LWR Pellet-Cladding Interactions: Materials Solutions to SCC

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Zirconium alloys are commonly used as fuel-cladding tubes in water reactors because of their inherent resistance to a variety of environmental conditions. One of the major fuel-reliability issues of the 1970s and early 1980s was pellet cladding interaction (PCI). The mechanism of PCI is one of stress corrosion cracking (SCC) by a combination of aggressive fission products and cladding stress from pellet expansion. The severity of the problem, in particular in boiling water reactors, led to the development of barrier cladding by co-extrusion of Zircaloy-2 with an inner iodide zirconium that essentially eliminated the PCI-related failures. However, the substantially lower corrosion resistance of the zirconium layer led to clad breach and failures by other mechanisms. The difference in corrosion resistance could lead to some dramatic differences in post-failure fuel operations. This article briefly summarizes how PCI-SCC factors led to the development of PCI-resistant fuel cladding and concludes with a note on future research needs.

INTRODUCTION

Zirconium alloys are used extensively in both light- and heavy-water reactors for such applications as fuel rods, intermediate grids, and calandria tubes. The two commonly used alloys, Zircaloy-2 and Zircaloy-4 (hereafter referred to as Zry-2 and Zry-4, respectively), are tin- and iron-based with slight alloying differences (Zry-2 contains 0.05% nickel while Zry-4 does not contain any nickel but has a slightly larger amount of iron). They are used as thin-walled tubes to clad fuel (UO_2) in both boiling-water reactors (BWRs) and pressurized-water reactors (PWRs). Many fuel vendors are currently offering (or at least considering) binary and ternary alloys of zirconium and niobium, particularly for PWRs where the improved strength, reduced irradiation growth, and reduced hydrogen pickup are more important. Current generation alloys such as Zirlo[1] and M5,[2] which are niobium-modified alloys, have the greatest experience base to date (aside from similar Russian alloys like E-110 and E-635). These materials have been shown to resist in-reactor corrosion and oxidation especially at long exposures, thereby making them valuable for extended burnup and longer times. Comstock et al.[3] reported the effect of processing variables on corrosion behavior of Zirlo®, along with the influence of alloy chemistry. Recent work by Murty et al.[4] summarizes texture, mechanical, and creep anisotropy, as well as transitions in creep mechanisms[5] in niobium-modified Zircaloys.

Zirconium alloys can be used in either a fully recrystallized annealed (RXA) or a cold-worked stress-relieved (CWSR) condition, depending on the desired properties related to irradiation growth, strength, and corrosion resistance. The processing route and heat treatment also depend on the composition of the alloy and the type of operating environment. Overall, zirconium-based fuel components have operated with a high degree of reliability under a range of conditions. To put this into perspective, typical failure rates today are less than ten per million rods in operation and the industry goal is on zero failures.

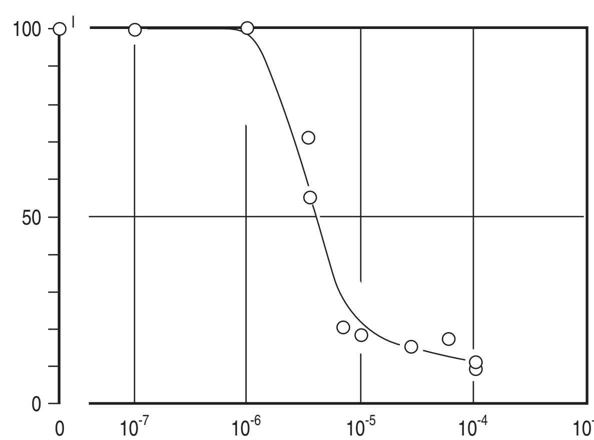

Figure 1. Effect of iodine (x-axis: g/cm^2) on relative uniform strain (y-axis: %).[10]

Fuel failures have been postulated or observed to occur under a number of conditions. Failure mechanisms that result in cladding perforation include[6,7] manufacturing defects, corrosion, hydriding, fretting, and pellet-cladding interaction (PCI). Through the use of robust engineering and improved manufacturing techniques, manufacturing defects have been practically eliminated by a number of fuel vendors. Likewise, failures by either corrosion or massive hydrogen uptake have also been nearly eliminated except in a few cases generally related to atypical operational conditions (e.g., coolant chemistry transients). This paper will deal with PCI failures, but it is interesting to note that, with the elimination of these failures, the most commonly observed failure mode has been debris fretting.

