calculations, and couple a coarse-mesh computational module to their pebble transport (movement) code; and ORNL will provide expertise in performing efficient and accurate transport calculations for both reactor types.

NUCLEAR ENERGY RESEARCH INITIATIVE

Advanced Extraction Methods for Actinide/Lanthanide Separations

Primary Investigator: Michael Scott, University of Florida

Collaborators: Argonne National Laboratory (ANL)

Project Number: 02-098

Project Start Date: September 2002

Project End Date: September 2005

The separation of An(III) ions from chemically similar Ln(III) ions is perhaps one of the most difficult problems encountered during the processing of acidic nitrate nuclear waste. In the 3+ oxidation states, the metal ions have an identical charge and roughly the same ionic radius. They differ strictly in the relative energies of their f- and d-orbitals, and in order to separate these metal ions, ligands will need to be developed that take advantage of this small but important distinction. If an efficient protocol can be developed for their partitioning, neutron bombardment can be employed to transmute actinides into products with significantly shorter radioactive lifetimes. Along with aiding the processing of acid nuclear waste streams, this methodology should ease some of the concerns expressed by the American public involving the long-term storage of nuclear waste.

With the intent to mimic the 3:1 CMPO:actinide stoichiometry of the extracted species in the TRUEX nuclear waste treatment process, a ligand system containing three pre-organized carbamoylmethylphosphine oxide (CMPO) moieties anchored onto a rigid three-fold symmetric triphenoxymethane platform has been developed for facile complexation of actinide ions and subsequent extraction from acidic nitrate nuclear waste streams. The CMPO arms on the ligands are oriented such that all three CMPO moieties can cooperatively bind a metal ion. Preliminary extraction experiments with simulated nuclear waste streams with solutions of the first generation of the ligand reveal a high affinity for the actinide thorium and a very low, constant affinity for the lanthanides across the series. Several different ligand derivatives have been prepared, and a series of distribution ratio measurements will be performed at Argonne National Laboratory with at least two lanthanides and americium to test the selectivity of the ligands for the 3+ metals. This information will be used to help design an improved ligand set.

One method to tune the actinide selectivity will be to influence the charge density of the metal and its coordination geometry. Accordingly, small alterations can be made to the ligand system to exploit these differences and further increase its affinity for actinides. Procedures have been outlined to incorporate modifications that alter the basicity of the CMPO oxygen donors as well as the distance between adjacent CMPO groups on the triphenoxymethane platform. With a wide variety of methods to alter the three arms, the binding attributes of the ligand can be subtly adjusted using the extraction data obtained from the group at Argonne to maximize the selectivity for the 3+ actinides.

The proposed work falls within the scope of fundamental chemistry program in the Nuclear Energy Research Initiative and funding from the Department of Energy will allow for a detailed examination of the An(III) binding properties of the ligands at Argonne National Laboratory. This information will be crucial for the further refinement of the C₃-symmetric actinide binder at the University of Florida. The continued cooperation between the two organizations should produce an advanced extraction process for the separation of the chemically similar actinides and lanthanides found in acidic nitrate nuclear waste streams. It is envisioned that the compounds prepared during this work can be used in a process to remove the An(III) ions from nuclear waste streams immediately following the PUREX process and perhaps after concentration of the An(III) and Ln(III) ions by the DIAMEX process.

NUCLEAR ENERGY RESEARCH INITIATIVE

Improving the Integrity of Coated Fuel Particles: Measurement of Constituent Properties of SiC and ZrC, Effects of Irradiation, and Modeling

Primary Investigator: Lance L. Snead, Oak Ridge National Laboratory

Project Number: 02-131

Collaborators: CEGA Corporation, Idaho National Engineering & Environmental Laboratory; Massachusetts Institute of Technology; General Atomics; Argonne National Laboratory

Project Start Date: September 2002

Project End Date: September 2005

The silicon carbide (SiC) layer integrity in the TRISO-coated gas-reactor fuel particle is critical to the performance, allowed burn-up, and hence, the intrinsic efficiency of high-temperature gas cooled reactors. While there has been significant developmental work on manufacturing the fuel particles, the effects of the complex in-service stress state combined with realistic materials property data under irradiation has on fuel particle survival are not adequately understood. Furthermore, there is virtually no experimental data on the effects of irradiation on the thermo-mechanical properties of zirconium carbide, which has been proposed as a higher-temperature replacement for SiC. The basic assertion behind this proposal is that a significant need exists for detailed fuel particle modeling including realistic, experimentally derived data on fuel particle constituent materials in the non-irradiated and irradiated condition. To perform this work will require advances in modeling, along with development of techniques for measuring materials properties at the small scale of the fuel particle.

