

NUCLEAR ENERGY RESEARCH INITIATIVE: THORIUM FUEL CYCLE PROJECTS*

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Abstract. The United States (U.S.) Department of Energy (DOE) is conducting four projects involving the use of the thorium fuel cycle. All four projects are based on an once-through, proliferation resistant, high burnup, long refueling cycle use of thorium in a light water reactor. Three of these projects are part of the Nuclear Energy Research Initiative (NERI) program. These are: "Advanced Proliferation Resistant, Lower cost Uranium-Thorium Dioxide Fuels for Light Water Reactor," with Idaho Nuclear Engineering and Environmental Laboratory as the lead organization; "Fuel for a Once-Through Cycle (Th,U)O₂ in a Metal Matrix," with Argonne National Laboratory as the lead; and "A Proliferation Resistant Hexagonal Tight Lattice BWR Fuel Core Design for Increased Burnup and Reduced Fuel Storage Requirements," with Brookhaven National Laboratory (BNL) as the lead. The fourth project is "The Radkowsky Thorium Fuel project," also under BNL lead. This paper describes the three NERI thorium fuel cycle projects.

INTRODUCTION

A new approach to the Department of Energy's (DOE) conduct of nuclear energy research and development (R&D) was recommended by the President's Committee of Advisors on Science and Technology (PCAST) "Panel on Energy Research and Development," in November 1997. As a result, DOE is making a fundamental change in the management of its nuclear energy research activities. DOE's new approach was initiated with the Nuclear Energy Research Initiative (NERI), which was started in 1999. NERI features a competitive, peer-reviewed, R&D selection process to fund researcher initiated R&D proposals from the universities, national laboratories, and industry. NERI receives guidance from the Nuclear Energy Research Advisory Committee (NERAC). NERAC's primary function is to assist DOE in effectively carrying out its role in nuclear energy research. The advisory committee consists of expert members from a wide variety of research backgrounds and perspectives.

The objective of the NERI program is to address and help overcome the principal technical and scientific obstacles to the future use of nuclear energy in the United States. These obstacles include issues involving proliferation, economics, nuclear waste, and safety. Technologies addressed by NERI include, but are not be limited to, work on proliferation-resistant reactors or fuel cycles; new reactor designs with higher efficiency, reduced cost, and enhanced safety to compete in the global market; lower output power reactors for applications where larger reactors may not be advantageous; and new techniques for on-site and surface storage and for permanent disposal of nuclear waste. NERI is also expected to help preserve the nuclear science and engineering infrastructure within the universities, laboratories, and industry to advance the state of nuclear energy technology and to maintain a competitive position worldwide. DOE believes that by funding creative research ideas under NERI, solutions to important nuclear issues will be realized, and a new potential for nuclear energy in the United States will emerge.

* 1999 meeting.

The budget requests and Congressional appropriated funding for the NERI program to date are the following:

In Fiscal Year 1999, the Administration requested \$24 million for NERI and Congress appropriated \$19 million.

In Fiscal Year 2000, the request was \$25 million and Congress appropriated \$22.5 million.

In the first year of the NERI program, DOE made awards for 46 projects. This involved issuance of 53 grants and 38 Interoffice Work Orders (IWOs). For information on the awards and the recipients see the NERI web site at "<http://neri.ne.doe.gov>."

Three of these NERI projects involve the thorium fuel cycle. These are:

- The Advanced Proliferation Resistant, Lower Cost, Uranium-Thorium Dioxide Fuels for Light Water Reactors Project. The lead organization for this project is the Idaho National Engineering and Environmental Laboratory (INEEL). The Principle Investigator is Phillip E. MacDonald.
- The Fuel for a Once-Through Cycle (Th,U)O₂ in a Metal Matrix Project. The lead organization for this project is Argonne National Laboratory (ANL). The Principle Investigator is Sean McDeavitt.
- A Proliferation Resistant Hexagonal Tight Lattice BWR Fuel Core Design for Increased Burnup and Reduced Fuel Storage Requirements. The lead organization for this project is Brookhaven National Laboratory (BNL). The Principle Investigator is Hiroshi Takahashi.

