Radiolytic Dissolution Rate of Silicon Carbide

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ABSTRACT:

The development of accident tolerant fuels (ATF) is aimed at avoiding the situation that occurred at Fukushima in 2011. In that station blackout condition, rapid oxidation of the Zircaloy cladding resulted in a highly exothermic zirconium-steam reaction. The rapid oxidation resulted in severe clad embrittlement and the production of hydrogen, also at a high rate, which later combusted and caused damage to the secondary containment. To avoid such occurrences in light water reactors (LWRs), the ATF program was initiated to focus on the replacement of zirconium-based alloys with materials that exhibit slower steam oxidation kinetics. One candidate structure being investigated is a silicon carbide (SiC) fiber composite, which is also proposed for next-generation reactor cladding designs because of its strength, high temperature stability, and neutron-transparency.

However, silicon carbide is known to dissolve, albeit slowly, into water under typical LWR operating conditions. To our knowledge there have been only two tests of dissolution in-reactor, and these suggest that the radiation field will enhance the SiC dissolution rate. It is an open question how this enhanced dissolution scales with the strength of the radiation field.

Clearly if SiC materials are to be qualified and used as cladding in existing LWRs, the dissolution behavior must be quantified and its mechanism must be understood as a function of temperature, radiation field, and water conditions of pH and hydrogen/oxygen concentration. We propose to quickly develop a matrix of dissolution rates for high purity SiC material using intense electron beam irradiation to provide the radiation field, and measure the products of dissolution (silicic acid and CO_2 (or CO)) in the water downstream of the irradiation zone. The method allows multiple measurements using the same SiC sample at various temperatures and with variable water conditions. A second method will be pursued in parallel, should the SiC dissolution rate prove too slow to be readily measured via the products. In this method, thin (submicron) films of SiC will be deposited on zircaloy substrates, and irradiated at high temperature with very high radiation field for most of a day. Subsequently the thickness of the exposed samples will be compared to the original thickness to determine how much material has been lost. The latter method is time consuming and more costly, but is certain to work, and produces more information about the mechanism of hydrolysis.

By the end of this project, we expect to have determined the rate of SiC dissolution, and gathered sufficient insight about its mechanism in LWRs, so that engineering and use of SiC/SiC composite materials for accident tolerant fuel cladding can proceed with confidence.