Four elements are proposed for this work:

- (1) **Modeling Work:** In recent years, a collaboration has been established between Idaho National Engineering and Environmental Laboratory (INEEL) and the Massachusetts Institute of Technology (MIT) to look into finite-element and other methods of modeling the stress state of fuel pellets. This work has been carried to the point where it is being limited by the lack of realistic material property input. Specific information on the statistical distribution of strength, creep, and swelling for SiC is poorly described in the literature. Data on the thermomechanical properties of pyrolytic ZrC is also very limited.
- (2) **Technique Development for Measuring Constituent Properties:** To this point, techniques to study the integrity of fuel particles have been relatively rudimentary, consisting of compression tests (crush or c-ring) of the particle or bare SiC overcoat. The objective of this element is to apply state-of-the-art techniques and to develop new techniques specifically for application to the TRISO system to generate realistic data for the modeling. These techniques would then become available for the community developing gas-cooled-reactor fuels. Specific tools will be developed to measure strength through internal pressurization, elastic modulus on the scale of the TRISO particle, creep relaxation, and PyC/SiC interfacial properties.
- (3) **Irradiated Materials Property Information:** An irradiation program will be coupled with the technique-development program to generate information on mechanical properties needed for modeling input. The irradiation program will include the model, non-fueled cylindrical and spherical TRISO structures, and spherical TRISO-containing, helium-producing boron carbide.
- (4) **Updated Materials Data Handbook for TRISO Fuels:** As part of this effort, a materials property

This new data will be generated and applied in spherical and cylindrical model geometry. The objective of this work is to use the new data to better describe the stress state of the TRISO particle under irradiation and to give a direct comparison of the integrity of SiC-v-ZrC for this application. Potential failure during pellet processing will also be addressed.

handbook will be developed. This handbook will include pertinent physical property information on all constituent materials of coated particle fuel. Sources of information will be the open literature on nuclear materials, reports dealing with HTGRs (e.g., CEQA-002820, Rev 1), and information developed as part of this proposal. This material will then be available to the larger fuels community.

NUCLEAR ENERGY RESEARCH INITIATIVE

Enhanced Thermal Conductivity Oxide Fuels

Primary Investigator: Alvin Solomon, Purdue University

Collaborators: Framatome ANP, Inc.

Project Number: 02-180

Project Start Date: September 2002

Project End Date: September 2005

The objective of the proposed research is to produce a novel oxide fuel form with superior thermal conductivity. The research is proposed under NERI's Materials Science area of emphasis and enhances the long-term viability and safety of nuclear energy systems and the stability of the spent fuel as a waste form. The resulting fuel will be applicable to existing light-water reactors, especially with high burn-up, high performance fuels. It is also expected that such fuel will provide superior performance in advanced reactors that would otherwise be fueled with low-conductivity oxide fuels.

Although UO_2 fuel has many desirable chemical characteristics and has served satisfactorily in light water reactors for many years, its low thermal conductivity imposes significant limitations on reactor operations in present and especially in high performance, high burn-up future reactors. An increase in the thermal conductivity would relax several of these limitations and provide the following significant benefits:

- (1) Operational safety would be enhanced because fuel performance during a LOCA (loss-of-coolant accident) would be improved. The amount of heat stored in the fuel would be reduced, so peak cladding temperatures during dryout would be reduced.
- (2) A reduction in fuel temperatures and stored heat would support reactor power up-rates and improve power production economics.
- (3) The production of high-level radioactive waste would be reduced because fuel burn-up could be increased since lower fuel temperatures would result in less fission gas release, and smaller amounts of fission gas could eliminate limitations on burn-up that are tied to internal pressure of the fuel rod.

- (4) Reduced temperatures and temperature gradients in the fuel pellets would reduce the stresses imposed on the cladding, reduce fuel cracking and relocation, and reduce life-limiting fuel swelling, so the effects of pellet-cladding mechanical interaction would be reduced.

