DEEP BURN TRANSMUTATION OF NUCLEAR WASTE

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ABSTRACT

Helium-cooled, graphite-moderated reactors with ceramic-coated fuel particles offer unique advantages for the destruction of transuranic materials discharged in Light Water Reactor spent fuel. This is accomplished by fission, and capture-followed-by-fission processes. Three major features make it practical: (1) ceramic-coated particles accommodate high levels of burnup in one pass, thus reducing the need for repeated reprocessing; (2) graphite moderation produces valuable opportunities for thermal and epithermal neutrons to interact with fissionable and non-fissionable materials respectively; and (3) ceramic-coated particle kernel sizes can be adjusted to control the rate of such interactions. In the transmutation scheme proposed here, virtually complete destruction of weapons-usable materials, and 95% destruction of all transuranic waste is achieved. Higher levels of destruction are possible by repeated reprocessing and recycling, but there is little incentive to do so since each reprocessing step generates new secondary waste. After transmutation, the impervious ceramic-coated fuel particles provide an ideal residual waste form.

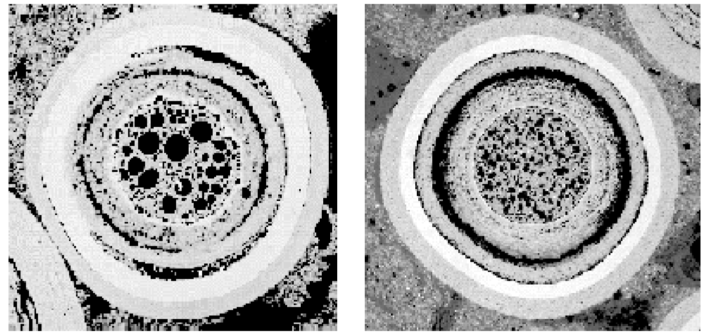
1. Introduction

To meet world power demands, the use and acceptance of nuclear power must continue to grow as a safe, reliable, and economical source of energy. A reason for concern in the U.S. is the fact that, at the rate waste is produced by the existing fleet of nuclear reactors, new repository capacity equal to the statutory capacity of the yet-to-open Yucca Mountain Geologic Waste Repository would be needed about every 20-30 years. Therefore, the ability to expand, or even maintain the nuclear power capacity in the U.S. may soon be limited, unless either additional disposal capacity is made available, or the volume, proliferation risk, and toxicity dose of the waste are significantly reduced. Based on this motivation, methods to reduce nuclear waste volume and toxicity have been proposed. However, while the potential advantages of such nuclear waste mitigation concepts are compelling, critics have argued that these methods would generate significant amounts of new waste, new opportunities for plutonium diversion would be created, deployment times would be long, and costs would be very high. In this paper, we propose a new option that addresses these objections. It is based on the use of modular helium-cooled reactor technologies.

2. The Deep Burn Concept

The concept described here, called Deep Burn transmutation, is based on the use of thermalized neutrons and high burnup fuel forms in modular helium reactor systems (MHRs). These reactors have annular graphite-moderated cores, and are designed to be passively safe at power levels up to 600 MW, such that there is no fuel failure or fission product release under any loss of coolant flow or pressure accident. They can also operate safely at very high temperatures resulting in electric power produced at close to 50% efficiency.

An essential feature of the deep burn transmutation concept is the use of ceramic-coated fuel particles (TRISO particles) that are strong and highly resistant to irradiation, thereby allowing very extensive destruction levels in one pass (figure 1). The ceramic coatings are also durable, and impervious to moisture for long times. Extrapolated corrosion test results by U.S. national laboratories indicate that the incremental waste exposure in the repository due to corrosion of the ceramic coatings is expected to be negligible for hundreds of thousands of years (figure 2). Thus, the discharge from the transmutation process provides a robust and attractive residual waste form. In addition, the MHR's fixed moderator (graphite), and neutronically transparent coolant (helium), provide inherent safety features and neutronics advantages for the destruction of nuclear waste that cannot be replicated in other design.



Pu Oxide
747,000 MWdays/tonne
>95% Pu-239, and
>65% all Pu transmuted

Th-Pu Oxide
183,000 MW-days/tonne
>95% Pu-239 Transmuted

Figure 1. Irradiated TRISO Particles

Another essential feature is that many more collisions are required to thermalize neutrons in an MHR than in water reactors. This produces valuable opportunities for epithermal neutrons to interact with non-fissionable materials. Specifically, since non-fissionable materials exhibit large neutron-capture resonances in the epithermal region (figure 3), they have a good chance to capture neutrons and become fissionable.