BACKGROUND

These thorium fuel cycle projects differ from earlier thorium fuel cycle work conducted in the U.S. to develop thorium cycle converter-reactor systems. Several prototypes, including the HTGR (high-temperature gas-cooled reactor) and MSRE (molten salt converter reactor experiment), have operated. A uranium-thorium seed blanket fuel arrangement was also used to demonstrate the light water breeder concept at the Shippingport Atomic Power Station. This reactor operated for five years from August 1977 to October 1982. At the end of this period, the core contained approximately 1.3 percent more fissile material after producing heat for five years than it did before initial operation. The only U.S. commercial thorium/uranium fueled HTGR was the Fort St. Vrain reactor near Platteville, Colorado. The reactor, with a capacity of 330 MW(e), began full operation in early 1979. The operation of this full-scale commercial HTGR was marked by intermittent operations resulting in low capacity factors.

While uranium technology in light water reactors has been demonstrated to be very dependable, the use of thorium technology has lagged ever since the closure of the Fort St. Vrain commercial HTGR in 1989. All currently operating commercial nuclear power plants in the United States use uranium

ADVANCED PROLIFERATION RESISTANT, LOWER COST, URANIUM-THORIUM DIOXIDE FUELS FOR LIGHT WATER REACTOR PROJECT

In addition to the lead organization, INEEL, with Phillip E. Macdonald as the Principle Investigator, the following organizations and investigators are participating in this project:

ABB Combustion Engineering Inc., George P. Smith, Jr.

Argonne National Laboratory, Dr. James C. Cunnane

Framatome Technologies, Steward W. Spetz

Massachusetts Institute of Technology (MIT), Prof. Mujid S. Kazimi and Prof. Michael J. Driscoll

Purdue University, Prof. Alvin Solomon

Seimens Power Corporation, Dr. Leo F. P. Van Swam

University of Florida, Prof. James S. Tulenko

Westinghouse Electric Corporation, Dr. E. J. Lahoda

The goal of this project is to develop a ThO₂-UO₂ fuel that is assembly-for-assembly compatible with existing light water reactors (LWRs). The fuel will be developed for a once-through fuel cycle in which in-reactor conversion of ²³²Th to ²³³U is maximized and plutonium production is minimized. The fuel will be taken to higher burnup than planned in previous work. No chemical processing of the fuel is considered. The durability of the fuel as a wasteform is important.

The objective of this project is to develop a fuel for the existing LWRs that is less expensive to fabricate than the UO₂ fuel, allows longer refueling cycles and higher sustainable plant capacity factors, is very resistant to nuclear weapons-material proliferation, results in a more stable and insoluble waste form, and generates less high level waste.

The fuel cycle economics of the fuel being investigated is influenced by a number of factors. Extended burnup reactor cores using conventional UO₂ fuel require high ²³⁵U enrichments and significant quantities of burnable poisons for reactivity control, which significantly increases costs. However, the reactivity in a ThO₂-UO₂-fueled reactor remains more constant during long irradiations than in a UO₂ core because of the high conversion ratio of the thorium. Calculations using the MOCUP code system indicate that the mixed ThO₂-UO₂ fuel, with about 5.8 wt% of the total heavy metal ²³⁵U, could be burned to 72 MW·d/kg using 30 wt% UO₂ and the balance ThO₂. The ThO₂-UO₂ cores can also be burned to about 86 MW·d/kg using 35 wt% UO₂ and 65 wt% ThO₂ with an initial enrichment of about 6.8 wt% of the total heavy metal fissile material.

Longer refueling cycles and higher plant capacity factors can be achieved with this fuel. ThO₂-UO₂ fuel has a significantly higher thermal conductivity at LWR operating temperatures and a lower rate of fission gas release. Therefore, ThO₂-UO₂ fuel can be operated to higher burnup with less difficulty than UO₂ fuel. With improved fuel, many of the U.S. plants could move to 24- to 36-month refueling cycles. An improvement to 24-month cycles is worth about 2.5 percent in plant capacity and an improvement to 36-month cycles would increase plant factors by about 5 percent. Having the same plants generate 5 percent more electricity would save U.S. utilities and thus taxpayers about \$1 billion per year.

