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Disposition of Plutonium as Non-Fertile Fuel for Water Reactors

Kenneth Chidester*, Stacey L. Eaton, and Kevin B. Ramsey

Abstract

This is the final report of a three-year, Laboratory Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). The original intent of this project was to investigate the possible use of a new fuel form as a means of dispositioning the declared surplus inventory of weapons-grade plutonium. The focus soon changed, however, to managing the larger and rapidly growing inventories of plutonium arising in commercial spent nuclear fuel through implementation of a new fuel form in existing nuclear reactors. LANL embarked on a parallel path effort to study fuel performance using advanced physics codes, while also demonstrating the ability to fabricate a new fuel form using standard processes in LANL's Plutonium Facility. An evolutionary fuel form was also examined which could provide enhanced performance over standard fuel forms, but which could be implemented in a much shorter time frame than a completely new fuel form. Recent efforts have focused on implementation of results into global energy models and development of follow-on funding to continue this research.

Background and Research Objectives

Current conditions have created an increasing worldwide inventory of unseparated plutonium, which is a growing concern with regard to nuclear weapons proliferation. Although much attention has been given to the disposition of excess weapons plutonium, which is the more immediate concern, eventual attention will have to be focused on the much larger quantities of plutonium found in spent nuclear fuel. Research at Los Alamos National Laboratory (LANL) was focused on developing a fuel form that enables a near-term ability to manage these unseparated plutonium inventories. This new fuel form, referred to as Evolutionary Mixed Oxide (EMOX), is a slight perturbation on standard Mixed Oxide (MOX) fuel.

EMOX is created by the addition of a nonfertile component (currently calcia-stabilized zirconia) to a uranium-plutonium oxide matrix. By controlling the nonfertile content, the amount of plutonium remaining in the spent fuel can be controlled. EMOX is unique in that it can evolve, through the addition of greater amounts of nonfertile material, from a standard MOX composition to EMOX compositions with increased plutonium destruction rates. Eventually, a pure nonfertile fuel (NF) form containing no uranium can

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be used for nearly complete plutonium destruction. The various compositions can be tailored to meet specific fuel-cycle goals, whether it be the stabilization or reduction of unseparated plutonium inventories.

Most of the EMOX fuel development activities to date at LANL have focused on two main areas. The first area involves the analysis of the concept from a reactor viewpoint using physics codes in conjunction with certain self-imposed constraints. One major objective of this program was to develop a fuel form that could be used in existing reactor designs. The fuel must also adhere to safety (in-reactor behavior) and economic (fuel lifetime) considerations.

The second area is the assessment of the feasibility of fabricating such a fuel. In the fuel development laboratory at the LANL Plutonium Facility, the objective of the research was to address the issues involved with the fabrication of the EMOX fuels, using both surrogate materials and actual plutonium. The objectives also included the examination of such properties as phase distribution, pore size, grain size, and degree of densification. The major constraint is to limit the fabrication process to existing processes, thereby enhancing its commercial acceptability. This effort was closely coupled to the analysis effort to jointly produce a feasible fuel form.

Recent efforts involved the examination of certain performance metrics. Such metrics include the effect on worldwide (or system-wide) plutonium inventories, economic impacts, and the use of the energy-potential that exists in the fuel. The results of all of these efforts have been used throughout the life of the project in the ongoing activity of follow-on project development.

Importance to LANL's Science and Technology Base and National R&D Needs

This work supported the basic Laboratory strategic direction and goals in the key areas of nuclear technologies and advanced materials and processing. Physics codes were used to confirm the feasibility of using a nonfertile component in nuclear reactor fuel to increase plutonium consumption and reduce overall plutonium discharged from a reactor core. A process capability was established and used to fabricate test quantities of plutonium-bearing nonfertile fuels. These capabilities are now available to support future DOE missions to develop and fabricate plutonium- and uranium-bearing nonfertile-type fuels.

This work also addressed technical issues concerning the long term disposition of excess plutonium and laid a foundation for work enabling reduction of future plutonium inventory growth in the worldwide commercial nuclear reactors. Currently, plutonium inventories around the world are growing at 50 to 70 tons per year, with projections of

growth near 200 tons per year unless something is done to curb it. One proposal is to introduce advanced nonfertile fuels and advanced reactor systems. The technology and capability developed under this work directly supports one of DOE/NE's proposals to begin work in FY 1999 on Advanced Proliferation Resistant Nuclear Technology in the areas of advanced proliferation resistant fuels and reactor systems.

Scientific Approach and Accomplishments

Fuel Performance

Extensive calculations have been performed to assess the feasibility of incorporating the EMOX fuel form into existing pressurized water reactor (PWR) systems. For these calculations, a PWR pin-cell model was developed for use in the HELIOS¹ code. HELIOS is a collision probability-based transport code, which supports general two-dimensional geometries and has the ability to perform burnup calculations. The cross-section library contains over 270 explicit materials and data for 115 fission products. A typical Westinghouse PWR² was used for the model development. As is done with conventional MOX in a UO₂ core, the fuel cycles examined consist of the addition of some fraction of EMOX (typically third-core, half-core, etc.) to a standard enriched UO₂ core. This EMOX core fraction comprises varying percentages of PuO₂, natural UO₂, and varying amounts of a nonfertile component of calcia-stabilized zirconia (the "nonfertile fraction").

