

NUCLEAR ENERGY RESEARCH INITIATIVE

5. ADVANCED NUCLEAR FUELS

R&D in the advanced fuels area is needed to provide measurable improvements in the understanding and performance of nuclear fuel with respect to safety, waste production, proliferation resistance, and economics in order to enhance the long-term viability of nuclear energy systems. This program element addresses the long-term R&D goal to develop improved performance and advanced fuel designs for existing light water reactors and advanced fuel designs and related fuel cycle requirements for advanced Generation IV reactor designs.

The scope of this long-term R&D includes a variety of thermal and fast spectrum power reactor fuel forms, including ceramic, metal, hybrid, (e.g., cermet, cercet), and liquid, as well as fuel types including 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. The R&D scope also includes development of higher density LEU (<20 percent U-235) fuels for research and development reactors.

Currently selected projects include innovative concepts for material preparation and production of nuclear fuels; inherently safe fuel designs and core response; understanding 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; and innovation in fuel design, composition, or other attributes that maximize energy production, optimize fissile material utilization, or reduce production costs.

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Development of Improved Burnable Poisons for Commercial Nuclear Power Reactors

PI: M.L. Grossbeck, Oak Ridge National Laboratory

Project Start Date: August 1, 1999

Projected End Date: August 31, 2002

Project Number: 99-0074

Research Objective

Burnable poisons are used in nuclear reactors to aid in reactivity control and to reduce power peaking. The materials used at the present time suffer from two common disadvantages. The first is that the elements currently used, such as gadolinium and boron result in a small residual negative reactivity. Ideally, the burnable poison should be entirely depleted by the time the fuel is depleted. In fact, some burnable poison or isotopes that result from neutron absorption in the burnable poison remain at the time of fuel depletion and serve to limit the amount of fuel that can be used. The second is that boron transmutes to helium, which creates undesirable internal fuel pin pressures. Elimination or reduction of these two effects will lead to higher fuel burnup and longer core life resulting in lower cost of operation.

For many absorbing elements, such as gadolinium, it is isotopes other than the primary absorber that lead to residual reactivity. A goal of this research is to investigate the possibility of separating isotopes to isolate the absorbing isotope of interest, thus reducing or eliminating the residual reactivity. Absorbing elements such as samarium, gadolinium, dysprosium, and other identified candidates are being considered. State of the art two-dimensional computer codes will be used to determine the effects of the new burnable poisons, in both homogeneous and self-shielded configurations, on reactivity and core safety parameters. The second phase of the project will investigate isotope separation by the plasma separation process, and test separations will be attempted. In the final phase of the project, product forms determined from phase one will be fabricated using techniques of ceramic processing.

Research Progress

The project is in the first phase where isotopes of strongly absorbing elements are examined. The main thrust of the project uses state of the art computer codes. Three-dimensional calculations are being performed using TALLY, MCNP4B, and ORIGEN2. Two-dimensional calculations are being performed with HELIOS. The core models use 8, 16, and in some cases, 64 burnable poison pins per fuel assembly in a PWR. Values of the reactivity coefficient (k_{eff}), normalized pin powers, and fuel and burnable poison burnup, as a function of time are calculated. Then reactivity of the burnable poison is

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calculated as a function of burnup. This residual reactivity due to the burnable poison is what is expected to be reduced through the isotope separation.

In conjunction with the detailed calculations, ORIGEN2 calculations are being performed on combinations of separated isotopes to examine their time behavior graphically. This series of calculations is used as a check on the Monte Carlo calculations and to point future directions. One group cross sections generated for the spectrum resulting from the burnable poison configuration are used in ORIGEN2. Reasonable agreement has been obtained for the case of homogeneous burnable poisons. Such calculations have demonstrated that removal of ^{154}Gd and ^{156}Gd results in about a factor of ten reduction in residual reactivity.

At the present time, Gd, Eu, Sm, Dy, Er, Yb, Hf, and Nd have been analyzed for the case of homogeneous burnable poison in the fuel. All have been found to have a self-shielding effect that extends the life of the burnable poison. Although this is usually a beneficial effect, it results in increased residual burnable poison. However, calculations are being done to evaluate burnable poisons in the form of coatings on the fuel pellets. In this configuration, several isotopes show promise if they are separated from the naturally occurring element. The isotopes ^{151}Eu , ^{164}Dy , ^{167}Er , ^{149}Sm , and ^{177}Hf have demonstrated improvement by placing them in a thin layer and enriching them isotopically. The residual reactivity as a function of layer thickness and burnable poison material density is also being investigated.

Calculational techniques have been refined to shorten the run time by a factor of ten. This has permitted rapid progress, and as a result, it is expected that the project will be completed within the time of the extension of phase one, by the end of January 2001. It must be remembered that isotopes cannot be developed. The isotopes provided by nature are all that can be used, making the project a high-risk endeavor. However, small improvements in fuel lifetime can result in large savings in fuel cost and reduction in waste.

Planned Activities

The above investigation has led to exploration of incorporating burnable poison in the fuel cladding. This achieves reduced self-shielding. It also changes the chemistry and compatibility issue to one of cladding rather than fuel. This is being explored at the present time, although results remain preliminary. The concept of incorporating burnable poison in the cladding has led to the production of a cladding/burnable poison alloy, which is now being examined.

Technical discussions have been initiated with a private company that is constructing a facility for separation of isotopes by plasma separation. This method promises to be significantly less expensive than methods used at the present time for isotope separation. In the second phase, it is expected to do a separation run of one or more isotopes identified in the first phase.

