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Radiation Effects on High Thermal Conductivity Fuel Surrogates

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ABSTRACT:

A suite of sustainable fuel cycle options that enable improved safety, economics and security for current and future nuclear reactors is of central interest for the DOE Fuel Cycle Research and Development program. High thermal conductivity fuels offer several potential advantages over current oxide-based fuel forms, including the potential for achieving higher power ratings, high fuel burnup, and overall improved performance while maintaining good reliability and potentially achieving enhanced safety during normal and transient conditions. Significant enhancements in thermal conductivity (up to a factor of about two) compared to monolithic oxide fuel (~2-4 W/m-K) have been observed in scoping research studies that explored the use of ~10 vol% BeO or SiC high conductivity additives.

There are several challenges that must be successfully resolved regarding the scientific feasibility of these promising fuel systems. High conductivity ceramics typically suffer rapid degradation in thermal conductivity during neutron irradiation due to pronounced phonon scattering off radiation–induced defect clusters. This degradation occurs relatively rapidly at low irradiation temperatures (large changes occurring within ~0.1 displacements per atom, dpa), and may result in a lower temperature window below which composite fuel systems are no longer suitable or attractive due to reductions in the composite fuel conductivity to values approaching irradiated monolithic UO_2. A second major radiation degradation issue for many high conductivity composite fuel systems is that candidate materials with anisotropic crystal structures such as hexagonal close packed (HCP) BeO, AlN, and Al_2O_3 are susceptible to pronounced grain boundary cracking during neutron irradiation to fluences above ~1 to 5×10^{20} n/cm^2, E>0.1 MeV (~1 to 5 dpa), due to anisotropic swelling in the basal (c-axis) vs. prism (a-axis) direction. This cracking can cause further pronounced degradation in thermal conductivity and strength reductions, with the degradation typically becoming increasingly pronounced with increasing irradiation temperature.

This project aims to examine three main scientific feasibility issues associated with high conductivity composite fuels. The first research task is to examine the role of nanoscale dimensions on the suppression of grain boundary cracking in irradiated HCP ceramic materials, in particular to determine if there is a critical minimum size in HCP phases that effectively suppresses microcracking due to anisotropic swelling (and thereby suppresses dramatic cracking-induced reductions in thermal conductivity). The second research task is to evaluate whether a suitable operational temperature window might exist for high thermal conductivity composite phases in fuel systems (generally determined by thermal conductivity degradations due to defect

cluster-phonon scattering at low irradiation temperatures and by grain boundary microcracking or cavity-phonon scattering at high temperatures). The third research task will explore the irradiated phase stability of fine-scale ceramic additives embedded in a surrogate fuel, including effects due to radiation mixing of the interface region between the fuel and inert matrix phase.

For the purposes of this exploratory investigation of scientific feasibility of high thermal conductivity composite fuel systems, we will focus on SiC and Al_2O_3 additives. The surrogate composite fuel materials in this project will be based on CeO_2 containing ~10 volume % SiC or Al_2O_3, respectively. CeO_2 is commonly used as a model material for UO_2 in explorations of fundamental behavior due to both materials having the relatively rare fluorite crystal structure with Fm3m space group. Crystalline HCP ("alpha phase") Al_2O_3 nanofibers with an average diameter ~100 nm will be mixed with high purity nanoparticulate CeO_2 powder and hot pressed to produce high density composites containing ~10 volume percent Al_2O_3 fibers. The CeO_2-SiC composites will utilize ~10 vol.% high purity polycrystalline SiC platelets with typical dimensions of ~10 mm diameter x ~1 mm thick. The composite samples will be irradiated with 20 MeV Ni ions at 300 to 700^{o}C to doses between 1 and 20 dpa. A single scoping neutron irradiation experiment will be performed using a fission test reactor irradiation capsule at a temperature near 450^{o}C to a dose of ~5 dpa. Postirradiation characterization will involve transmission electron microscopy examination of the general microstructure (dislocation loops, etc.) and potential microcracking along the interface between the high conductivity ceramic platelets or platelets and the CeO_2 matrix, as well as potential grain boundary cracking within the ceramic platelets or whiskers due to radiation-induced differential swelling. The bulk neutron irradiated fuel surrogate samples will be examined following irradiation to measure key properties relative to the initial unirradiated values including overall swelling, thermal diffusivity, and compressive strength.

The proposed research will provide fundamental knowledge on the general radiation stability of surrogate composite fuel forms at LWR-relevant irradiation conditions and produce an irradiated composite fuel thermo-mechanical model that can be used to predict the suitability of numerous candidate inert matrix fuel composites. In particular, the role of reduced lateral dimensions on suppressing grain boundary cracking in anisotropic (HCP) inert matrix ceramics such as BeO, AlN and Al_2O_3 will be evaluated using the obtained experimental results and thermomechanical model; suppression of the grain boundary cracking that normally occurs in bulk forms of these anisotropic ceramics is crucial for establishing their feasibility for LWR fuel system applications since the cracking produces large degradation in thermal conductivity. The potential operating temperature windows for composite fuel systems will be evaluated, based on thermal conductivity reductions due to strong phonon-defect scattering at low temperatures and phonon-cavity scattering or grain boundary or interface cracking at high temperatures. The overall phase stability of fuel-inert matrix phases will also be examined, taking into account prolonged radiation mixing effects. The proposed work will thereby provide insight on important scientific feasibility issues for high thermal conductivity fuels based on BeO or SiC additives.