- (5) Proliferation resistance would be enhanced because the rate of production of ^{239}Pu decreases as burn-up increases.

The goal of enhancing the thermal conductivity of sintered oxide fuels will be achieved by a new process of penetrating a highly conductive solid second phase into the open or interconnected porosity of sintered fuel. This project would focus on UO_2 for the present program because the technology can be immediately applied to current LWR fuels.

The process first involves developing sintering schedules to produce a desired open pore structure (approximately 90 percent TD). The second critical step requires intrusion into the open porosity of the high conductivity phase. Although penetration of the porosity with a molten metal under pressure is conceivable, this requires very high processing temperatures for high-melting-point refractory metals or even zirconium alloys. A new, more attractive alternative method would be to infiltrate a liquid silicon carbide (SiC) polymer precursor and pyrolyze it at modest temperatures leading to an interconnected SiC phase with superior thermal conductivity for fully-dense polycrystalline material comparable to that of commercial purity silver (approximately 400 W/m-°K at room temperature). Although several infiltrations and pyrolyzing steps may be necessary, they could be readily performed in a simple batch process. Thus, it is proposed that the second approach be pursued.

Besides high thermal conductivity, necessary characteristics of a second phase include small neutron capture cross sections; high melting point; resistance to creep; compatible thermal expansion coefficients; and lack of chemical reactions with UO_2 , fuel cladding, or water. SiC has been identified as an excellent material from most of these perspectives. For example, its thermal conductivity at 1,000°K is roughly ten times that of UO_2 , and improves at higher temperatures. Moreover, the methodology for deposition of SiC carbide has been developed.

Standard UO_2 fuel has a very large temperature drop between the center and surface of the fuel pellets during irradiation. A drop of 1,000°C is not unusual during full power operation. The primary reason for the large temperature drop is the low thermal conductivity of UO_2 . Substantial increases in thermal conductivity are possible with modest volume loadings of the conductive phase. Preliminary model calculations indicate that if the second phase has a high thermal conductivity compared to UO_2 , a 10 percent volume loading of a continuous, randomly oriented second phase would increase the thermal conductivity of the fuel by about 50 to 100 percent, depending on temperature, and reduce the peak centerline fuel temperature in the hot channel bundle with a linear power of 29.8 kW/m by 800°C. It would be even greater for the peak pin.

The first technical challenge in producing an enhanced-conductivity fuel is first to model the heat-conducting performance of various microstructures. Three aspects of the microstructure are essential to good fuel thermal performance. First, the second phase must be finely dispersed. Widely spaced conductive paths would be less effective in conducting heat from the fuel and could produce localized hot spots on the cladding, resulting in nonuniform corrosion and hydrogen migration in the cladding. On the other hand, if the porosity is too fine and finely dispersed, penetration becomes difficult and thermal conductivity may be reduced by phonon scattering in the conductive phase. Second, the conductive phase must be continuous. Although there is a slight improvement of heat transfer even from discrete particles of a highly conductive material, much larger benefits are derived if the conductive material forms a continuous path. Third, the conductive network must penetrate the entire fuel pellet so that heat can be conducted efficiently from the center to the surface. Recent advances in SiC processing make it possible to produce SiC that has a very high thermal conductivity because of its high purity and good crystallinity. Doping with appropriate n-type elements can further increase the conductivity to above 480 W/m-°K. Silicon carbide also has a melting point of 2,700°C consistent with that of UO_2 .

NUCLEAR ENERGY RESEARCH INITIATIVE

Use of Solid Hydride Fuel for Improved Long-Life LWR Core Designs

Primary Investigator: Ehud Greenspan, University of California

Project Number: 02-189

Collaborators: Massachusetts Institute of Technology; Westinghouse Savannah River Company; University of Tokyo