Yet another feature is that the size of the ceramic-coated particle kernels can be adjusted to create self-shielding so that fissionable materials, such as Pu-239, interact more readily with thermal neutrons, whereas non-fissionable minor actinides interact more readily with epithermal resonant neutrons. This produces enough neutrons to destroy fissionable materials by direct fission, and non-fissionable minor actinides by a capture-followed-by-fission process. Minor actinides behave as fertile fuel, helping maintain nuclear stability.

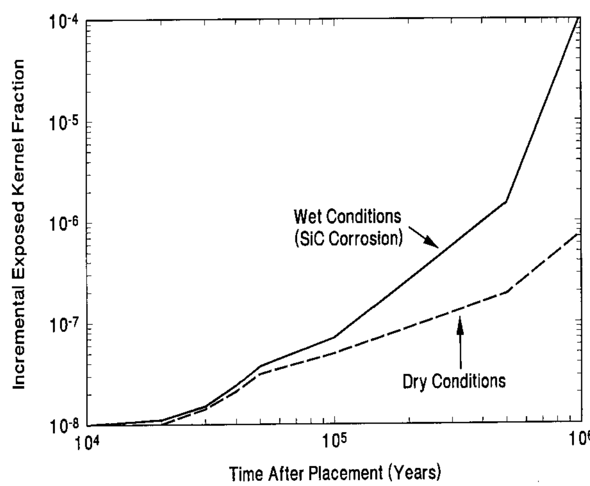


Figure 2. Durable Particle Integrity

3. The Process

The Deep Burn process starts with the separation of uranium and fission products from the LWR spent fuel. Plutonium and neptunium are then separated from the rest of the transuranic elements, and used to make TRISO Driver Fuel (DF), with kernels sized large to maximize the use of thermal neutrons. This maximizes fission and production of next-generation neutrons. The rest of the minor actinides are packaged in TRISO particles as Transmutation Fuel (TF), with kernels sized small to maximize the efficacy of epithermal resonant neutrons. This produces

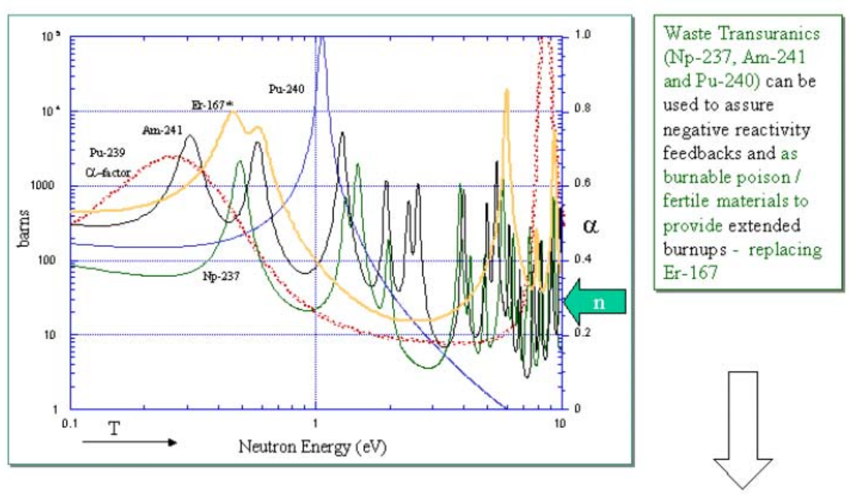


Figure 3: Capture cross-sections of selected minor actinides

transmutation into fissionable elements, which then it is followed by fission. We have selected block type fuel in an annular core configuration. The use of block fuel is important because the blocks stay exactly where they are placed until they are picked up. This makes the core configuration more predictable, allowing more control of flux distributions, better fuel utilization, and more predictable performance. It also reduces the opportunity for diversion, increasing proliferation resistance. One third of the Drivel Fuel (DF) is replaced with fresh DF at the end of each refueling cycle. It was originally proposed that each of these cycles would last a year. However, more recent calculations suggest that the refueling cycles may be as long as two years. The spent DF from the reactor is then reprocessed to remove fission products, mixed with the rest of the minor actinides left from the initial separation, fabricated into fresh Transmutation Fuel (TF), and reloaded in the reactor. In this reprocessing step, there is very little plutonium-239 handling since it is largely burned in the three preceding refueling cycles. At the end of each refueling cycle, one third of the TF (having been irradiated for three cycles) is replaced with fresh TF. The discharged TF may then be further irradiated (without reprocessing or refabrication) in an accelerator-driven subcritical MHR, or disposed of in a Geologic Repository.
This process has been evaluated using montecarlo and deterministic calculations combined with burnup calculations. The results show that virtually complete destruction of weapons-usable materials, and roughly 95% destruction of all transuranic waste is achieved (figure 4). Higher levels of destruction are possible by repeated reprocessing and recycling, but there is little incentive to do so as each reprocessing step generates new secondary waste.