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

PI: Sean M. McDeavitt, Argonne National Laboratory

Collaborators: Purdue University

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0095

Research Objective

Metal-matrix cermet nuclear fuels have potential for use in a once through, high-burnup, proliferation resistant fuel cycle. This project combines the advantages to be gained from cermet fuel with the resources extension potential of the thorium oxide fuel cycle and the inherent proliferation resistance of mixed oxide ceramics. These advantages fit well with the DOE's focus on the development of Generation IV nuclear power systems and proliferation resistant fuel cycles. The goal of this project is to demonstrate the feasibility of a metal-matrix fuel comprising (Th,U)O₂ microspheres in a zirconium matrix that can achieve high-burnup and be directly disposed as nuclear waste.

Research Progress

For Task 1, Processing and Characterization of (Th,U)O₂ Microspheres, spray drying and sintering methods are being developed to fabricate the (Th,U)O₂ microspheres. The first year has been spent in obtaining authorization, setting up equipment, and establishing approved procedures. Approvals were obtained from the U.S. Nuclear Regulatory Commission and from the Purdue Radiological Control Committee to receive and use thorium. Using a surrogate alumina powder, slurry dispersion methods were studied and slurry viscosities were measured as a function of shear rate, dispersion methods, and pH. Spray drying procedures were developed using alumina powder and spray dried alumina microspheres were characterized. In addition, the facility and space for performing the spray drying of (Th,U)O₂ has been prepared, and the bioassay methods have been established.

In Task 2, Metal Matrix Development and Fuel Fabrication Development, fabrication experiments were completed to investigate processing factors such as surrogate oxide particle size, metal matrix particle size, minimum and/or optimum metal matrix content, mechanical mixing techniques, and drawing methodology. The powder-in-tube drawing techniques appear appropriate for consolidation and forming of the fuel pins. Microscopic analyses of finished parts show a sufficient degree of oxide and metal mixing can be achieved mechanically. However, the use of similar sized oxide and metal precursors seems to preclude the desired phase distribution of isolated oxide particles within a continuous metal matrix. Efforts are focused on methods to coat the oxide particles with metal prior to in-tube processing.

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For Task 3, Neutronics and Thermal Design Analysis, research has focused on code acquisition and benchmarking, on preliminary fuel pin neutronics design, and on fuel thermal performance modeling. The fuel lattice code HELIOS was acquired and successfully benchmarked with a Monte Carlo burnup code. Preliminary neutronics design of a thorium metal matrix fuel pin was then performed with the HELIOS code. A tight pitch, “fat” fuel pin was designed with 60 percent heavy metal and 40 percent Zirconium. Slightly different pin designs were developed for pressurized and boiling water reactors, but both designs produced a burnup of greater than 100 GWd/t for a 3-batch core. The fuel thermal performance modeling work involved developing an effective conductivity model for metal matrix composite fuel types. A computer code was developed and was used to perform fuel temperature profiles for use with the HELIOS neutronics code.

Planned Activities

Regarding Task 3 efforts, neutronics design work will continue and will include detailed fuel assembly and equilibrium cycle core burnup calculations. Task 4, Property, Behavior, and Performance Assessment, preliminary activities are underway to prepare for the measurement of fuel pin properties and performance modeling. Purdue University has obtained a metal matrix version of the fuel performance code, DART, and will begin adapting the code for the proposed fuel type. In addition, an examination of the literature related to interactions of uranium and thorium dioxides with zirconium and Zircaloy is being conducted to guide future investigations into the potential interaction between the (Th,U)O₂ microspheres and the zirconium matrix.

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Fundamental Mechanisms of Corrosion of Advanced Zirconium-Based Alloys at High Burn-up

PI: Randy G. Lott, Westinghouse Electric Company LLC

Collaborators: The Pennsylvania State University; Argonne National Laboratory – West; Idaho National Engineering and Environmental Laboratory

Project Start Date: August 1999 Projected End Date: September 2002

Project Number: 99-0128

Research Objective

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 burnup core designs. The objective of this project 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 burnup) and predict the effects of changes in operating 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 goal of this project is to identify these differences and understand how they affect corrosion behavior. To do this, several microstructural examination techniques 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) are being employed.

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 project is to link the characterization and theoretical modeling efforts to yield improved alloy specifications.

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Research Progress

The effort in the first year of the project has focused primarily on developing the procedures, tools, and expertise required to accomplish the project objectives. The microstructural examination studies have pioneered new techniques for characterizing corrosion oxides. An experimental facility for studying radiolysis has been designed and is being fabricated. A structure for a comprehensive corrosion model has been developed and the base model is operational. These efforts provide the basis for successful completion of this three year project.

A unique aspect of the project is the development and use of novel techniques. Project researchers from Pennsylvania State University and Westinghouse are leading an effort to characterize oxides on zirconium alloys using synchrotron radiation. Interfaces at the Advanced Photon Source at Argonne were established along with the completion of a key milestone when the project was granted use of a critical beamline following a competitive peer review of the proposed work. An initial micro-diffraction run was completed. In that run, a narrowly focused 0.25 μ by 2 μ beam was scanned across a cross-sectioned oxide to characterize the crystal structure as a function of depth. The studies indicate that the oxide is predominantly monoclinic with a small amount of tetragonal oxide also present. The tetragonal oxide is concentrated near the metal/oxide interface.

Corrosion oxides have also been characterized using EIS. There is a direct relationship between the electrical properties of the oxide and the corrosion process. The EIS studies indicate the presence of both a continuous layer of highly insulating oxide at the inner surface and a more porous deteriorated oxide on the outer surface. The project is now positioned to characterize the corrosion oxide of different zirconium-based alloys with varying corrosion resistance using this technique and the work has been initiated.