This thorium fuel cycle also offers a high level of nuclear weapons-material proliferation resistance. The uranium is calculated to remained below 20 wt% total fissile fraction throughout the cycle, making it unusable for weapons. Total plutonium production per MWd was a factor of 3.2 less in the ThO₂-UO₂ fuel than in the conventional fuel. ²³⁹Pu production per MWd was a factor of 4.2 less in the ThO₂-UO₂ fuel than in the conventional fuel. The plutonium produced

was high in ^{238}Pu , leading to a decay heat rate 3.7 times greater than that from plutonium derived from conventional fuel and 29 times greater than that from weapons grade plutonium. The decay heat in spent $\text{ThO}_2\text{-UO}_2$ fuel is high enough to melt and render ineffective the explosives commonly used in nuclear weapons, unless the weapon is actively cooled. Spontaneous neutron production for plutonium from $\text{ThO}_2\text{-UO}_2$ fuel was 1.75 times greater than that from conventional fuel and 12 times greater than that from weapons grade plutonium. High spontaneous neutron production drastically limits the probable yield of a crude weapon.

The fuel investigated in this project has improved waste form stability. Spent UO_2 fuel fragments react and disintegrate relatively rapidly (about 1 percent per year) with water containing Yucca Mt. contaminants. ThO_2 is the highest oxide of thorium and does not depart significantly from its stoichiometric composition when exposed to air or water at temperatures up to 2000 K. Heavily oxidized high thoria solid $\text{ThO}_2\text{-UO}_2$ solutions contain urania structures only between UO_2 and U_4O_9 and, therefore, retain their mechanical integrity. The thoria stabilizes the UO_2 and prevents oxidation beyond U_4O_9 .

The fuel investigated also has a high level of waste minimization. Use of higher burnup fuel will result in proportionally fewer spent fuel bundles to handle, store, ship, and permanently dispose. The facility operating portion of the planned system to dispose of the nation's spent nuclear fuel and high-level waste has been estimated to be about \$13.6 billion over about 40 years, or \$32,000 per Boiling Water Reactor (BWR) fuel bundle and \$60,500 per Pressurized Water Reactor (PWR) fuel bundle. Approximately 4,000 BWR and 3,400 PWR fuel assemblies are discharged each year in the United States. If the equilibrium cycle discharge burnups in the United States could be increased to 75 MWd/kg, for example, the government could save more than \$100 million per year.

This project includes four tasks.

Task 1: Fuel-Cycle Analysis will evaluate the economic viability of a $\text{ThO}_2\text{-UO}_2$ fuel cycle in commercial reactors operating in the U. S. Framatome Technologies will add cross-sections for thorium and related isotopes to its SCIENCE nuclear code package and then perform two- and three-dimensional fuel-lattice calculations and calculate power distributions in a typical PWR 17×17 core. Finally, costs for $\text{ThO}_2\text{-UO}_2$ and conventional uranium cycles will be compared. MIT will try to further optimize the core design by investigating such things as fuel rod geometry, metal-water ratio, and $\text{ThO}_2\text{-UO}_2$ ratios using the CASMO-4 and SIMULATE lattice codes. Both MIT and the INEEL will perform benchmark quality calculations at the rod, cell, and assembly levels using the Monte Carlo code MOCUP, which combines MCNP and ORIGEN.

Task 2. Fuel Manufacturing Costs will determine if the current nuclear fuel fabricators in the U.S. have the capability to manufacture $\text{ThO}_2\text{-UO}_2$ fuel economically. Westinghouse will generate process flow sheets; identify equipment, process, safety, and licensing issues and the required plant modifications to current uranium based manufacturing facilities; and determine the projected capital and operating costs. Criticality and radiological safety are particularly important issues that must be addressed for this type of fuel. Purdue will evaluate fabrication issues associated with co-precipitation of the powder and with pressing, sintering and grinding $\text{ThO}_2\text{-UO}_2$ fuel pellets and investigate manufacturing techniques to produce low cost fuel.

Task 3. Fuel Performance will evaluate the thermal, mechanical, and chemical aspects of the behavior of $\text{ThO}_2\text{-UO}_2$ fuel rods during normal, off normal, and design basis accident conditions. $\text{ThO}_2\text{-UO}_2$ fuel has different properties than UO_2 fuel:

- slightly higher decay heat,
- higher thermal conductivity at normal operating temperatures and lower thermal conductivity at high temperatures,
- higher fission gas production per fission, but a lower rate of release of fission gases, and
- higher melting temperature.