The PCI mechanism is now known to be in-reactor stress corrosion cracking (SCC) influenced by fission products such as iodine and cadmium. Failures due to iodine SCC (ISCC) were identified as early as 1965 during an exploratory test of performance limits of LWR fuel where the fuel rods were operated at very high power (~55 kW/ft). The chronological events based on the experience of the General Electric Company were summarized by Armijo et al.[8] Fission-product iodine was found to be the main culprit after zirconium iodide was observed near the pinholes.[9] The PCI-related fuel failures were noted in a commercial reactor in around 1969, and subsequent laboratory tests revealed the underlying mechanism to be ISCC. PCI failures are not unique to BWRs, and all water reactor types revealed such incidents:[7] PWRs—Maine Yankee, Obrigheim, Point Beach; BWRs—Dresden, Gundremmingen, Oskarshamn, Peach Bottom, Quad Cities; heavy water reactors—Douglas Point, Pickering. However, the relatively severe ramp conditions in BWRs have attracted more attention to this problem.

The search for PCI mitigation led, in the mid 1970s, to the development of barrier fuel with a thin, pure zirconium layer on the inside surface of the tubing that essentially eliminated PCI-related failures. Various research and simulation programs were followed by unrestricted operation in commercial reactors in the mid-1980s,[8] marking a major accomplishment in improved reliability and operation flexibility.

While the fuel reliability continued to improve following the introduction of barrier fuel, there was a period in the

1980s and 1990s when even a single failure could disproportionately affect the baseline activity and offgas of the plant. It was observed that, following a primary failure, the integrity of some fuel rods began to degrade, allowing further access of the coolant to the UO_2 fuel and accelerating the rate of fuel release to the coolant. This behavior, called secondary degradation, was later shown to relate to the corrosion resistance of the zirconium barrier. This led to a new set of studies aimed at improving the corrosion resistance of the barrier without sacrificing the PCI resistance.

This article describes the PCI mechanism and related aspects while summarizing the salient features of some of these advanced alloys.

PCI-SCC MECHANISM

When fission-product iodine was identified as the culprit for the PCI failures, extensive research began into characterizing the influence of iodine on the fracture behavior of Zircaloys with varied compositions and thermo-mechanical treatments. Enormous amounts of data were collected on the influence of iodine concentration, test temperature, and applied stress, and extensive research was conducted on various test techniques. (The reader is referred to the proceedings of various symposia, in particular by ASTM on "Zirconium in the Nuclear Industry" from the late 1970s through the 1990s. Such information may also be found in many International Atomic Energy Agency and American Nuclear Society proceedings.) The range of parameters considered is quite extensive and includes texture, cold work, radiation, stress state, and stress level, in addition to those listed earlier. Tests ranged from simple tensile tests under an iodine environment to internally pressurized, closed-end tubing with iodine to full simulations with internally expanding ceramic segments to mimic fuel loading.

Figure 1 depicts the effect of iodine concentration on uniform hoop strain at 673 K at a hoop stress (σ_θ) of 250 MPa.[10] With the express intention of characterizing the competing effects of creep and time for crack initiation and propagation, Peehs et al.[10] reported the effects of superimposed stress level as well as the applied strain rate on ductility in the presence of iodine. A threshold level of iodine concentration (partial pressure) is reported to be about 2×10^{-6} g/cm^2, around which a precipitous drop in ductility is noted. (There has been extensive debate on the quantification of iodine concentration in analyzing these types of results.) Once the role of iodine in laboratory SCC tests was clearly identified, the next step was to compare and correlate the crack morphologies observed in laboratory tests on unirradiated and irradiated Zircaloys with those observed in PCI-related in-reactor fuel failures. When both unirradiated and irradiated (7×10^{24} n/m^2, E>MeV) tubing samples that fractured following exposure to iodine vapor at 573 K were compared with a clading section that failed after an increase of power in a test reactor and with an element that failed after an increase of power in a commercial reactor, the results showed that PCI failures are related to ISCC. Stress-versus-time-to-failure plots were generated to identify the threshold stress in an iodine atmosphere. Radiation exposure is clearly more important than other factors, such as the thermal and mechanical processing variables (Figure 2).[11] Processing effects are difficult to compare due to the complex combination of surface condition, texture, and/or residual stresses; the effects of texture and stress state will be described in the sidebar.