The physical structure of the corrosion oxides can be examined by TEM. However, preparation of appropriate TEM specimens of these oxides requires carefully developed procedures. To reveal the structure of the oxide, TEM specimens must be prepared in cross-section with the extent of the thin area in a plane perpendicular to the corroding surface. Techniques for successfully preparing cross-sectional specimens were developed. Preliminary studies reveal an extremely fine grain structure with a combination of equi-axed and columnar grains.

The effect of radiolysis on zirconium corrosion is characterized by the local environment rather than the bulk water conditions. The pores and microcracks that form in the oxide layer provide a pathway for water intrusion that create a local aqueous environment. A conceptual design for an experiment facility that simulates the local aqueous environment within the porous oxide film was completed.

A modeling effort was undertaken to provide a framework for relating the results of the characterization work to their impact on corrosion. The basic structure of the corrosion model has been implemented and tested. The modular structure of the model provides

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flexibility to incorporate individual effects, such as heat flux and continuous oxide growth on corrosion behavior.

Planned Activities

Progress during the first year of the program has provided the technology to conduct detailed characterizations of corrosion oxides. The primary objective of the second year will be to utilize this technology to characterize oxides on a variety of Zr alloys. Experience with preliminary studies indicates that it is important to understand both the effects of alloy type and the corrosion environment on the type of oxide that forms. APS, EIS and TEM techniques developed in the first year will be employed to characterize oxides grown in both pure water, steam and Li doped water. Characterizations of existing alloys will be completed and the number of materials under consideration expanded. Where required, additional autoclave exposures will be initiated to provide appropriate sources of oxide specimens.

The current model considers growth of the continuous oxide and breakdown of the oxide in separate modules. The primary outputs of these modules are the continuous oxide growth rate and the critical oxide thickness for breakdown. Emphasis in the second year will shift to modeling of the critical oxide thickness. As data on the oxide structures is accumulated, the model will be adjusted to match the observations.

The oxide characterization and theoretical modeling efforts will eventually produce improved alloy specifications. The project is designed to be evolutionary—lessons learned in the initial phases will be applied in the later phases. This evolutionary process is embodied in the alloy selection and development portion of the project. As the project proceeds, alloy specifications for improved corrosion resistance will be identified.

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

PI: Philip E. MacDonald, Idaho National Engineering and Environmental Laboratory

Collaborators: Argonne National Laboratory, University of Florida, Purdue University, Massachusetts Institute of Technology, ABB-Combustion Engineering Inc., Westinghouse Electric Corp., Framatome Technologies, Siemens Power Corp.

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0153

Research Objective

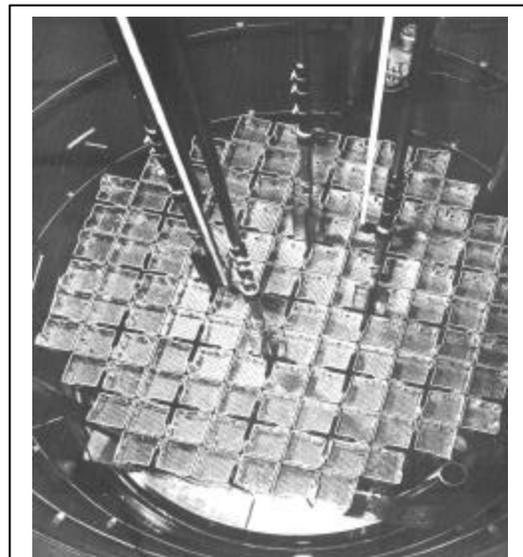
The overall object of this project is to evaluate the efficacy of high burnup mixed thorium-uranium dioxide ($\text{ThO}_2\text{-UO}_2$) fuels for light water reactors (LWRs). A mixed thorium-uranium fuel that can be operated to a relatively high burnup level in current and future LWRs may have the potential 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 .

One of the important goals of this project is to study fuels that would be assembly-for-assembly compatible with the fuel in current LWRs. This implies that both utilities and vendors would find this fuel acceptable for manufacturing and use in current LWRs, if the economics prove to be desirable.

Research Progress

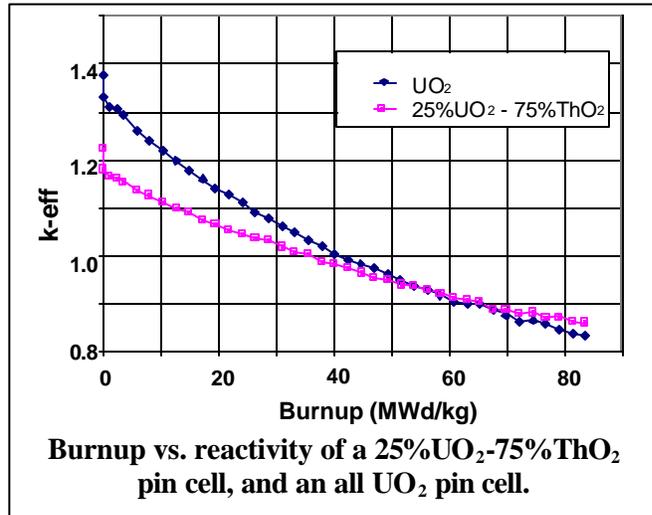
Reactor Core Neutronic Analysis and Fuel Cycle Design: Fuel cycle analysis performed during Phase I included benchmarking several computer models against available critical data (one of the critical experiments is shown to the right) and then analysis to determine the neutronic and economic viability of using a homogeneously mixed, thorium-uranium