Three organizations will be involved in the evaluation of the performance of ThO₂-UO₂ fuel: INEEL, MIT, and Purdue. Purdue will make additional material property measurements including thermal conductivity, creep, and gas induced swelling. MIT will develop a fission gas release model for ThO₂-UO₂ fuel and evaluate innovative ThO₂-UO₂ fuel designs. All three collaborators will do the property correlation work and some steady state analysis; the final transient analysis will be done at the INEEL.

Task 4. Long-Term Stability of ThO₂-UO₂ Waste will determine whether thorium-uranium fuel is superior to uranium as a fuel waste form. The objective in Year 1 of this task is to determine the oxidation rates in air and in oxygen saturated water of ThO₂-UO₂ fuels with various ratios of thorium and uranium. The objective in Year 2 is to determine the corrosion and dissolution release rates of ThO₂-UO₂ fuel in synthetic ground water. These experiments will be continued in Year 3, along with experiments in a hot cell with 50 MW/kg Shippingport fuel to benchmark the out-of-pile work. The cold laboratory work will be done at the University of Florida, and Argonne will do the hot cell work in collaboration with the University of Florida.

FUEL FOR A ONCE-THROUGH CYCLE-(TH,U)O₂ IN A METAL MATRIX PROJECT

The lead organization for this project is Argonne National Laboratory, and the Principle Investigator is S. M. McDevitt. He is assisted by M. C. Hash. In addition to ANL, Purdue University is participating in this project. The Purdue investigators are A. A. Solomon, T. J. Downar, & S. T. Revankar.

The concept for this fuel is a dispersion of (Th,U)O₂ particles that are 50 to 100_μm diameter. The fuel would have a density of 80 to 90 percent and is expected to have low swelling. The fuel particles would be dispersed in a zirconium matrix that has high density, high thermal conductivity, and provides fission product containment. The fuel matrix would be enclosed in a tubular Zircaloy shell that would serve as the powder packing form. The shell would be drawn for the proper density and shape and would be compatible with current Light Water Reactors. The concept is shown in the following Figure 1.

There are two tasks for this 3 year NERI project:

Task 1. Proof-of-Principle Activities will include ceramic microsphere fabrication, dispersion fuel rod fabrication, and fuel modeling of neutronic and thermal properties.

Task 2. Fuel Performance Estimates will be based on past data from dispersion fuels. It will include performance modeling, bounding calculations, and preparation for irradiation experiments.

