NUCLEAR ENERGY RESEARCH INITIATIVE

Enhanced Thermal Conductivity Oxide Fuels

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The objective of the proposed research is to produce a novel oxide fuel form with superior thermal conductivity. The research is proposed under NERI's Materials Science area of emphasis and enhances the long-term viability and safety of nuclear energy systems and the stability of the spent fuel as a waste form. The resulting fuel will be applicable to existing light-water reactors, especially with high burn-up, high performance fuels. It is also expected that such fuel will provide superior performance in advanced reactors that would otherwise be fueled with low-conductivity oxide fuels.

Although UO_2 fuel has many desirable chemical characteristics and has served satisfactorily in light water reactors for many years, its low thermal conductivity imposes significant limitations on reactor operations in present and especially in high performance, high burn-up future reactors. An increase in the thermal conductivity would relax several of these limitations and provide the following significant benefits:

(1) Operational safety would be enhanced because fuel performance during a LOCA (loss-of-coolant accident) would be improved. The amount of heat stored in the fuel would be reduced, so peak cladding temperatures during dryout would be reduced.

(2) A reduction in fuel temperatures and stored heat would support reactor power up-rates and improve power production economics.

(3) The production of high-level radioactive waste would be reduced because fuel burn-up could be increased since lower fuel temperatures would result in less fission gas release, and smaller amounts of fission gas could eliminate limitations on burn-up that are tied to internal pressure of the fuel rod.

(4) Reduced temperatures and temperature gradients in the fuel pellets would reduce the stresses imposed on the cladding, reduce fuel cracking and relocation, and reduce life-limiting fuel swelling, so the effects of pellet-cladding mechanical interaction would be reduced.

(5) Proliferation resistance would be enhanced because the rate of production of ^{239}Pu decreases as burn-up increases.

The goal of enhancing the thermal conductivity of sintered oxide fuels will be achieved by a new process of penetrating a highly conductive solid second phase into the open or interconnected porosity of sintered fuel. This project would focus on UO_2 for the present program because the technology can be immediately applied to current LWR fuels.

The process first involves developing sintering schedules to produce a desired open pore structure (approximately 90 percent TD). The second critical step requires intrusion into the open porosity of the high conductivity phase. Although penetration of the porosity with a molten metal under pressure is conceivable, this requires very high processing temperatures for high-melting-point refractory metals or even zirconium alloys. A new, more attractive alternative method would be to infiltrate a liquid silicon carbide (SiC) polymer precursor and pyrolyze it at modest temperatures leading to an interconnected SiC phase with superior thermal conductivity for fully-dense polycrystalline material comparable to that of commercial purity silver (approximately 400 W/m·°K at room temperature). Although several infiltrations and pyrolyzing steps may be necessary, they could be readily performed in a simple batch process. Thus, it is proposed that the second approach be pursued.

Besides high thermal conductivity, necessary characteristics of a second phase include small neutron capture cross sections; high melting point; resistance to creep; compatible thermal expansion coefficients; and lack of chemical reactions with UO_2, fuel cladding, or water. SiC has been identified as an excellent material from most of these perspectives. For example, its thermal conductivity at 1,000°K is roughly ten times that of UO_2, and improves at higher temperatures. Moreover, the methodology for deposition of SiC carbide has been developed.

Standard UO_2 fuel has a very large temperature drop between the center and surface of the fuel pellets during irradiation. A drop of 1,000°C is not unusual during full power operation. The primary reason for the large temperature drop is the low thermal conductivity of UO_2. Substantial increases in thermal conductivity are possible with modest volume loadings of the conductive phase. Preliminary model calculations indicate that if the second phase has a high thermal conductivity compared to UO_2, a 10 percent volume loading of a continuous, randomly oriented second phase would increase the thermal conductivity of the fuel by about 50 to 100 percent, depending on temperature, and reduce the peak centerline fuel temperature in the hot channel bundle with a linear power of 29.8 kW/m by 800°C. It would be even greater for the peak pin.

The first technical challenge in producing an enhanced-conductivity fuel is first to model the heat-conducting performance of various microstructures. Three aspects of the microstructure are essential to good fuel thermal performance. First, the second phase must be finely dispersed. Widely spaced conductive paths would be less effective in conducting heat from the fuel and could produce localized hot spots on the cladding, resulting in nonuniform corrosion and hydrogen migration in the cladding. On the other hand, if the porosity is too fine and finely dispersed, penetration becomes difficult and thermal conductivity may be reduced by phonon scattering in the conductive phase. Second, the conductive phase must be continuous. Although there is a slight improvement of heat transfer even from discrete particles of a highly conductive material, much larger benefits are derived if the conductive material forms a continuous path. Third, the conductive network must penetrate the entire fuel pellet so that heat can be conducted efficiently from the center to the surface. Recent advances in SiC processing make it possible to produce SiC that has a very high thermal conductivity because of its high purity and good crystallinity. Doping with appropriate n-type elements can further increase the conductivity to above 480 W/m-°K. Silicon carbide also has a melting point of 2,700°C consistent with that of UO_2.