$\text{ThO}_2\text{-UO}_2$ Critical Experiment

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dioxide fuel. Several mixed thorium-uranium fuel cases, ranging from a 75 percent ThO_2 -25 percent UO_2 base case to a 65 percent ThO_2 -35 percent UO_2 case, were evaluated for reactivity swing with burnup and isotopic concentrations. These cases were compared to all uranium cases with similar U-235 enrichments in the heavy metal (thorium + uranium, see figure to the right). Preliminary analyses of the enrichment and fabrication costs of the homogeneous thorium-uranium fuels have resulted in a cost per MW-hr about 20-25 percent higher than the all-uranium fuel. While the economics do not favor the homogeneous thorium-uranium fuels that have been studied thus far, other indicators do favor thorium-uranium fuel. For instance, the end-of-life isotopic concentrations strongly favor the thorium-uranium fuel based on total plutonium and minor actinide content, leading to a more proliferation resistant fuel as compared to uranium fuel. Also of interest is the reduction (or elimination) of burnable and soluble poisons needed with a thorium-uranium fuel due to the smaller reactivity swing with burnup. Other factors also show an advantage to using thorium-uranium fuel, such as better fuel performance and waste characteristics, and less coolant corrosion control problems due to the minimal use of soluble poisons. In addition, the preliminary studies on the use of a micro-heterogeneous ThO_2 - UO_2 fuel in LWRs show that this fuel type appears to increase the burnup by 15 percent to 25 percent for the same fissile loading. This increase in burnup is significant enough to close the economic gap, and may show that thorium-uranium fuel can be economically competitive with current all-uranium fuels.



Fuel Manufacturing Costs: First year conclusions are as follows:

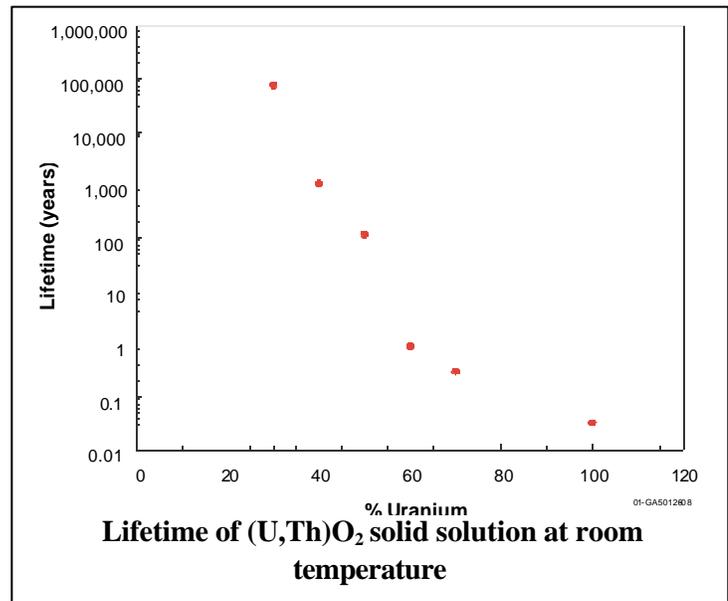
- Current uranium-only nuclear fuel manufacturing facilities can be used to manufacture mixed thorium/uranium fuels.
- Extensive changes are required in the wet conversion areas that manufacture the ThO_2 and the UO_2 to account for the possibility that 20 percent U-235 could be accidentally sent through the conversion line. Due to this rather conservative approach, the resulting facility will be able to handle any fuel composition up to a pure U-235 enrichment of 20 percent.
- Moderate changes will be required in the powder and pelleting areas of the manufacturing facility. The main changes will be in the size of the bulk powder containers. All equipment is currently able to accept powder of any composition up to a pure 20 percent U-235 enrichment level.
- Changes in monitoring for airborne contaminants will be required to account for the much lower Th limit and for the high U activity.

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- There are adequate supplies of thorium available on the commercial markets to support use of a mixed ThO_2 - UO_2 fuel cycle.

Fuel Performance: 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 are being evaluated. During normal operation, ThO_2 - UO_2 fuel may operate with somewhat lower fuel temperatures and internal gas pressures than UO_2 fuel at corresponding powers and burnups. During an accident such as a large break loss-of-coolant accident (LOCA), ThO_2 - UO_2 fuel will have less stored energy but a slightly higher internal heat generation rate than UO_2 fuel at similar power levels. Correlations, as a function of temperature, thorium and uranium concentration, burnup, and other appropriate parameters have been developed from the existing literature for the following material properties: thermal conductivity, specific heat, thermal expansion, emissivity, modulus of elasticity, and melting temperature.

Long Term Stability of ThO_2 - UO_2 Waste: The first year work focused on measuring the kinetic parameters for the air oxidation of $\text{Th}_x\text{U}_y\text{O}_2$ where $x + y = 1$ and x had values of 1.0, 0.7, 0.6, 0.5, 0.4, 0.3, and 0. A non-isothermal thermogravimetric analysis method was used to determine the kinetic parameters. The research showed that air oxidation rates of the solid solution UO_2 and ThO_2 samples were much lower than for pure UO_2 . An example of the results for a 5 percent conversion of various thorium-uranium compositions is shown to the right. Note that material with less than about 50 percent UO_2 lasts for a relatively long time. This reduction in oxidation rate has advantageous storage implications, if air oxidation is the rate-controlling step in transport of spent fuels into the environment.