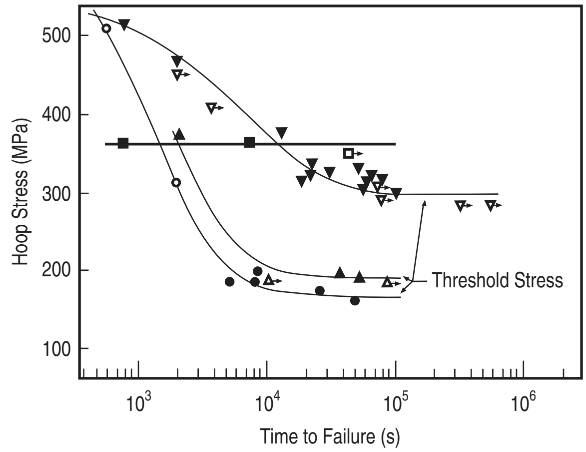

Figure 2. Effect of stress on time-to-failure.[11]

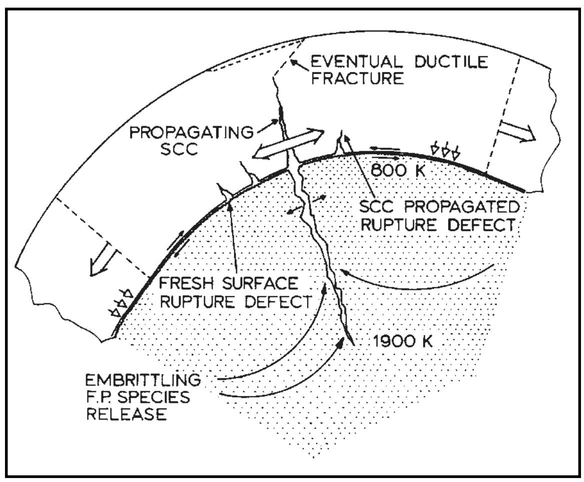

Figure 3. PCI-SCC model (after Roberts[12]).

These observations support the PCI model, illustrated in Figure 3, that depicts salient features of a cladding breach under PCI conditions.[12] Upon a power ramp, the ceramic fuel expands and cracks due to steep thermal gradient from the center line of the fuel pellet to the outer surface. The cladding is placed under hoop tension while the cracks allow additional access of fission products to the inside surface of the cladding.[13] The presence of an aggressive environment of fission products, such as iodine and cadmium, along with localized stresses and strains at the positions where PCMI exists, leads to SCC. The interaction is further assisted by the biaxial-stress state that exists from the axial restraint of the fuel column on the cladding.

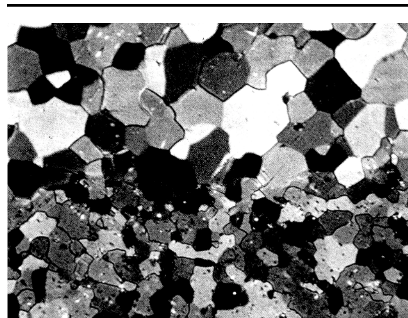
Figure 4. An example of the excellent bonding of Zr-liner with Zircaloy-2.[18]

PCI-RESISTANT CLADDING

A potential solution to minimize PCI-related failures was to decrease the power-ramp rate to allow sufficient time for hoop stresses to relax. However, such power-ramp restrictions result in loss of power availability and operational flexibility and, thus, other remedial solutions were sought. A number of solutions were considered. One such idea was to coat the inside surface of the cladding with graphite. This was later shown to be successful in the CANDU reactors, but was not believed to be practical in BWRs. One obstacle was the practicality of applying the coating on the inside of a tube that was about 4.5 m long (versus about 0.5 m for the CANDU). More importantly, though, the environment in a BWR, with greater linear heat rates and higher burnups, required a more robust design. The concept that received the most attention as a cladding solution was a metallurgical barrier.

