

HTR2008-58325

PROJECT DEEP-BURN: DEVELOPMENT OF TRANSURANIC FUEL FOR HIGH-TEMPERATURE HELIUM-COOLED REACTORS

Robert M. Versluis
U.S. Department of Energy
Germantown, MD

Francesco Venneri
LOGOS Technologies
Arlington, VA

David Petti
Idaho National Laboratory, ID
Idaho Falls, ID

Lance Snead
Oak Ridge National Laboratory
Oak Ridge, TN

Donald McEachern
General Atomics
San Diego, CA

1. INTRODUCTION

The helium-cooled, graphite-moderated Very High Temperature Reactor (VHTR) has become the centerpiece of the U.S. Department of Energy's (DOE) Next Generation Nuclear Plant (NGNP) program. The NGNP program aims to construct a VHTR prototype, with the participation of industry, by the year 2021.

The NGNP program has made significant strides in the development of the reactor for both high-efficiency electric power and high-temperature process heat applications, however, the fuel cycle role of the VHTR has not received the same kind of attention. Studies of the VHTR fuel cycle must involve: (a) use of light-water-cooled reactor (LWR) spent fuel as kernel feedstock; (b) recycle of spent VHTR fuel; (c) use of the VHTR in the management of transuranics (TRU); and, (d) the geologic storage performance of spent VHTR fuel. Examination of these aspects of the fuel cycle and defining a resolution path for any technology gaps is essential for determining future deployment options of the VHTR.

This paper describes a project initiated by the Office of Nuclear Energy (NE) to establish the technological foundations supporting the role of the VHTR in the nuclear fuel cycle. The project will include a quantitative assessment of the scope, cost, and schedule implications of incorporating various fuel cycle considerations into a prototype-VHTR project. The work will be coordinated with other NE programs to ensure synergism and to avoid duplication of efforts.

2. BACKGROUND

Deep-Burn. The concept of destruction of spent fuel TRU in a TRISO-fueled (TRIStructural ISotropic) gas-cooled reactor is known as Deep-Burn. The term "Deep-Burn" reflects the large fractional burnup of up to 60-70% fissions per initial metal atoms (FIMA) that may be achievable with a single-pass, multi-cycle irradiation in VHTRs. Spent TRISO

fuel from Deep-Burn can be either placed directly into geologic storage to provide long-term containment of the residual radioactivity, or recycled into fast reactor fuel. Deep-Burn rapidly and effectively reduces the inventory of TRU from spent fuel without the need for repeated re-cycles, destroys weapons-usable materials contained in spent fuel, and precludes the possible weapons-related use of the residuals thereby providing strong proliferation barriers. In the past three years, with the help of neutronics models developed for the gas-cooled reactor, the physics of the Deep-Burn VHTR (DB-VHTR) concept has been thoroughly analyzed and to some extent validated.

The DB-VHTR concept is particularly attractive because it employs the same reactor design that is used for the NGNP program, with the same potential for highly efficient electricity and hydrogen production. All engineering elements of the DB-VHTR concept that relate to the reactor core and the power production are common to the NGNP and are being addressed in the NGNP program and the National Nuclear Security Administration's (NNSA) Gas Turbine Modular Helium Reactor (GT-MHR) program for weapons-plutonium disposition.

TRISO Fuel. The key to the VHTR behavior and performance with respect to TRU management lies in its TRISO fuel. This fuel form is made of small particles having a 200-400 micron diameter core composed of heavy metals (HM), surrounded by carbon and silicon carbide structural layers that can contain the fuel and fission products under all postulated normal and off-normal reactor events, thus giving rise to the "inherently safe" character of the reactor concept.

During Deep-Burn irradiation, the various TRU constituents of the Deep-Burn TRISO (DB-TRISO) fuel particle are progressively destroyed according to their cross section for fission or capture of neutrons. For instance, already at the 50% FIMA burnup level, almost all the Pu-239 (~90%) has already been destroyed thereby eliminating the

risk of future diversion of spent fuel for weapons production. Of crucial importance to the Deep-Burn concept is the interplay between Pu-239, Pu-240 and Pu-241. Pu-239 is a strong fissile isotope, while Pu-240 is a strong burnable poison that is also a fertile isotope producing Pu-241. The utilization of the short-lived Pu-241 in the high-burnup TRISO fuel without reprocessing is a special feature of Deep-Burn. The sequence 239-240-241 produces a very steady reactivity behavior over long periods of time leading ultimately to the achievement of better than 60% FIMA burnup without exceeding allowable TRISO fuel fluence limits.