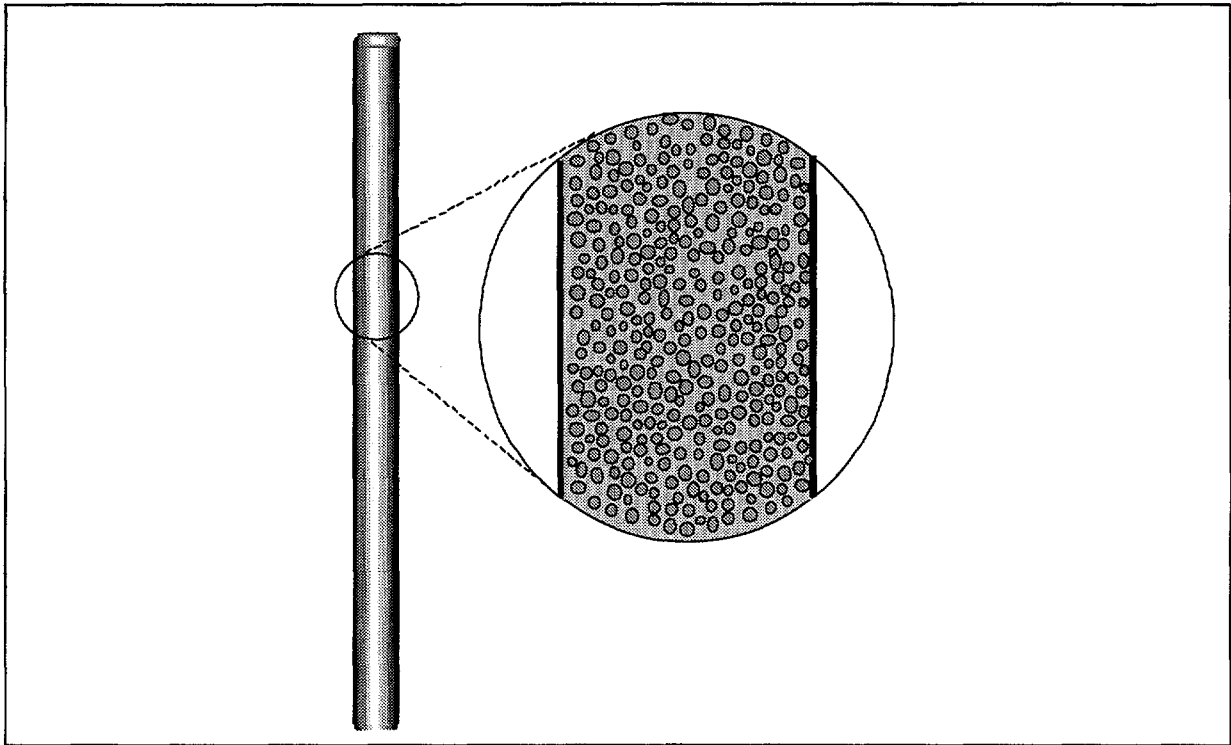


Figure 1. (Th, U)O₂ Metal Matrix Concept

The potential benefits of the metal matrix fuel include:

- High Actinide Burnup. The ²³²Th to ²³³U conversion extends the fuel life.
- Proliferation Resistance. Mixed oxides prevent direct chemical separation of ²³³U and ²³⁹Pu.
- Improved Irradiation Stability. Reduced centerline temperature results in stronger physical properties.
- Minimal Waste Treatment. The concept uses direct disposal of spent fuel from the once-through cycle.
- Low Fabrication Cost. Low temperature and simple industrial methods can be used.

PROLIFERATION RESISTANT HEXAGONAL TIGHT LATTICE BOILING WATER REACTOR (BWR) FUEL CORE DESIGN FOR INCREASED BURNUP AND REDUCED FUEL STORAGE REQUIREMENTS PROJECT

The lead organization for this project is Brookhaven National Laboratory, and the Principle Investigator is Hiroshi Takahashi. Upendra S. Rohatgi is also a BNL investigator. Other participating organizations are Purdue University and Hitachi Ltd. The investigator for Purdue University is Thomas J. Downar.

The design objectives of the High Conversion, Boiling Water Reactor (HCBWR) concept are to achieve a high conversion of Th to ²³³U, reduce accumulated inventory of plutonium while producing useful energy, develop very high burnup BWR fuel using a high concentration of plutonium and a large rate of ²³³U production, minimize potential for proliferation of weapons grade fissionable materials, maximize inherent safety features of reactor, maximize plant capacity factor, and minimize cost of electricity generation.

The HCBWR is a proliferation resistant, economically competitive concept. It has a very tight lattice with relative small water volume fraction and will operate with a fast reactor neutron spectrum. It has a radially and axially segmented core design. A thin annulus of neutron moderating and absorbing materials separating core and blanket segments provides negative reactivity feedback for high core voiding. Preliminary design study parameters for the HCBWR Reactor are given in the following Table I.

Table I. HCBWR Reactor. Preliminary Design Study Parameters

Parameter	Value
Reactor Type	Boiling water cooled Pu oxide Th-233 U oxide high burn up fast reactor
Core Layout	Segmented design (radial and axial) tight hexagonal lattice
Power Level	Range: 600 MW(e) to 1350 MW(e)
Primary System Pressure	~ 8 MPa
Fuel Material	Pu oxide fuel and ^{233}U plus thorium fertile (^{233}U) oxide
Blanket Material	Thorium oxide
Coolant	Boiling water
Blanket Design	(1) Radial and axial blankets (2) Internal blankets
Working Fluid	Water and superheated steam

The HCBWR is expected to have a very high proliferation resistance. The design is constrained so that no natural uranium is incorporated into the fuel feedstock at any time in the fuel cycle. Uranium-233 produced from conversion will not be separated from other isotopic products. The Uranium-232 will be retained with the Uranium-233 to provide handling difficulty resulting emitted radiation and internal heat generation.