Both soft copper and unalloyed zirconium barriers on the inside surface of the tubing were considered. These were thought to provide a material that was inherently more resistance to SCC than

THE EFFECTS OF TEXTURE ON SCC

As noted previously, the investigations leading to the characterization of the behavior of Zircaloys under simulated environmental and mechanical loading have identified several parameters which include material condition, irradiation level, inside surface condition of the cladding, fission product concentration, and crystallographic texture[14] of the tubing material. The fracture of Zircaloy under fission product SCC conditions occurs by cleavage on the basal or near-basal planes of the hexagonal crystal. In particular, Peehs et al.[10] have shown that Zircaloys, with a preponderance of basal poles in a near-perpendicular orientation to an applied load, are relatively more resistant to SCC than the material with an applied load parallel to the basal poles, meaning that tubing with radial basal pole texture resists SCC.

Tubing cut out from a sheet with ideal normal texture was exposed to iodine during an internal pressure test. After a certain time under pressure, the tubing sample was examined for cracks in the transverse plane, and Figure A shows the profile of crack density along the tube circumference. The majority of the cracks were seen at positions where basal poles are mainly along the hoop (transverse) direction; maximum crack density occurred at basal pole angles between 50° and 70°. Thus predominant radial texture resists ISCC as has been observed in tubing fabricated using high precision tube reduction (HPTR) process versus the normal Pilger milling. This is also evident in the study by Schuster and Lemaignan,[15] who investigated the ductility loss in iodine atmosphere of Zircaloy-4 as a function of the basal peak pole angle (ϕ). A precipitous loss in ductility occurred at $\phi \geq 35°$ and the thermal treatment (CWSR vs RXA) apparently did not have much impact on the data.

Adams et al.[16] showed the superiority of radial basal texture to cleavage using a quantitative texture analysis in conjunction with appropriate cleavage stress criterion based on the existence of a threshold stress for SCC above which environmentally induced embrittlement occurs. Both crack initiation and propagation were considered to occur when the effective stress normal to the basal plane is greater than a threshold value. A simplified analysis based on the distribution of basal poles (<0002>) revealed stress variation of SCC-susceptibility akin to the commonly observed stress-time curves in SCC tests. The influence of texture was investigated by considering all possible basal pole textures from circumferential to radial distributions.[17] Figure B depicts the model predictions in terms of inverse SCC-susceptibility (equivalent to exposure time) versus the applied stress level (represented here as β) for equibiaxial stress ($\sigma_\theta = \sigma_z$ or α = 1) state for three different textures—purely radial, purely transverse (or circumferential), and the typical bimodal texture observed following a CWSR anneal. These results are very similar to the σ-t_r plots typically observed in SCC tests.

The model predictions clearly indicated the superiority of the radial basal pole orientation to resist cracking to stress corrosion as in the experimental results of Peehs et al.[10] In addition, Murty et al.[17] were able to predict the stress-state (σ_z/σ_θ) dependence of the threshold stress for CWSR and recrystallized Zircaloy claddings.

Figure C depicts the stress-state dependence of the threshold stress normalized to uniaxial-hoop (σ_z = 0) loading for CWSR Zircaloy-4, and the experimental results of Cubicciotti et al.[11] are compared with the predictions based both on the crack initiation and propagation models. We note a good correlation and these results illustrate the complex dependence of SCC susceptibility on stress-state, crack initiation, and propagation characteristics.

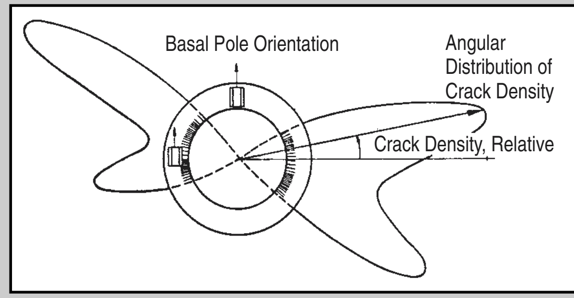

Figure A. Influence of basal pole orientation on crack (iodine SCC) density.[10]

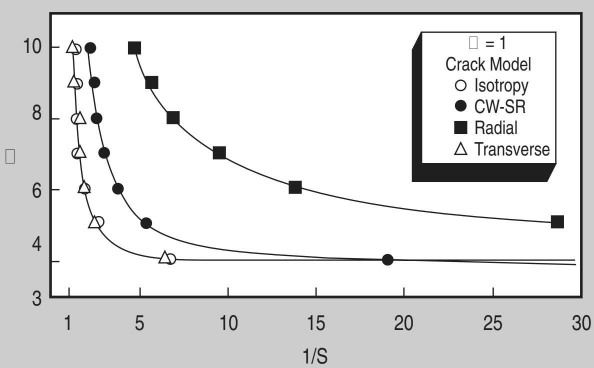

Figure B. Texture model predictions of stress-level (β) versus rupture time (inverse of SCC susceptibility parameter, 1/S).[16]