Fuel Cycle. Although the DB-TRISO fuel shares common elements with the TRISO fuel proposed for the NGNP and the NNSA's Plutonium (Pu)-TRISO fuel, TRISO fuel is not being manufactured in the U.S. at present in commercial quantities. There is however a considerable experience base for large-scale manufacturing of high-quality fuel both in and outside the U.S., and the NGNP program is in the process of re-establishing this capability in the U.S.. Manufacturing of DB-TRISO will require the additional implementation of remote handling techniques and the development of fuel designs specifically geared to very high burnup.

Spent TRISO fuel, be it from VHTR Low-Enriched Uranium (LEU) operations or fabricated from recycled LWR-TRU fuel, can be directly disposed of in a repository, or further recycled in fast reactors. For the geologic disposal option, a clear understanding of the performance of TRISO fuel in the repository will be required, so that a repository strategy for TRISO fuel can be developed taking into account, on one hand, the larger volumes and, on the other hand, the enhanced capability to retain the radioactive isotopes. In the recycle option, the addition of a TRISO head-end process, such as grind-and-leach or METROX, to the main partitioning schemes (UREX+ or PYRO) would be required and the relevant technology would need to be developed.

Preliminary studies show that up to 20 times more energy can be extracted from Deep-Burn TRISO (DB-TRISO) fuel in VHTRs than from mixed oxide (MOX) fuel in LWRs. If these results can be validated, the Deep-Burn concept would create a completely different paradigm for the near-term economics of closed fuel cycles. The cost of spent LWR fuel reprocessing would be partially or fully offset by the value of the recovered TRU in a Deep-Burn VHTR (DB-VHTR) producing power at competitive cost.

3. OBJECTIVES

Establishing fuel cycle options for the VHTR would enable determining the VHTR role in any global nuclear growth scenario, both for once-through and recycle options. It is likely that this would provide cost-effective recycle options for LWR spent fuel with a minimum of reprocessing and rapid and significant reduction of spent-fuel TRU stockpiles, particularly the weapons-usable fraction.

The current project aims at establishing the technological foundations supporting the role of the VHTR in the nuclear fuel cycle. This includes a quantitative assessment of the scope, cost, and schedule implications of incorporating various fuel cycle considerations into a prototype-VHTR project. The

current statement of work, which was started in mid-2008 on one-year funding, lays the groundwork for additional awards if additional funding becomes available in the future. The medium-term objectives, which would take several years to achieve, are:

- Analysis of the DB-VHTR as a TRU burner, including neutronics, thermo-hydraulics and safety aspects;
- First-principle model of DB-TRISO fuel for incorporation in the VHTR design tool;
- Production of sufficient amounts of high-quality TRU-bearing DB-VHTR fuel to begin irradiation experiments;
- Flowsheet development for aqueous and pyro-chemistry reprocessing methods for VHTR spent fuel;
- Quantitative assessment of waste management and geologic storage options for VHTR spent fuel;
- Cost analysis and roadmap development for the VHTR fuel cycle options, including recommended interfaces with NGNP and GNEP/AFCI programs.

4. SCOPE OF WORK

Neutronic, Thermo-Hydraulic, and Safety Analysis of the DB-VHTR as a TRU Burner.

Gas reactors have an advantage over light-water reactors in terms of their ability to burn TRU because the use of robust particle fuel, a solid moderator and a neutronically transparent coolant enables the use of fully enriched TRU TRISO fuel, and the attainment of very high burnups (~ 500,000 – 700,000 MWD/tHM). Thus, the overall amount of TRU burned in a single recycle can be much greater in a DB-VHTR than an LWR. In addition, the higher thermal efficiency of the VHTR increases the amount of electricity produced during consumption of the TRU. Preliminary assessments of the DB-VHTR indicate that fuel cycle lengths of 1 to 1.5 years are feasible and that the reactivity swing over the cycle could be managed. There is a need for additional analysis to develop a self-consistent fuel management, thermal-hydraulic, and fuel performance approach, including burnable poisons, for the DB-VHTR to determine that the TRU burnups of such magnitude are achievable with sufficient margins of power peaking, peak fuel temperature, and fast fluence to acceptable limits.