The EMOX performance was assessed as a function of several variables, including the fraction of the core that is loaded with the new fuel, the initial plutonium loading in the EMOX component, and the nonfertile fraction used in each composition. Figure 1 shows a sample of the type of results achieved through these evaluations. Performance is measured in terms of percent change in the core-averaged plutonium inventory, a positive value indicating a plutonium-breeding system. Results show that greater plutonium destruction rates can be achieved with the addition of increasing amounts of nonfertile material. Greater destruction rates are also achieved as the EMOX core fraction increases, and the systems can be utilized as plutonium consumers at lower initial plutonium loadings. The pure nonfertile cases provide the best performance, with the EMOX cases providing a smooth transition between that and the standard MOX initial fuel.

A four-assembly, 2-D, lattice-physics, color-set model was developed allowing analysis of EMOX compositions and neutronic properties in more realistic flux environments. The four-assembly model also allows investigation of the impact of varying the core fraction of EMOX. A spectrum of nonfertile and uranium-nonfertile

Fig. 1. Change in plutonium inventory for various third-core EMOX compositions as a function of initial plutonium loading.

compositions were analyzed. One-fourth core loading results indicate two trends with increasing plutonium weight percent compositions in the fresh EMOX: a desirable increase in the net total plutonium destroyed per cycle is observed (Fig. 2), but unfortunately an increase in the residual plutonium discharged in the spent fuel is also observed (Fig. 3). An optimum fresh EMOX plutonium loading of 7 weight percent was found.

Fig. 2. Change in plutonium inventory for various quarter-core EMOX compositions as a function of initial plutonium loading.

Fig. 3. Residual plutonium inventory for various quarter-core EMOX compositions as a function of initial plutonium loading.

Fuel Fabrication Demonstration

A parallel effort was ongoing to assess the feasibility of fabricating EMOX and nonfertile fuel within the bounds of existing fabrication processes. This effort was designed to address the issues involved with the development of a complete fabrication process including the examination of such final sintered fuel pellet properties as phase distribution, pore size, grain size, and degree of densification.

The decision to use calcia-stabilized zirconia ($\text{ZrO}_2\text{-CaO}$) as the nonfertile component of the EMOX and nonfertile fuel was made based on the substantial amount of information published from previous experimental and developmental programs. Experience shows that irradiation of $\text{UO}_2\text{-ZrO}_2$ fuel will result in rapid densification or void collapse due to phase transformation.^{3,4,5} In the Shippingport Pressurized Water Reactor, side-by-side irradiation was performed on ternary ($\text{UO}_2\text{-ZrO}_2\text{-CaO}$), binary ($\text{UO}_2\text{-ZrO}_2$), and uranium oxide (UO_2) plate-type elements.⁶ The UO_2 and ternary fuels performed similarly, with no reported fuel failures. The binary fuel, however, exhibited rapid densification due to the induced phase instability. Solid ternary ($\text{UO}_2\text{-ZrO}_2\text{-CaO}$) fuel pellets were irradiated at the Power Burst Facility, a transient test reactor, and no fuel rod failures occurred with over 1000 hours of operational experience.⁶ Finally, the Light Water Breeder Reactor Fuel Development Program performed side-by-side irradiation of ternary ($\text{UO}_2\text{-ZrO}_2\text{-CaO}$), binary ($\text{UO}_2\text{-ZrO}_2$) and UO_2 duplex fuel pellets (annular pellets with thorium center for breeding).⁶ Again, the UO_2 and ternary fuels performed similarly, both being successfully irradiated to high burnup.

EMOX and nonfertile fuels containing weapons-grade plutonium (obtained from the hydride/oxidation process at LANL) were fabricated using the solid-state reaction method. Specifically, a nonfertile fuel (PuO_2 - 2.69 wt% - ZrO_2 - 87.19 wt% - CaO - 10.12 wt%) and EMOX fuel (UO_2 - 83.94 wt% - PuO_2 - 10.97 wt% - ZrO_2 - 4.63 wt% - CaO - 0.462 wt%) were fabricated using the method shown in Figure 4. The results of microstructural and x-ray diffraction (XRD) analysis indicate formation of a solid solution for both fuel forms. Specifically, XRD analysis of the nonfertile fuel indicates formation of a face-centered cubic structure with a lattice constant of 5.147 Angstroms. For the EMOX fuel, analytical results indicate formation of a face-centered cubic structure with a lattice constant of 5.440 Angstroms. Density for sintered pellets ranged from 90% to 97% of theoretical.

Systems-Level Evaluation

The EMOX analysis was used to feed a global energy model being developed as part of LANL's Nuclear Vision project. The EMOX data will also serve as a reference for future systems studies on the impact of nonfertile fuel incorporation into global nuclear fuel cycles. These fuel and cycle studies are central to devising strategies for future reduction of global plutonium inventories and the associated current and future nuclear danger.

Program Development

The EMOX systems analysis and reactor physics work served as the foundation for a proposal submitted through LANL's Nuclear Vision Project to DOE/NE for an 8-year, 44 million dollar, proliferation-resistant fuel and cycle development program scheduled to begin in FY 1999. Presentations at DOE received positive feedback and the proposal has made it through the first series in the down-select processes. A DOE/NE FY 1999 proposed budget included \$3M for proliferation resistant fuel and cycle development. As a result of this LDRD work, LANL is well positioned to be a significant player in this and any other future proliferation resistant fuel development programs.

Fig. 4. Flow diagram for fabrication of EMOX fuel pellet.

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