Project Start Date: September 2002

Project End Date: September 2005

The general objective of this proposal is to assess the feasibility of improving the performance of PWR and BWR cores by using solid hydride fuels or solid hydride inserts. The concentration of hydrogen in the hydride fuel is comparable to that of hydrogen in liquid water of LWR cores. The introduction of part of the hydrogen needed for neutron moderation within the fuel volume permits attainment of optimal neutron spectrum while using a relatively small water volume fraction—just that amount of water that is required for comfortably safe cooling of the fuel. This feature enables the core to be designed to have optimal moderation, in terms of the attainable discharge burn-up, and to have a smaller volume or higher total power than a LWR core that uses oxide fuel. Moreover, thorium hydride fuel, one of the hydride materials to be examined, has a higher heavy metal (HM) density than oxide fuel. As a result of this higher HM concentration and larger fuel-to-water volume ratio, U-ThH₂ or Pu-ThH₂ fueled cores can be designed to have a significantly higher energy generation per core loading and significantly longer core life than the corresponding oxide-fueled cores. Preliminary estimates indicate that both the energy per fuel loading and core life could increase by more than a factor of 2. The core power level can also be significantly increased. The net outcome is expected to be improved economics, improved resource utilization, reduced waste, improved proliferation resistance, and improved safety.

This study may lead to the development of new fuel and core designs for LWRs that could have one or a combination of the following advantages relative to contemporary LWRs and those under development:

- (a) Reduced capital cost by virtue of compaction and/or increased power output;
- (b) increased discharge burn-up;
- (c) increased core-life;

- (d) increased energy generation per fuel loading;
- (e) reduced fuel cycle cost;
- (f) reduced waste volume and toxicity due to higher discharge burn-up and to partial utilization of Th;
- (g) increased utilization of Pu relative to MOX fueled cores due to the higher discharge burn-up possible with hydride fueled cores of acceptable power density;
- (h) utilization of thorium fuel resources;
- (i) simplified design of BWR fuel assemblies and control systems along with improved stability against power oscillations;
- (j) improved safety due to the large negative temperature coefficient of reactivity of hydride fuel;
- (k) improved capability to dispose of plutonium in LWRs by using fertile-free PuH₂ or Pu-Zr hydride fuel; (the large prompt negative temperature coefficient of reactivity of hydride fuel may compensate for the lack of large negative Doppler reactivity effect due to absence of fertile fuel); and
- (l) improved proliferation resistance due to enhanced destruction of Pu and use of thorium.

One hydride fuel being considered is uranium-zirconium hydride, similar to that developed by General Atomics (GA) for TRIGA reactors. This fuel has been in use for more than 40 years in many reactors around the world, both in constant power and pulsed power operating conditions. It has been extensively studied, and tested in reactors, and it has an impressive record of safety. Fuel for high power TRIGA reactors has been operating under conditions that in many aspects meet or exceed LWR fuel performance requirements. Relative to UO₂ fuel in LWRs,

this TRIGA fuel operates at close to twice the average linear-heat-rate, and reaches more than twice the discharge burn-up. The thermal conductivity of the TRIGA fuel is nearly six times larger than that of UO_2 so that its peak fuel temperature under typical LWR operating conditions is estimated to be below $700^\circ C$, which is acceptable and provides a comfortable margin. Some of the novel core designs with hydride fuel being proposed will feature lower linear heat rate and hence, lower peak fuel temperatures. This will result in a large margin to accommodate transients that lead to fuel temperature increase. Uranium-zirconium hydride fuel was also used in a sodium-cooled SNAP space power reactor developed by Atomics International.

Another hydride fuel proposed for consideration is uranium-thorium hydride. It was proposed as fuel for nuclear reactors by the late Dr. Masoud Simnad, the developer of the TRIGA fuel. $U-ThH_2$ is even more stable than $U-ZrH_{1.6}$ fuel and can operate at higher temperatures. More importantly, the HM density in the $U-ThH_2$ fuel can exceed the U density in UO_2 . It is estimated that by using $U-ThH_2$, it is possible to load more than twice the amount of HM into a core of a given volume than in the corresponding well-moderated UO_2 (or MOX) fueled core. This implies that it might be possible to extend the time between refueling by more than a factor of two, or, in principle, double the core power level.

Prof. M. Yamawaki of the University of Tokyo has recently developed and tested a $U-Th-Zr-H$ fuel and carried-out performance tests including in-core irradiation. The maximum permissible linear heat rate limit of that hydride fuel is estimated to be 500 w/cm , more than is needed for economic LWR operation. Prof. Yamawaki will collaborate on this study and provide needed data. Before passing away several months ago, Dr. Simnad had also expressed great interest in participating in this proposed project.

Another, smaller part of the study will assess the feasibility of designing PWR and BWR cores to have long life and large discharge burn-up using very compact lattices incorporating small amount of non-fuel containing solid hydride. The function of the solid hydride is to limit the spectrum hardening upon 100 percent voiding of the water coolant, and thereby help achieve a negative void coefficient.