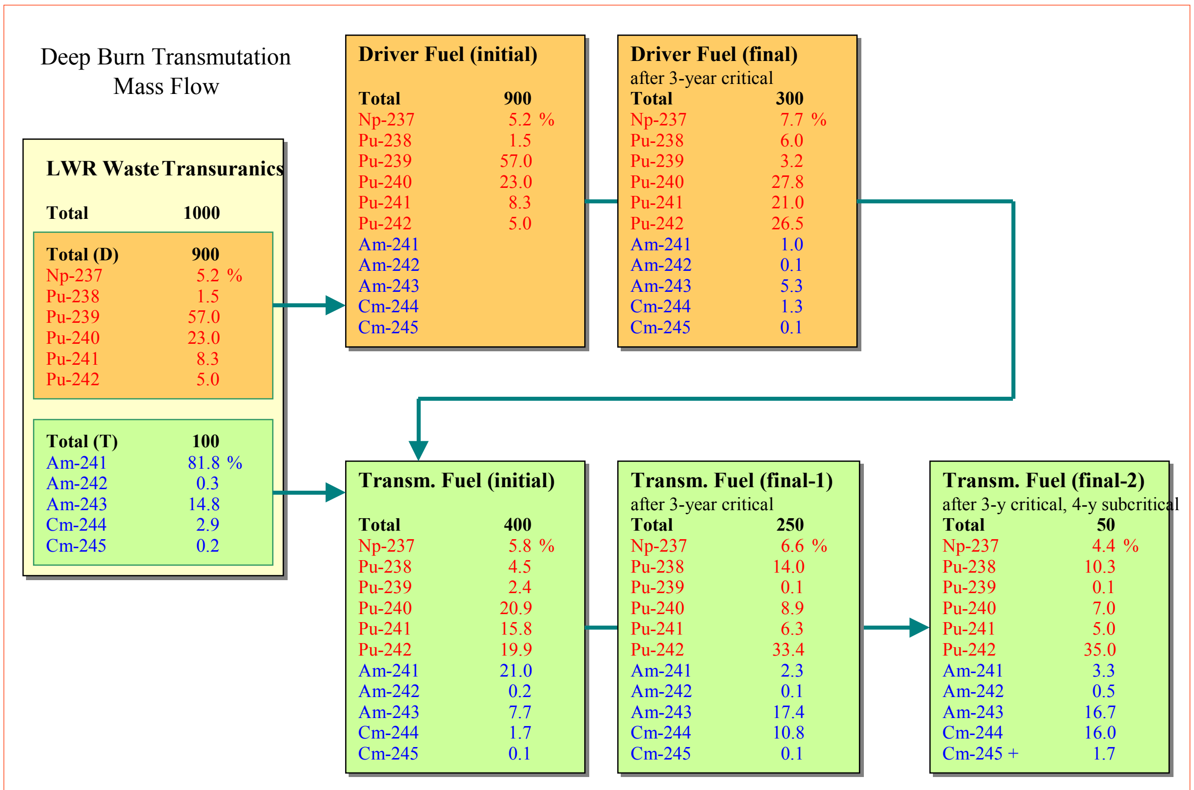


Figure 4 From LWR Waste to Final Waste after Deep Burn Transmutation: mass flow results from the Deep Burn Transmuter studies. The mass flow of the waste transuranics is followed through the Driver and Transmuation Fuel stages, the critical (MHR-DT) and subcritical (MHA-DT) irradiation periods.