Planned Activities

- Optimize the fuel form to increase the economic competitiveness of thorium-uranium fuel.
- Determine the projected capital and operating costs of fuel manufacturing.
- Evaluate fuel fabrication issues associated with co-precipitation of the powder and with pressing, sintering, and grinding ThO_2 - UO_2 fuel pellets.

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- Evaluate the behavior of $\text{ThO}_2\text{-UO}_2$ fuel during both normal operation and accident conditions, and compare the results with the behavior of current UO_2 fuel and USNRC licensing standards.
- Determine the corrosion and dissolution rates of both fresh and previously irradiated $\text{ThO}_2\text{-UO}_2$ fuel in synthetic ground water.

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

PI: Hiroshi Takahashi, Brookhaven National Laboratory

Collaborators: Purdue University, Hitachi, Ltd.

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0164

Research Objective

The objective of this research is to advance the well-developed water-cooled reactor technology in order to make efficient use of the abundant thorium resources and enhance the proliferation resistance of the nuclear fuel cycle. Considerable effort has been invested in development of the sodium cooled fast reactor to breed fissionable Pu-239 from natural uranium. Much less effort has been expended in development of alternative technologies that take advantage of the considerable experience with light water reactors (LWR) to provide a hard neutron spectrum for converting thorium into the fissile U-233 isotope.

This project investigates the feasibility of a plutonium-thorium (Pu-Th) fuel cycle for a new type of proliferation resistant, economically competitive, high conversion, boiling water reactor (HCBWR). The technology will be developed to burn existing stocks of plutonium, while converting the fertile thorium to fissile U-233. The high conversion will take place in a fast neutron spectrum, which results from minimizing the volume of water in very tight fuel assembly lattices. High fuel burnup will be possible as a result of the continuous generation and fission of U-233 as the plutonium is consumed. Inherent safety will be designed into the reactor because of the favorable feedback neutronics characteristics of thorium and by the use of innovative core heterogeneities. This will insure a negative void coefficient for those accident sequences, which result in off-normal coolant boiling.

The major technical objective of the proposed project is to develop a reactor design that will:

- Minimize the potential for proliferation of weapons grade materials.
- Maximize the inherent safety features of the reactor.
- Maximize the achievable fuel burnup and plant capacity factor.
- Minimize the cost of electricity generation.

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Research Progress

The light water reactor with a hard neutron energy spectrum has been studied in the past, both in the U.S. and in Europe. More recently, there has been considerable interest in the Japanese nuclear industry and research organizations. This research project has evaluated several of the reactor designs proposed by the Japanese industry and research institutes. These designs included: (1) high conversion BWR, (2) long burnup cycle BWR (LBBWR), (3) BWR without blanket, (4) high conversion pressurized water reactor (PWR), and (5) PWR with Pu multi-cycle.

Detailed analysis of high conversion cores have been carried out in Japan over the last two years and the general conclusions were that in order to achieve high burnup, a reactor with a hard neutron energy spectrum is required. A tight lattice BWR core can provide the hard neutron energy spectrum; however, the coolant void coefficient tended to be positive. In order to achieve safe core operation, the analysis showed that a negative void coefficient could be achieved by enhancing neutron leakage during coolant heat up.

From this research project's preliminary analysis of the above reactors, the LBBWR type reactor has been selected for the initial design study. Because nonproliferation is one of the key factors in the NERI program, thorium will be used as fertile material instead of the U-238 which is used in the Japanese study. Use of thorium as the fertile material can also eliminate fissile plutonium more efficiently. In addition, the thorium fuel cycle does not accumulate a significant inventory of minor actinides that have very long half-lives and as such does not exacerbate the high level waste disposal problem.

Code acquisition and benchmarking work has been completed. Both stochastic and deterministic analysis codes are being used in the effort. The Brookhaven National Laboratory Monte Carlo burnup code MCBURN has been benchmarked for high conversion lattices. The collision probability lattice codes HELIOS was purchased from Studsvik/Scandpower and also benchmarked for thorium depletion with hexagonal lattices. A basic neutronics study was performed comparing performance of thorium and uranium fuels in both standard LWR and high conversion lattices. Preliminary design of the HCBWR fuel pin and fuel lattice was then completed using HELIOS and MCNP. Initial results show very favorable fuel burnup performance as well as good safety characteristics. The void coefficient was negative for all cases examined with thorium/plutonium fuel. Preliminary thermal-hydraulics analysis was also performed for the tight pitch channel using the RELAP5 code. Preliminary results are encouraging but there are concerns about adequacy of constitutive relationships for the tight lattice cores.

Planned Activities

Work has progressed on schedule and some of the tasks on the second year have been initiated during the first year in order to facilitate the analysis. Specifically, the development of a hexagonal lattice capability in the nodal code PARCS was begun in the fourth quarter so that core burnup studies and core safety analysis could be started at the beginning of the second year.

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To study the reactor safety of tight lattice Pu-Th reactor the reactor parameters associated with transient kinetic behavior will be calculated with the MCBURN code and provided in the PARCS code calculation.

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Development of a Stabilized Light Water Reactor (LWR) Fuel Matrix for Extended Burnup

PI: Brady D. Hanson, Pacific Northwest National Laboratory

Collaborators: University of California - Berkeley

Project Start Date: August 1, 1999 Projected End Date: September 2002

Project Number: 99-0197

Research Objective

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 burnup while improving safety margins and reliability for present operations. Burnup is currently limited by the fission gas release and associated increase in fuel rod internal pressure, fuel swelling, and by cladding degradation. Once fuels exceed a threshold burnup, a “rim” or high burnup 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 burnup and even allow higher operating power for increased efficiency.