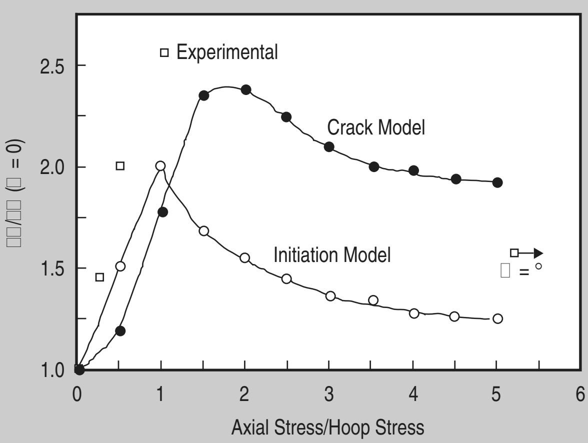

Figure C. Effect of stress ratio ($\sigma/_z/\sigma_\theta$) on threshold-SCC susceptibility. Model predictions compared with experimental results on CWSR Zircaloy-4.[17]

Zry-2 and to reduce the local stresses in Zry-2 near pellet cracks (Figure 3). The advantage of considering the composite cladding approach is PCI resistance without compromising the properties of both the fuel and cladding. Both copper and zirconium barriers were initially considered feasible.[13] The copper barrier was produced by electroplating, and the zirconium barrier by co-extruding concentric cylinders of Zry-2 and pure zirconium into a tube shell geometry.

Both expanding mandrel and closed-end internal pressurization tests (with iodine and cadmium) were performed in aggressive environments on liner cladding. In unirradiated testing, both iodine and cadmium exhibited superior performance when compared to nonbarrier Zry-2 cladding. However, following irradiation, the same tests demonstrated the advantages of zirconium over copper.[14] The decrease in resistance of the irradiated copper barrier was attributed to the brittle intermetallic layer that formed at the barrier-Zry-2 interface. Of additional concern was how to uniformly coat the 5 m long rod on the internal surface with copper as well as the creation of a bi-metallic galvanic cell.

The excellent PCI resistance of zirconium barrier fuel was attributed, in part, to zirconium's superior resistance to iodine compared to the resistance of Zircaloy. In addition, zirconium can withstand far higher strains prior to the embrittlement by the aggressive atmosphere. Because of the co-extrusion of the composite tube reduced extrusions (TREX), an inherent metallurgical bond could be established between the liner and the bulk Zircaloy. Figure 4 demonstrates that the various cold-work and intermediate anneal steps during the tube manufacture essentially eliminated any physical boundary between the liner and the bulk clad.[18] Because recrystallization and grain growth occur more rapidly in pure zirconium, a relatively large and easily discernable grain structure is developed compared to bulk Zircaloy. In addition, the higher alloying content of Zry-2 leads to a higher density of second-phase intermetallic particles (Figure 4). Since there is no oxide and no surface or grain boundary between zirconium and Zircaloy, the reliability of bond and adhesion are unquestionable. Production of zirconium-barrier fuel and irradiation demonstrations in operating commercial reactors commenced sometime in 1981. The results from these various reactors resulted in the elimination of operational limits by around 1985.[8]

ADVANCED BARRIERS

A major improvement in fuel reliability and operational flexibility was achieved with zirconium-lined barrier cladding, which mitigated the PCI failure mechanism in BWRs. However, failed rods have exhibited an increased tendency for relatively long cracks.[19–21] The mechanism was shown to be a multi-step process.[22] Beginning with an initial failure, often by debris fretting, steam entered the rod and began to react with the inside surface of the cladding and the fuel. As the corrosion reaction proceeded, the steam became continuously depleted of oxygen and enriched in hydrogen. At some distance from the primary defect, the gas on the inside of the rod became rich enough in hydrogen to rapidly absorb into the cladding. The hydrogen then formed a brittle second phase with zirconium (ZrH_x). Application of stress to this region (e.g., during a power change) could then lead to fracture and a new, secondary defect. The secondary defect could often propagate into a longer crack due to the stress concentration at the new defect and a new mechanism related to available hydrogen in the cladding. On occasion, the cracks in secondary defects reached 2–3 m in length. The mechanism of crack advance has been very similar to the delayed hydride cracking experienced in CANDU reactors, with the higher temperatures in BWRs conducive to even higher crack propagation rates.[22,23]

