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CHARACTERISTICS OF HYDRIDE PRECIPITATION AND REORIENTATION IN SPENT-FUEL CLADDING*

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DISCLAIMER

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SUMMARY

The morphology, number density, orientation, distribution, and crystallographic aspects of Zr hydrides in Zircaloy fuel cladding play important roles in fuel performance during all phases before and after discharge from the reactor, i.e., during normal operation, transient and accident situations in the reactor, temporary storage in a dry cask, and permanent storage in a waste repository. In the past, partly because of experimental difficulties, hydriding behavior in irradiated fuel cladding has been investigated mostly by optical microscopy (OM). In the present study, fundamental metallurgical and crystallographic characteristics of hydride precipitation and reorientation were investigated on the microscopic level by combined techniques of OM and transmission electron and scanning electron microscopy (TEM and SEM) of spent-fuel claddings discharged from several boiling and pressurized water reactors (BWRs and PWRs). Defueled sections of standard and Zr-lined Zircaloy-2 fuel claddings, irradiated to fluences of $\approx 3.3 \times 10^{21}$ n cm^{-2} and $\approx 9.2 \times 10^{21}$ n cm^{-2} (E > 1 MeV), respectively, were obtained from spent fuel rods discharged from two BWRs. Sections of standard and low-tin Zircaloy-4 claddings, irradiated to fluences of $\approx 4.4 \times 10^{21}$ n cm^{-2}, $\approx 5.9 \times 10^{21}$ n cm^{-2}, and $\approx 9.6 \times 10^{21}$ n cm^{-2} (E > 1 MeV) in three PWRs, were also obtained. Microstructural characteristics of hydrides were analyzed in as-irradiated condition and after gas-pressurization-burst or expanding-mandrel tests at 292-325°C in Ar for some of the spent-fuel claddings. Analyses were also conducted of hydride habit plane, morphology, and reorientation characteristics on unirradiated Zircaloy-4 cladding that contained dense radial hydrides. Reoriented hydrides in the slowly cooled unirradiated cladding were produced by expanding-mandrel loading.

Results of TEM characterization of the BWR and PWR spent fuel cladding revealed a high density of "microscopic" delta hydrides that were too

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small to be resolved by OM or SEM. The microscopic hydrides were 20-100 times smaller than the normal "macroscopic" hydrides that are readily resolved by OM. The distribution of microscopic hydrides, short dislocations, and dislocation loops produced during irradiation in reactor was analyzed by stereo TEM, and the three-dimensional distribution of these features was determined from stereopairs of bright- and dark-field images. The results showed that the microscopic hydrides were bulk hydrides and that they were nucleated preferentially in association with <c>-type dislocation loops and short dislocations. The number density of microscopic hydrides increased significantly with increased fluence and was at least a few orders of magnitude greater than that of macroscopic hydrides in the PWR cladding that was irradiated to a fluence of 5.9×10^{21} n cm^{-2} ($E > 1$ MeV).

The habit plane of precipitation of the microscopic hydrides was the basal plane in the as-irradiated BWR and PWR cladding. Because of the characteristic texture of cladding (i.e., strong propensity for basal pole alignment ≈30° away from the radial direction), most of the microscopic hydrides were aligned ≈30° off the tangential direction of the cladding. The microscopic hydrides seem to aggregate or form a stringlike cluster to minimize the surface and strain energies associated with the metal-hydride boundaries, eventually forming a long circumferential hydride that is readily resolved by OM. It has been generally agreed that the habit plane of hydride precipitation in unalloyed α-phase Zr is $\{100\}_{Zr}$, i.e., the prism plane. However, the habit plane of macroscopic hydrides in unirradiated unstressed Zircaloy-2 and -4 has been reported to be $\{107\}_{Zr}$, a plane very different from the prism plane but rather close to the basal plane. In agreement with this report, macroscopic hydrides observed in the "as-spent" BWR and PWR claddings exhibited a habit plane very close to $\{107\}_{Zr}$, a plane 14.7° away from the basal plane. Based on these observations, we propose a model that explains the relationships among the habit planes of the microscopic and macroscopic hydrides, cladding texture, and the orientation of the macroscopic hydrides. Radial macroscopic hydrides were observed in significant numbers on the prism plane in the burst BWR and PWR cladding specimens and in the unirradiated mandrel-loaded and slow-cooled specimens. This observation indicates that in contrast to the $\{107\}_{Zr}$ habit plane of the macroscopic circumferential hydrides, the habit plane of reoriented radial hydrides is the prism plane.

No broken or partially cracked hydrides were observed regardless of its morphology (macroscopic or microscopic) or orientation (radial or circumferential), under the slow-strain-rate mandrel-loading condition. Significantly bent macroscopic hydrides were observed in the PWR cladding that was stressed to failure at ≈325°C by mandrel expansion. These observations indicate that hydrides were ductile at the mechanical test temperatures (i.e., 290-325°C). Dense dislocations were frequently observed at the metal-hydride boundaries, indicating that localized dislocation motion occurring at the metal-hydride boundary (probably promoted by the high concentration of hydrogen in solution in the metal and the mixed tensile and shear stress present at the metal-hydride boundary) is the primary deformation mechanism that leads to failure.

Possible implications of the results from the present investigation are discussed to provide a better understanding of several fuel performance issues. These issue are long axial splitting in high-burnup cladding under reactivity-initiated-accident situations, susceptibility of high-burnup cladding to hydride reorientation and potential failure during dry-storage and repository conditions, and the peculiar hydride dissolution and precipitation behavior in spent-fuel cladding (i.e., significant difference in the solubility limits of hydrogen measured during heatup and cooldown of the cladding).