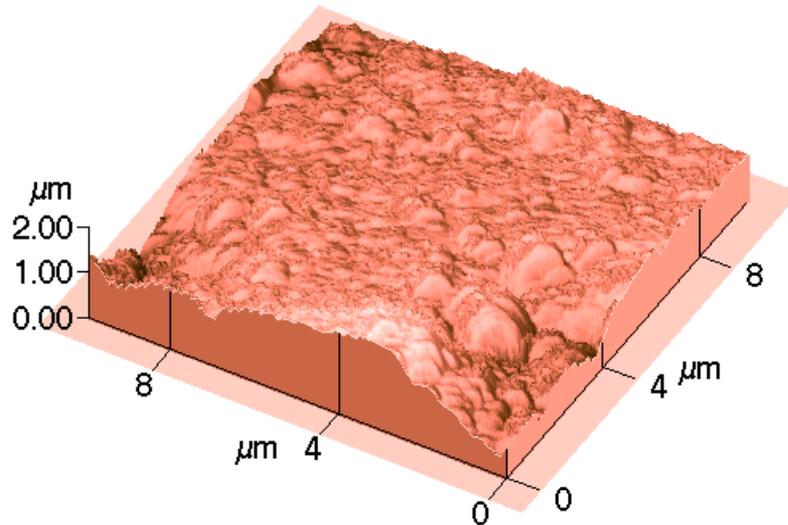
Research at Pacific Northwest National Laboratory (PNNL) has demonstrated that the soluble fission products and actinides present in the matrix of irradiated (spent) fuels stabilized the fuel matrix with respect to oxidation to U_3O_8 . The higher the soluble dopant concentration, the more resistant the fuel has been to undergoing the restructuring of the matrix from the cubic phase of UO_2 to the orthorhombic U_3O_8 phase. In this project, the attempt is to utilize the changes in fuel chemistry that result from doping the fuel to design a fuel that minimized HBS formation. The use of dopants that can act as getters of free oxygen and fission products to minimize fuel-side corrosion of the cladding is also being studied.

In addition to the use of dopants, project researchers are studying techniques such as the use of large grain sizes and radial variations in enrichment to minimize HBS formation and fission gas release. In this project, a combination of modeling and experimental studies is being used to determine the optimum design.

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Research Progress

Task 1 (Understand and Model HBS Formation) focused on performing a comprehensive literature review to guide future efforts. Five different high burnup fuels, including two MOX fuels, were sectioned and prepared for examination of the HBS. Atomic force microscopy (AFM) has been used and appears to be able to map the HBS as a function of radius. An example of the morphology of the HBS as viewed using AFM is seen in the figure below.



**3-D AFM Image (10 x 10 mm) of LWR Spent Fuel
Showing Restructured Grains in the HBS Region**

The successful use of AFM on these highly radioactive samples is believed to be the first such effort. A scanning electron microscope (SEM) and x-ray diffractometer (XRD) were both readied for examination of these spent fuel samples. Modifications to the Resonance Absorption Burnup (RABURN) model, used to calculate the radial concentrations of fission products and actinides, have been made to incorporate true cylindrical geometry.

Task 2 (Develop Matrix Stabilization Model) efforts went into preparing the SEM and XRD units for testing of the spent fuels and candidate fuels. The SEM is now fully operational and project researchers are waiting for their turn to use the equipment. All of the necessary paperwork to adhere to administrative regulations governing the use of fuel enriched in ^{235}U has been completed.

Task 3 (Design and Test Advanced Fuel Matrix) work involved an extensive literature review on the effect of dopants on the UO_2 lattice. This review was aimed at guiding the fuel design efforts. All of the equipment necessary to fabricate pellets based on the “recipes” of dopants developed in the other two tasks has been purchased. Most items,

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including the high temperature tube furnace (for grain growth experiments and sintering of pellets) and the pellet press have been installed. Commercial samples of gadolinia-doped fuels have been ordered for use in baseline tests of physical and chemical properties. The fuels developed for this project will be compared against these baselines. Finally, discussions with Argonne National Laboratory on the use of the Advanced Photon Source or Transmission Electron Microscopy (TEM) facilities to determine the location of dopants in the fuel matrix have been initiated.

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Continuous Fiber Ceramic Composite Cladding for Commercial Water Reactor Fuel

PI: Herbert Feinroth, Gamma Engineering Corporation

Collaborators: Massachusetts Institute of Technology (MIT), McDermott Technology, Inc., Northwestern University, Swales Aerospace, Inc.

Project Start Date: August 1, 1999 Projected End Date: June, 2001

Project Number: 99-0224

Research Objective

The objective of this project is to study the use of advanced ceramic materials as cladding for water reactor fuel elements, and to determine, via engineering type tests, the feasibility of substituting such advanced ceramic materials for the Zircaloy cladding now in use. The ceramic materials to be developed and tested in this research program are known as oxide-based continuous fiber ceramic composites (CFCCs).

Oxide-based CFCCs have three main characteristics that recommend them for water reactor nuclear fuel cladding application. First, because CFCCs consist of very strong, micron sized fibers in a dense ceramic matrix, they do not behave in a brittle manner. Instead they have a failure mode that is non-catastrophic and similar to metals. Second, CFCCs retain their strength to much higher temperatures (e.g. >2000°F) as compared to metals such as zircaloy, which lose much of their strength above 1000°F. And third, oxide-based CFCCs (as opposed to carbide and nitride based CFCCs) remain chemically passive in high temperature steam. Thus, they do not react violently with water during a hypothetical Loss of Coolant Accident (LOCA), they do not produce heat during such an accident, and they do not produce hydrogen gas. Such characteristics, if applied to cladding in commercial water reactors, would lead to significant reductions in the consequences of low probability core overheating accidents, such as LOCAs. This could lead to improved and simplified reactor plant designs, simplified regulatory criteria, and improved public acceptance of nuclear power resulting from real reduction in residual risk.