The role of barrier cladding in this process was found to be twofold.[23,24] The reduced corrosion resistance of unalloyed zirconium when compared to Zry-2 resulted in a more rapid generation of hydrogen. More importantly, though, the higher corrosion rates also provided cladding stress. From the Pilling-Bedworth ratio, the volumetric expansion of stoichiometric zirconium oxide (ZrO_2) is 1.56. Therefore, every 10 μm of zirconium oxidized closes the pellet-clad gap by 5.6 μm (actually, the gap closure is even higher than this ratio because the density of unalloyed zirconium oxide is quite low). A closed pellet-cladding gap in a defected rod removes operational margin since even small changes in local power can result in significant cladding stress. It was also found that barrier corrosion could continue even after the pellet-cladding gap was nominally closed, presumably a result of existing pathways through the less-than-fully dense oxide.[23] This results in a growing oxide capable of providing a stress on the cladding even in the absence of a change in rod power. This mechanism explained the propagation of the longer secondary cracks, where the stress from any change in power would have decayed before a crack could reach a given length.

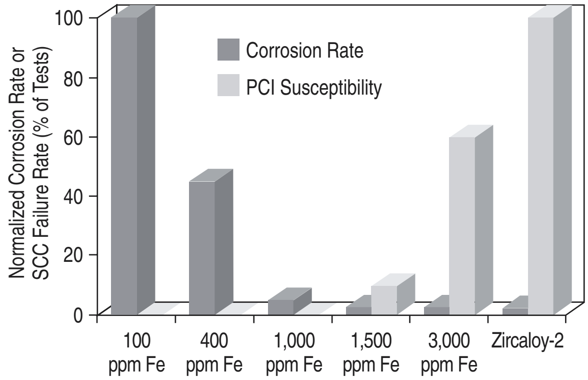

Figure 5. PCI and corrosion resistance as a function of iron alloying.

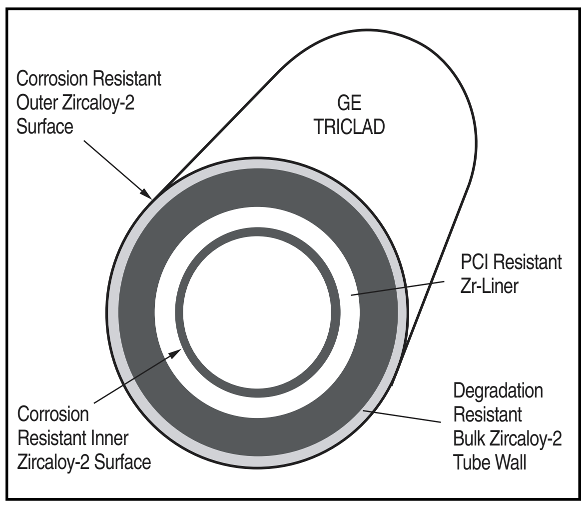

Figure 6. Cross-section of GE's Triclad Zircaloy-2 lined zirconium-barrier cladding.

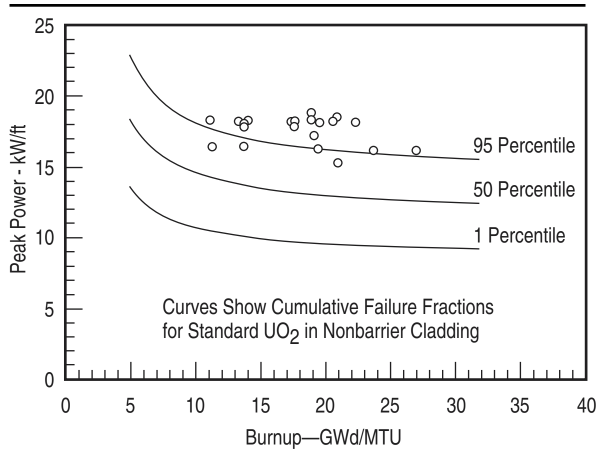

Figure 7. Typical ramp test results with additive fuel in nonbarrier cladding, where the lines represent the failure probabilities from similar ramp tests of non-additive fuel. None of the additive tests failed.