The economics of Deep Burn have been analyzed using plant and fuel fabrication cost estimates from previous MHR reactor studies, particularly those related to plutonium burning. Separations and reprocessing costs were obtained from the DOE "ATW Roadmap" report of 1999. There are significant uncertainties about the cost of fabricating minor actinide fuel, and the cost of fabricating plutonium fuel is significantly higher than the cost of fabricating uranium fuel. However, despite this, it is interesting to note that the MHR Deep Burn feature leads to the production of much more electricity for a given amount of fabricated fuel, which leads to a fuel cost per kWh no much higher than the uranium fuel cost. More specifically, the total estimated cost is 3 ¢/kWh for plant amortization and Operations &

Maintenance, and 0.9 ¢/kWh for fuel and separations, for a total of 3.9 ¢/kWh. These costs include the cost of an accelerator for the subcritical part of the process, and suggest that the Deep Burn concept has the potential to be economically viable, particularly if it is carried through the completion of the critical phase only, or if the government is willing to pay for the separation step.

From the proliferation risk standpoint, Deep Burn requires no multiple or repeated reprocessing. There is the initial separation step where LWR fuel is processed to remove Uranium and fission products. However, fissionable actinide isotopes are not individually isolated in this step: Whole Plutonium is extracted simultaneously with whole Neptunium, and radioactive fission products could be left in the fuel to deter diversion at this stage. Later in the process, there is another intermediate reprocessing step, but it takes place at a point when there are essentially no Pu-239 or weapons grade elements. The whole process may be characterized as “separate and burn”. There is no continuous or repeated reprocessing of weapons-grade material. This significant anti-proliferation advantage is made possible by the Deep Burn feature of the MHR design.

From the toxicity standpoint, Deep Burn and the isolation provided by the corrosion-resistant ceramic-coated particles lowers the ingested toxicity risk in a repository below that of natural uranium from the start (figure 5).

4. The Fuel

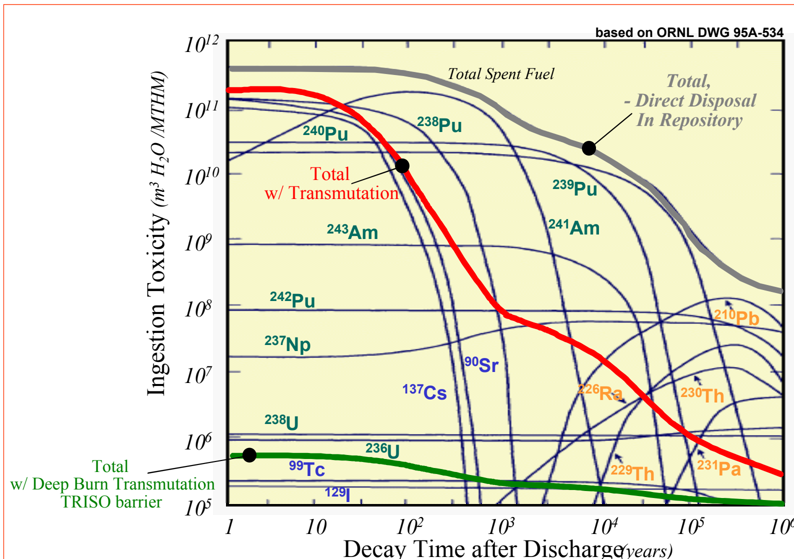


Figure 5. Toxicity in Repository after Deep Burn is Lower than that of Natural Uranium

TRISO Fuel particles are an ideal form of fuel for transmutation (and other missions) because they can accommodate deep levels of transmutation (Deep Burn), make useful use of epithermal neutrons, and can be adjusted in size to preferentially expose minor actinides to epithermal neutrons and fissionable materials to thermal neutrons. These features are in addition to their ability to sustain high temperatures, which allows production of electricity at high efficiencies and unmatched safety.

There are two kinds of sizes of particle kernels envisioned for transmutation: A large DF kernel and a smaller TF kernel.

To achieve Deep Burn design and performance requirements, the DF should be in a form to provide initial self shielding of some of the fissionable material. This self-shielding will reduce beginning of life excess reactivity and allow power to be maintained throughout the planned fuel lifetime, and requires a large, dense kernel.

The neutrons from the fissioning of DF are used to convert the initially largely non-fissile isotopes into fissionable isotopes and then to fission them. At the same time the non-fissile nuclides in the TF have resonance capture cross sections at neutron energies slightly higher than the neutron energy characteristic of the operating moderator temperature. Capture in the TF provides a negative temperature coefficient throughout the cycle. The capture of neutrons in the non-fissionable actinide nuclei therefore performs the useful function of converting the non-fissile nuclei into fissionable nuclei (this is in contrast to the use of burnable poisons, such as erbium, to provide negative temperature coefficient where the neutrons captured do not further the transmutation function). The new fissionable nuclei can then be fissioned, thus destroying long-lived transuranics. It is important that all of the material in the nuclei in the TF be available to the neutrons, i. e., no self-shielding. This is accomplished with a small diameter kernel, or a diluted or low-density kernel.