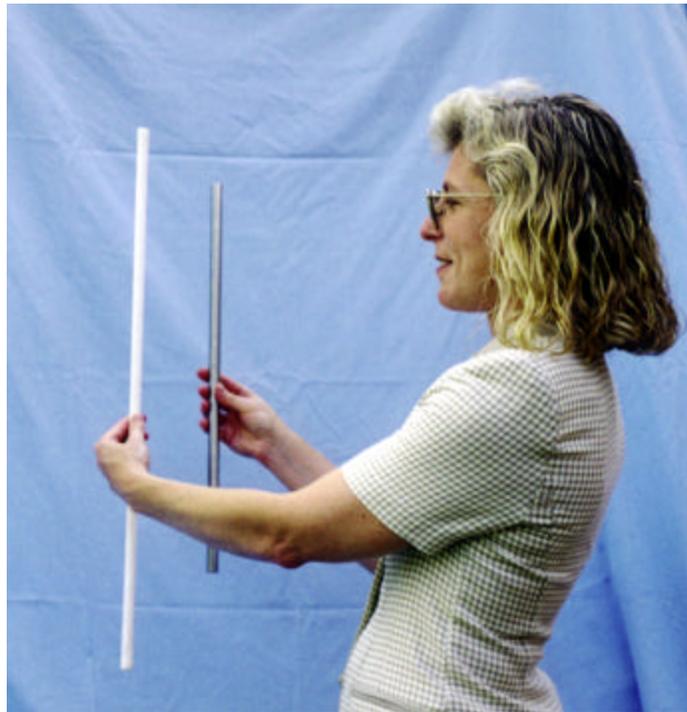
This project seeks to address the feasibility of using CFCCs as reliable fuel cladding. Its aim is to determine the feasibility of providing an improved water reactor fuel element, which is significantly more resistant to damage during a LOCA accident than is the current water reactor fuel element. Specifically, the goals for the project are to (1) evaluate and select two or three specific oxide based CFCC materials which have the potential to meet LWR fuel cladding requirements, (2) fabricate LWR fuel clad test specimens from such materials using advanced CFCC fabrication techniques, (3) conduct in-pile corrosion tests on these specimens, along with standard zircaloy specimens, and

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(4) expose such specimens to simulated LOCA test conditions to confirm their superior performance during LOCA accident conditions.

Research Progress

Approximately sixteen feet of high-density alumina-yttria ceramic composite cladding has been fabricated and delivered to Gamma Engineering Corporation for final sectioning and assembly into the test fixtures at Swales Aerospace and Massachusetts Institute of Technology. A portion of these test samples will be reserved for further characterization and archival storage. The tubes are a composite of high-density alumina fibers (Nextel 610 by 3M), embedded in a matrix of dense yttria-alumina. Some of the tubes (about 4 feet) are then chemical vapor infiltration (CVD)-coated by Northwestern University with either pure alumina, or a mixture of 90 percent alumina, 10 percent chromia to enhance the surface condition and performance of the CFCC material.



Clad Test Samples

MIT completed the design of a test fixture for reliable mounting of ten 3-inch long specimens in a special loop to be inserted in their test reactor. The hardware for the fixture is currently under procurement, and assembly of the fixture with the test specimens was planned during the month of August 2000. Swales has completed the assembly and trial use of a special LOCA test rig designed to quench typical clad specimens (3 inches in length) from high temperature (up to 2500°F) into room temperature water.

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Planned Activities

Phase 2 of this project involves two test programs, and further characterization of the fabricated tubes.

Irradiation Test Program: Ten clad specimens (each about 3 inches long) will be irradiated in a special high temperature (600 °F) flow loop in MIT's 5 MW nuclear reactor.

LOCA - Thermal Shock Test: Three inch long test specimens will be heated in a special furnace to three different "accident" temperatures: 1000°F, 1800°F, 2500°F. After stabilization at these temperatures, each specimen will be quenched in room temperature water in a special apparatus.

Further Characterization Tests: Visual and microstructure examination will be conducted after the irradiation and thermal shock tests. In addition, permeability tests will be performed on some of the pre-irradiated coated specimens to compare with pre-irradiated uncoated specimens.

Thus far, the project has addressed many of the technical issues associated with the use of CFCCs as cladding in commercial reactors. Some of the issues have been resolved, and others remain for further development. A key factor in determining whether further work is warranted on applying this promising new material to nuclear fuel cladding use will be the results of the planned thermal shock and irradiation testing.

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An Innovative Ceramic Corrosion Protection System for Zircaloy Cladding

PI: Ronald H. Baney, University of Florida

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0229

Research Objective

The operational lifetime of current Light Water Reactor (LWR) fuel is limited by thermal, chemical, and mechanical constraints associated with the fuel rods being used to generate nuclear heat. A primary limiting factor of this fuel is the waterside corrosion of the zirconium based alloy cladding surrounding the uranium pellet. This research project intends to develop thin ceramic films with adhesive properties to the metal cladding in order to eliminate the oxidation and hydriding of Zircaloy cladding. The corrosion protection system will allow fuel to operate safely at significantly higher burnups resulting in major benefits to plant safety and plant economics.