For these reasons, all fuel vendors began to develop barriers with improved corrosion resistance while attempting to maintain adequate resistance to PCI. For example, ABB conducted test-reactor ramping and decided to add tin.[19] The company's current standard barrier cladding now contains a tin-alloyed liner. The approach of both GNF and Siemens has been to alloy with iron. The current barrier offering of GNF (designated "Process 7") contains more than 1,000 ppm iron,[25] while that of the Siemens "High Fe" liner is close to 4,000 ppm.[26] (The iron level in nominally pure zirconium barriers ranged from about 100 to 400 ppm.) The difference between the two products reflects the difference in strategy. Given that previous studies have shown the addition of iron is detrimental to PCI resistance, the lower iron level would be expected to have more margin to PCI failure. Therefore, GNF has taken a more conservative step from the experience base. On the other hand, while corrosion resistance of both 1,000 ppm and 4,000 ppm is quite good, the 4,000 ppm should have slightly more margin in terms of corrosion. The balance between corrosion and PCI resistance can be measured in a number of ways, and the answer is partially a function of how it is measured. The measurements by GNF are shown in Figure 5. This chart shows the improvement in corrosion resistance with increasing iron overlaid with the laboratory PCI resistance as a function of iron. While the test is clearly able to demonstrate the effect of iron on PCI resistance, no correlation between the laboratory test and in-reactor behavior is currently available. In-reactor experience will ultimately show how much margin to PCI has been retained by the advanced barrier designs.

Another approach that was developed by GNF, along with its alloyed barrier, is the Triclad product.[27] This product is similar to standard barrier cladding except that a thin layer of corrosion-resistant Zry-2 is bonded to the inner surface of the zirconium barrier. Figure 6 is a schematic of Triclad that, counting the microstructural gradient from heat treating, contains a total of four layers: an inner layer of corrosion-resistant Zry-2 to slow oxidation and hydrogen generation and to delay local hydride formation in the case of rod perforation; a zirconium barrier for PCI resistance to blunt cracks nucleated at the inner surface; bulk parent Zry-2 tubing with large second-phase particles to resist changes under irradiation; and an outer layer of Zry-2 processed for high resistance to uniform and nodular corrosion. As per the conventional barrier tubing, the bonding at both surfaces is continuous. Williams et al.[28] summarized various mechanical, corrosion, and fracture test results on Triclad. Autoclave tests with axial slits of 2.5 cm containing UO_2 pellets in 673 K steam at 6.8 MPa for five days clearly indicated the superiority of the Zry-2 liner in restricting hydrogen pickup and hydride formation. The inner Zry-2 liner restricted hydrogen pickup to ten percent of that observed in the zirconium-barrier cladding sample. Laboratory tests have also shown that Triclad has greatly improved resistance to PCI when compared to nonbarrier, although the results are similar for other alloyed barriers. Again, operational experience will ultimately determine the success of this concept. Experience to date has been excellent, with no incidences of degradation. One disadvantage of Triclad, however, is that it is relatively expensive to produce, so the number of operating reloads is limited.

ADDITIVE FUEL

The future of robust LWR fuel designs could be additive fuel in nonbarrier cladding. The development experience for additive fuel is nearly as extensive as for barrier cladding.[29] GE, in collaboration with development partners at Hitachi and Toshiba, conducted extensive laboratory and in-reactor studies to establish the characteristics of additive fuel. The additive studies consisted of an alumina-silica glass added to standard UO_2. The glass, being virtually insoluble in UO_2, forms a second phase around all the UO_2 grain boundaries. Both in-reactor and laboratory tests have demonstrated the additive increases of the creep rate of the fuel. This, in turn, reduces the stress on the cladding. A second effect of the glassy phase is that it is able to trap fission gases and presumably delays fission product release to the cladding surface where it can participate in the PCI mechanism. A graph comparing in-reactor ramp test experience of additive fuel with similar barrier tests is shown in Figure 7. The resistance of the additive fuel is at least comparable to barrier cladding.

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