The study will address primarily reactor physics, and thermal-hydraulic, safety, fuel-cycle, and economic considerations. Material compatibility research will be undertaken as a follow-on study provided the conclusions from the present study justify doing so. To ensure that the present study will adequately take into account material-compatibility issues, two material specialists, Prof. Olander of the University of California at Berkeley, and Prof. Yamawaki of the University of Tokyo, an expert on hydride fuels, have been included on the team. A complete list of the collaborators, their institutions, and their areas of expertise follows:

- (1) Greenspan, University of California at Berkeley, neutronics
- (2) Olander, University of California at Berkeley, material compatibility
- (3) Todreas, Massachusetts Institute of Technology, thermo-hydraulics and safety
- (4) Petrovic and Garkisch, Westinghouse, practical fuel and core design considerations and economics
- (5) Yamawaki, University of Tokyo, hydride properties and compatibility

The funding for Prof. Yamawaki's involvement will be provided by Japanese sources.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Advanced Methods for Pebble-Bed Reactor Neutronics: Design, Analysis, and Fuel Cycle Optimization

Primary Investigator: A.M. Ougouag, Idaho
National Engineering and Environmental Laboratory
(INEEL)

Project Number: 02-195

Project Start Date: September 2002

Collaborators: Georgia Institute of Technology;
Pennsylvania State University; Imperial College of
London, PBMR (Pty) Ltd.; University of Arizona

Project End Date: September 2005

The goal of this project is to develop a comprehensive suite of computer codes for the design of pebble-bed reactors (PBRs) and the management of their fuel cycles. The PBR concept is a leading candidate for near-term deployment and for further development as a Generation IV reactor, yet the neutronics methods available to design and analyze PBRs are several generations behind the state of the art. Existing PBR analysis codes use finite-difference or statistical methods, which are slow and thus unsuitable for the repetitive simulations needed for optimization. Thus, an efficient deterministic method is needed for design and optimization of the PBR fuel cycle.

The INEEL has developed a new deterministic method for the neutronics analysis of PBRs. The method accounts for the pebble flow explicitly and couples the flow to the neutronics. It can model once-through cycles and cycles that allow recirculation of pebbles through the core. The method is implemented in the INEEL code PEBBED. At present, PEBBED uses a finite-difference neutronics solver and simplistic depletion and cross-section computation techniques; these methods were applied to prove the viability of the basic algorithm in the code, but they should be replaced by modern techniques that fully complement the advance achieved by that algorithm.

The project will incorporate the needed modern techniques; beyond that, it will extend the state of the art in computational neutronics. A new method will be developed to carry out long depletion steps while maintaining high fidelity in the model nuclear data. The method will be an essential tool for the reliable design of a PBR or any reactor core of cylindrical geometry. No method or code exists that contains the capabilities proposed here.

The new method and tools will greatly enhance the scientific and computing infrastructure in the United States. The resulting codes will be essential to cost-effective PBR design, and they will give unprecedented fidelity to PBR modeling. Thus, the project will directly support two areas of the NERI work scope: F-1, Advanced nuclear energy systems, and F-3, Nuclear fuels/fuel systems. The project conforms to the NERAC Long-Term Nuclear Technology R&D Plan in advancing reactor physics technology for a Generation IV reactor concept, and it will enable assessment of the proliferation potential of PBRs as advocated by the NERAC TOPS report. It is also consistent with the National Energy Policy of 2001, which specifically mentions the PBR as a promising advanced reactor concept.

The following paragraphs explain the specific needs addressed in this project.

Implementation of a 3-D (r- θ -z) nodal/coarse mesh multigroup neutron diffusion solver: A way has been found to circumvent the mathematical impasse that has frustrated all previous efforts to find a mathematically rigorous formulation of an analytical nodal method in 3-D cylindrical geometry. In real PBRs, the presence of control rods or control elements, uneven burn-up, and uneven build-up of poisons will impose azimuthal asymmetry (i.e., θ -dependence). To perform core physics analyses efficiently in most reactor systems, nodal methods are generally applied. However, current nodal methods in cylindrical geometry do not include the θ -dependence rigorously because the transverse integration procedure (from which nodal method developments start) in r leads to a mathematical impasse. In this project, a new mathematically rigorous approach will be implemented into a nodal code for r - θ - z geometry.