A major technical challenge for coating a metal with a ceramic protection system is to develop a cohesive bond between the two materials. The differences between the thermal expansion of the ceramic coating and the thermal expanding metal can, if not properly addressed, interfere with the ceramic's ability to maintain a bond and thus maintain a protective layer.

Research Progress

The following ceramics were investigated: alumina, boron carbide, graphite, mullite, silicon carbide, spinel, tungsten carbide, zirconia, and zirconium carbide. A background search was performed to identify their current uses in industry, processing methods, potential cost, their availability, and their suitability as a protective coating material for Zircaloy cladding in a LWR.

The mechanical properties of the coatings, such as coefficient of thermal expansion (CTE) and thermal conductivity, were compared with that of zirconium (Zircaloy) in order to determine the best candidate. A hand-calculated thermal analysis was performed to show temperature changes owing to the different coatings as a function of coating thickness and the validity of these calculations was established by a comparison with the thermal hydraulic computer program COBRA. A neutronic analysis was also performed using the Monte Carlo N-Particle Transport Code (MCNP) to show reactivity changes and unit fuel cell thermal flux changes caused by the different coatings as a function of thickness. The CASMO computer code was utilized to show burnup effects and changes in plutonium concentrations owing to an alumina coating of 50 to 100 microns. As a

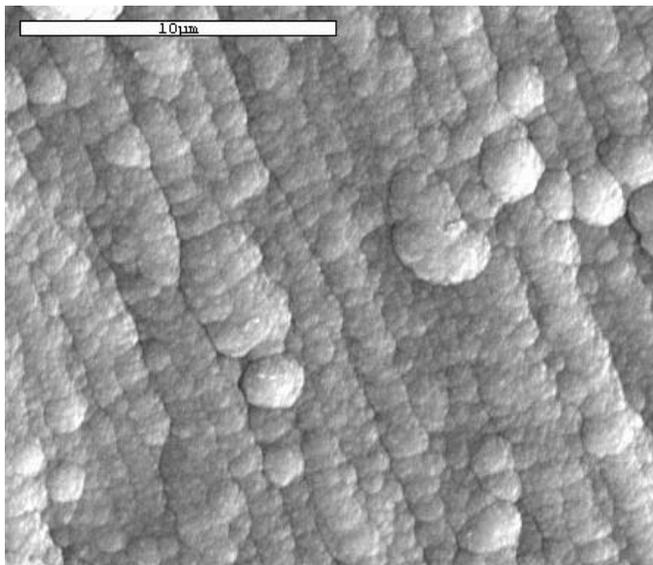
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result of these studies, the two best coating options were identified as alumina and silicon carbide and initial samples were placed in an autoclave to simulate upper limit reactor conditions (water that was at 700F and 3000 PSIA pressure) in order to observe the coatings' corrosive and chemical stability.

Alumina's CTE is within 12 percent of zirconium's throughout the entire temperature range, while silicon carbide has the highest thermal conductivity of 78.8 W/m-K. All of the materials were analyzed thermally in order to calculate the rise in temperature of the fuel centerline, pellet surface, and outer and inner clad surfaces. At 50 microns, none of the coatings caused more than a 0.13 percent rise in cladding temperature and 0.05 percent rise in fuel centerline temperature. Even with a thickness of 500 microns, none of the materials caused more than a 1.5 percent increase in temperatures in any of the regions. Due to the extensive computation time required, only silicon carbide, alumina, zirconium carbide, and zirconia were analyzed neutronicly. At the most likely coating thickness between 10 and 50 microns, there was less than a 0.09 percent decrease in reactivity and a 0.5 percent decrease in the thermal flux in the fuel and a 1.7 percent decrease in the thermal flux in the water.

Owing to the displacement of water caused by the coating, there was a hardening of the spectrum and therefore an increase in plutonium production as the fuel is being used. With a coating of 50 microns of alumina, there was as 1.9 percent increase in Pu-239 at 60 GWD/MTU. This increase in plutonium production has the added benefit of extending the reactivity lifetime of the fuel as plutonium becomes used as fuel.

A variety of coating processes for coating carbon, silicon carbide and alumina onto Zircaloy were explored. Carbon, diamond like carbon (DLC), and silicon carbide (SiC) were coated onto Zirc-4 coupons by plasmas-assisted chemical vapor deposition (PACVD). The figure below shows a SEM image of one such coating.



SEM image of SiC thin films on unpolished Zircaloy deposited at 400C using Silacyclobutane by electron cyclotron resonance (ECR) plasma assisted (PA) chemical vapor deposition (CVD)

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Silicon carbide was also coated by a laser ablation deposition (LAD) process. Alumina was coated by an ultraviolet assisted chemical vapor (UVCVD) process. Alumina was also coated by a spin coating sol-gel process. A thick layer of alumina was coated onto a sheet of Zirc-4 by physical vapor deposition (PVD) using a plasma spray technique.

The quality of the coatings were assessed by visible inspection for uniformity and physical adherence of the coating and by Auger Electron Spectrographic (AES). The quality of the ceramic coating was assessed by the depth of the coating and the thickness of the coating.

Planned Activities

The corrosion testing of alumina and other promising coatings will be studied in detail. Further autoclave testing and accelerated corrosion testing will be required in order to complete a performance analysis of the ceramic coatings applied to zirconium alloy. Further investigation is also required to verify bonding throughout the fuel lifetime for the various coating materials. Coating processes will be optimized to give the highest quality coatings.