Core Design Studies. Using generic NGNP-type prismatic and pebble-bed reactor cores, the project will perform physics calculations using benchmarked models to evaluate the capabilities of this reactor to perform destruction and utilization of LWR spent-fuel TRU and to improve the performance of the geologic repository or synergistically complement fast recycling reactors. The objectives are to establish DB-VHTR concept feasibility and optimize fuel management performance; continue analytical work on the design of the DB-VHTR; and to produce required data for fuel cycle evaluation (mass flows, inventories, operational parameters).

The work will factor in preliminary feasibility work on the Deep-Burn concept that was performed in 2005 and 2006 by General Atomics, Oak Ridge National Laboratory, and Argonne National Laboratory, through the Advanced Fuel Cycle Initiative.

DB-TRISO Fuel Microanalysis. Starting from data

generated by the core design studies, the project will analyze the behavior of DB-TRISO fuel at burnup levels exceeding 50% FIMA. This enables the development of highly detailed, full 3-D models of hexagonal block and pebble fuels for physics calculations that account for kernel-scale self-shielding effects on local depletion and temperature distribution in the kernel. The project will generate data of neutron fluence, power production, and fission product gas production for use in fuel performance and design tools to determine the feasibility of the specific deep burn fuel designs.

DB-VHTR Fuel Modeling, Fabrication, and Testing

Modeling. While DB-TRISO fuel can in principle be irradiated to very high burnups of 40-50% FIMA within previously-achieved fluence limits of TRISO fuel, DB-TRISO fuel is expected to greatly exceed historically-achieved burnups with large quantities of fuel. The chemical and physical changes to the fuel kernel during Deep-Burn will be extreme and necessitate a deeper understanding of the fuel behavior during irradiation. Kernel, coatings, and fission product transport behavior will be modeled using leading-edge computational methods to provide predictive capabilities for integration with high fidelity nuclear models of the gas-cooled reactor.

Fabrication. There is a long history of international TRISO-coated particle fuel fabrication up to and including production scale. Remote fabrication at each step (kernel, coating, compacting) of such fuels has been studied as part of other DOE fuel programs. Both MOX fuel and Curium targets have been made via sol-gel technology. A pilot-scale remote operating coater was designed and operated 'cold' as part of the U/Th gas reactor fuel program. Automated compacting technology was developed as part of Ft. St. Vrain development. These technologies have been demonstrated at the proof-of-principle stage and designs exist for larger-scale remote systems. Engineering-scale demonstrations are needed to assess technical and economic feasibility of remote fabrication of DB-VHTR fuel. Of specific concern is the ability to perform remote maintenance on the chemical vapor deposition (CVD) coaters and the need to produce high quality, low-defect fuel in the remote environment.

The project will initiate steps towards the manufacture of testable amounts of DB-TRISO fuel and to establish the processes for large scale fabrication. Designs and plans for permitting and installation of coating and compacting facilities for TRU fuel will be developed. The project will establish surrogate coating operations and functional and operational requirements for the Pu and Am-241 equipment. The use of zirconium carbide (ZrC) will be considered, either on the kernel itself or on the buffer layer, or as a replacement for silicon carbide. The project will produce the first "sparse" kernels containing TRU materials (a few particles with the required amounts of americium among many kernels) and perform hot tests, though not in the first year.

Testing. Small quantities of TRISO fuel containing limited quantities of radioactive TRU such as Pu-239 and Np-237 have been successfully fabricated and tested in the past. Studies on the disposition of weapons-grade Plutonium indicate that extremely high burnup can be achieved by using

oxide fuels, oxygen getters and perhaps dilution with inert materials. LEU and Highly Enriched Uranium (HEU) TRISO fuel have been extensively tested both with respect to fluence and burnup.

The project will establish a similar database for the Deep-Burn fuel by irradiating DB-TRISO fuel manufactured in the course of this program in high-flux facilities in statistically significant sample sizes and perform post-irradiation testing to provide feedback to the fuel designers. The project will perform an analysis of existing irradiation facilities and develop a plan for irradiation and accident testing. This plan will include plans for the required irradiation campaigns, the testing at off-normal conditions, and the modifications to test equipment and facilities needed to test DB-TRISO fuel.

Recycle of Actinides from VHTR Fuel

In the early days of VHTR technology, a crush-burn-leach process was proposed to reprocess VHTR fuel. This process produced large quantities of carbon dioxide that needed to be trapped. A new head-end process consistent with the UREX+ separation technologies has been identified and demonstrated at the proof-of-principle level for TRISO fuel in the past few years. The process flow consists of separation of the compacts from the graphite block, disposal of the graphite block as low-level waste, grinding and jet-milling of the compact components (matrix, coatings, fuel kernels) into a fine powder to support chemical separation, and leaching, to dissolve the TRU for aqueous separation or a novel electrochemical process termed METROX for the pyroprocessing separation.