Development of an advanced, spatially detailed depletion capability: A new nodal depletion method will be developed in this work, extending to cylindrical geometry the techniques in the NOMAD-BC code developed by one of the PIs,. The current standard methods used for PBR design and analysis rely on finite-difference neutronics solvers. Consequently, they are limited either to very low-fidelity modeling (by a very coarse computational grid) or to very long computational times (if the computational mesh is refined). Such methods in detailed design and safety studies would impose an unacceptable bottleneck by modern standards. The nodal/coarse-mesh-based depletion capability proposed in this project would eliminate this difficulty, because nodal methods have the potential to increase fidelity while greatly reducing computational times. With the new method, complete design analyses could be completed in a matter of seconds. Optimization studies would also be commensurately rapid.

Development of a new method for generation of diffusion-theory parameters: The nodal diffusion solver and nodal depletion method will be complemented by a cross-section perturbation method that will be developed in collaboration with Prof. Farzad Rahnama and his students at the Georgia Institute of Technology. This method will be an extension to cylindrical geometry of prior work at Georgia Tech. The method will be supported by the EVENT code developed by Prof. Cassiano de Oliveira at the Imperial College of London; Prof. Oliveira will collaborate as an unfunded participant. The new method extends modern equivalence theory past the present state of its art. Besides homogenized diffusion constants, the method will produce perturbation parameters, used to update nuclear data during changes in the reactor core that develop during long time steps, without resort to data-intensive, pre-computed tables or rehomogenization. This capability is unique and extremely well-suited to first-of-a-kind reactor design and analyses; it is also ideal for efficiency and fidelity in the analysis of existing reactor types.

Development of a method of feedback parametrization for temperature and depletion: In order to account correctly for the effects of temperature on diffusion-theory parameters, a feedback model must be included among the methods. Prof. Kostadin Ivanov at the Pennsylvania State University (PSU) has developed and validated a new adaptive high-order table look-up model for cross-section parametrization. PSU will modify their model to be compatible with PEBBED and with the methods developed

at Georgia Tech for generating diffusion-theory data.

Development of an automatic optimization routine for efficient and accurate sensitivity studies, design, and fuel management: An automatic optimization routine, based upon a genetic algorithm, will be developed and integrated with PEBBED to perform core design optimization. The algorithm will generate core size and flow parameters for PEBBED input and evaluate user-specified objective functions based upon the resulting PEBBED output.

Exploration of the implications of inhomogeneities in pebble packing: Packing in a cavity filled with identical spheres is not uniform, but varies according to a function that resembles a damped oscillation from zero at the walls to an asymptotic value several sphere diameters away. In small PBRs, where the oscillations may extend over a large fraction of the core diameter, the effects of the oscillations may be significant. Calculations will be performed to assess these effects, and an empirical model will be developed for inclusion in the homogenization codes.

Incorporation of code components into PEBBED: The neutronics analysis of PBRs differs from that of light water reactors because PBR fuel moves and the neutronics and depletion equations are coupled to this motion. The PEBBED code solves this coupled problem. All the new methods and codes developed in the tasks described above will be coupled to or implemented within PEBBED. The resulting code suite will be benchmarked by Prof. Barry Ganapol of the University of Arizona. Verification and validation will be performed using data supplied by the South African company, PBMR (Pty), Ltd., who will be participating as unfunded collaborators.

The team assembled for this project has a unique ability to perform the proposed work. The PIs have developed the PEBBED code and derived the θ -dependent analytical nodal formulation of the diffusion equation. The collaborators at Georgia Tech, with the support of Prof. deOliveira, have a unique and ideally suited approach to homogenization. The collaborators at PSU have a unique and ideally suited approach to data parametrization. Prof. Ganapol is known worldwide as an expert in all kinds of benchmarking problems. PBMR (Pty) Ltd. will provide relevant data from the world's foremost PBR design project.

The project will produce a set of interrelated codes to perform neutronics design and in-core fuel cycle optimization for a PBR quickly and efficiently. In addition, the project will lead to publications for conferences and

journals. The new capabilities will be demonstrated on sample optimization problems (PBR fuel cycle and non-proliferation characteristics of discharged pebbles).