Developing fuel for high burnup and high fast neutron fluences containing undiluted fissile material (as required by the driver fuel) has been carried out with highly-enriched uranium as part of the Fort St. Vrain Nuclear Generating Station and subsequent commercial work in the U. S. The FTE-13 test in Peach Bottom demonstrated that TRISO-coated undiluted plutonium fuels could be irradiated successfully to high burnup. The behavior of high burnup fuels during core heat-up events has been studied at GA and KFA Jülich and the effect of burnup is included in the Goodin-Nabielek model applicable to uranium fuels. The ranges of burnup and fast neutron fluence covered by previous tests are compared to the requirements for the Driver and Transmutation Fuel in figure 6. The next step in the development of Driver and Transmutation fuels is to build on this technology to meet the projected fuel requirements. Oak Ridge National Laboratory (ORNL), with GA, will develop processes and equipment to fabricate DF and TF test samples for irradiation, accident simulation, and other testing. ORNL has extensive prior experience developing coated particle fuels, as part of the HTGR and Thorium Recycle programs, and relevant experience fabricating TRU targets containing neptunium, plutonium, americium, and curium as part of their on-going program for production of transuranic isotopes. The process development and test program will begin with uranium fuels contained in hoods, progress to glovebox containment for the development of the DF, and culminate with the development of the remote-handled TF in shielded cells. Irradiations are planned to be done in US thermal reactors, post-irradiation examinations in the ORNL hot cells, and accident simulations in the ORNL coated particle Core Conduction Cool down Facility. The uranium facility is currently under design and the DF and TF facilities are in the planning stages. Alternates to conventional TRISO coatings are included in the program. Tests of the performance of the TF in the repository are also an important part of the fuel development program.

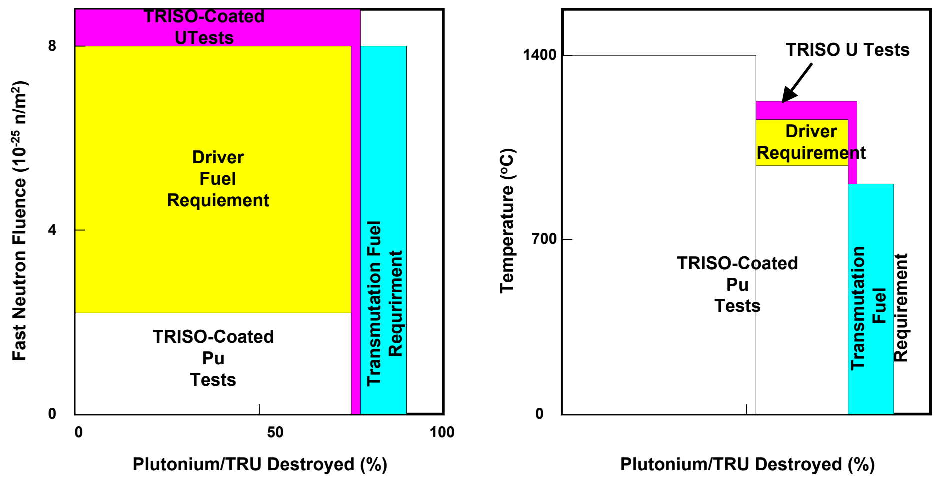


Figure 6.Comparison of DF and TF Requirements with the Irradiation Database

Development plans include meeting as-manufactured quality for glovebox and shielded-cell fabrication, high temperature processing of americium and curium, achieving in-service performance at high burnup and high fluence, and determining the limiting accident temperatures for DF and TF.

5. Conclusions

Deep Burn Transmutation allows the virtually complete destruction of weapons-usable materials, and roughly 95% destruction of all transuranic waste. Higher levels of destruction are possible by repeated reprocessing and recycling, but there is little incentive to do so as each additional reprocessing step will generate new secondary waste, which may eventually exceed the incremental destruction level achieved.

The Deep Burn concept would allow reductions in the volume of high level nuclear waste, and in toxicity and proliferation risk, safely, and in a potentially economic manner. Deep Burn MHR systems could be available for deployment in the near term to make a positive contribution to the solution to the waste problem. In addition to the deep burn transmutation capabilities, the MHR can, of course, burn uranium fuel or thorium in a high conversion ratio mode. This could lead to nuclear power scenarios that do not involve plutonium production and processing, and still maintain long-term sustainability.