The project will develop a full flowsheet for TRISO recycling using both aqueous and non-aqueous reprocessing, particularly as it pertains to spent DB-TRISO fuel. The process of crushing the ceramic coatings and exposing the spent-fuel kernels to dissolving agents will be brought up to today's standards of low secondary waste streams and process losses. The project will study the crush-leach flowsheet to minimize waste, establish and test a laboratory filtering system, and study the suitability of the fuel solution for liquid separation. Lab-scale tests of the equipment for separation of the solid coating and compact material from the fuel solutions will be performed.

The Project will investigate the METROX process, a promising head-end coupling for TRISO fuel into pyroprocessing for metallic reactor fuel. Early studies successfully demonstrated process feasibility with uranium-based fuel but additional study is required to qualify the process for TRU-bearing fuel. The Project will construct a complete mass balance flowsheet including identification of waste form types and quantities, and conduct experimental studies needed to validate process chemistry.

Waste Management and Repository Performance of VHTR Spent Fuel

TRISO fuel provides very strong barriers to the dispersal of the long-lived components of radioactive spent fuel. The TRISO SiC coating acts as a pressure vessel to contain the helium produced during storage from the alpha decay processes. Preliminary measurements have indicated that the corrosion rate of both pyrocarbon and silicon carbide coatings

in the repository are extremely slow in air, moist air and water, to the point that there is reasonable expectation for containment of radioactivity over millions of years. Also, the use of a DB-VHTR to recycle LWR spent fuel could reduce the heat load to the repository by a factor of 2-3, given the high destruction rate of TRU in the Deep-Burn irradiation. Additional testing is needed to characterize degradation of the fuel compact and fuel particle under repository conditions and long-term modeling of repository behavior.

The project will study the long-term behavior of spent TRISO in dry and wet environments and develop a strategy for the geologic storage of DB-TRISO. The project will develop a plan for measuring corrosion rates of pyrocarbon and silicon carbide coatings in repository atmospheres. Relevant tests representing coating corrosion in the repository will be defined and exploratory testing to refine protocols will be performed.

Integration of the VHTR in the Overall Nuclear Fuel Cycle

The most likely role of the VHTR is to operate on LEU fuels for nuclear power or process heat and hydrogen-production applications. In this mode, the VHTR offers some fuel cycle advantage compared to LWRs because the rate of TRU production per energy generated is reduced by a factor of 2-3; this behavior is attributed to improved thermal efficiency, together with higher fuel enrichment and burnup in the VHTR.

The current TRU destruction scenario adopted by GNEP/AFCI is termed the single-tier approach; it is the simplest demonstration of closing the fuel cycle. In this case, spent fuel from LWRs is sent directly to the Advanced Burner Reactor (ABR) for destruction. Our studies suggest that DB-VHTRs can have a synergistic relationship with the ABR when operated in dual-tier mode. This synergy allows relaxed operating parameters for the two reactor types and a smaller inventory of recycling TRU relative to the single-tier approach. It would also reduce the number of fast reactors by a factor of 3 as compared to the LWR two-tier scenario (i.e., thermal to fast reactor ratio of 9 to 1 rather than a 3 to 1 ratio).

The project will assess the future role for VHTRs in delivering energy products including consideration of its fuel cycle and TRU-management function. As gas-cooled reactors may play an important role in the sustainable expansion of nuclear energy and in the TRU-management for the whole nuclear energy system, the project will perform a dynamic analysis of the evolution from today's reactor park to future reactor parks. Such dynamic analysis will focus on the correct assessment of the mass-flows, waste inventory and arisings, separated-TRU inventories, the delay times in deployment, and the overall economic impacts.

The project will assess the role of Pu and Minor-Actinide burning in DB-VHTRs in the context of a fuel cycle that includes LWR and fast-reactor fuels. The role of DB-VHTRs in burning Pu both from the stockpile and LWR reprocessing will be examined. The project will formulate a range of scenarios in which the VHTR burner are operated in conjunction with today's reactors to achieve fuel cycles in which waste discharges for geological disposal are minimized.

As part of the dynamic analysis, the project will explore the economic viability of the whole fuel cycle infrastructure required to support the introduction of the VHTR. Based on the mass-flow analysis and the technical description of fuel cycle infrastructure, proliferation and physical security risks will be evaluated.