LOCA Results for Advanced-Alloy and High-Burnup Zircaloy Cladding

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The ANL test program is directed toward providing LOCA-relevant data for unirradiated advanced-alloy cladding, unirradiated Zircaloy-2 (Zry-2) and Zircaloy-4 (Zry-4) cladding, and fueled high-burnup Zry-2 and Zry-4 cladding. The purpose of the advanced-alloy program is to determine the post-quench ductility of Zr-1Nb (M5) and Zr-1Sn-1Nb (ZIRLO) alloys, as compared to the ductility of Zry-4 and Zry-2 oxidized and quenched under the same time-temperature conditions. The Zry-4 and Zry-2 data also serve as baseline data for the performance of fueled high-burnup segments subjected to LOCA integral and post-quench ductility tests. The data are provided to NRC and the nuclear industry for their independent assessment of the adequacy of licensing criteria (10 CFR 50.46) for loss-of-coolant accident (LOCA) events. The current embrittlement criteria limit peak cladding temperature (PCT) to 2200°F (1204°C) and maximum oxidation (expressed as equivalent cladding reacted, ECR) to 17% to ensure core coolability during and following emergency core cooling system quench. The presumption is that cladding oxidized to =17% ECR at =1204°C PCT will retain enough ductility to resist fragmentation during and following quench. The retention of post-quench ductility is a more limiting requirement than survival of thermal stresses during quench.

The advanced-alloy test program includes: 17×17 low-tin Zry-4, ZIRLO, and M5 PWR cladding; 10×10 Zry-2 BWR cladding; and Russian E110 (Zr-1Nb) PWR tubing and cladding. The test matrix specifies two-sided steam oxidation of Zry-2, Zry-4, ZIRLO and M5 at 1000-1260°C up to times corresponding to calculated ECR values of 5-20%. Following oxidation, the specimens are slow-cooled to 800°C and then rapidly quenched. The measured pre-test cladding wall thickness and specimen weight gain are used to calculate the experimentally determined ECR. For selected samples, oxygen and hydrogen concentrations are measured and metallographic analyses are performed to aid in the interpretation of test results. Room-temperature (RT) diametral compression tests are conducted on rings cut from oxidized-and-quenched specimens. Load-displacement data are analyzed to determine plastic ductility by the off-set displacement method. For samples that exhibit nil RT ductility at =20% ECR, compression tests are repeated at 135°C to determine the nil-ductility ECR relevant to post-quench coolant temperatures. LOCA integral tests – one per alloy per oxidation temperature – are conducted to generate ballooned-and-burst specimens at =17% ECR, which are then subjected to four-point-bend ductility tests. Post-bending diametral compression tests are planned for rings cut from axial locations outside the ballooned region. Oxygen and hydrogen concentrations are measured near bend-test failure and ring locations.

Tests have been completed for Zry-4, ZIRLO and M5 specimens oxidized to =20% ECR at 1100°C and at 1000°C. Diametral compression tests and hydrogen-pickup determinations have been completed for the 1100°C samples. Characterization and compression tests are in progress for the 1000°C specimens. Post-quench-ductility data for 1200°C and 1260°C specimens will be generated during this calendar year. In addition, over 40 E110 specimens have been oxidized at 950-1200°C – mostly at 1000°C – to determine the evolution of oxide instability leading to local and global breakaway oxidation, spallation, delamination, high-hydrogen pickup and embrittlement at low ECR values. The effects on oxide instability of E110 surface roughness and chemistry have also been studied.

Test results for Zry-4, ZIRLO, and M5 oxidized at 1100°C indicate that all three alloys exhibit about the same weight-gain kinetics, low hydrogen pickup, and retention of ductility up to 20% ECR. The M5 and ZIRLO ductility decrease with increasing ECR is comparable to that of Zry-4 and consistent with classical oxygen embrittlement (i.e., decrease in prior-beta layer effective thickness with increasing oxidation; decrease in prior-beta layer ductility with increasing temperature). M5 oxidation tests at 1000°C confirm its low oxygen pickup rate as compared to Zry-4. For all tests conducted to date, the three alloys exhibit intact, black oxide layers, which are associated with protective oxide, parabolic oxidation kinetics and low hydrogen pickup.

In contrast, E110 oxidized at 1100°C exhibits nodular breakaway oxidation – white and cracked in appearance – leading to oxygen-and-hydrogen embrittlement at <13% calculated ECR. E110 is more challenged at 1000°C: local oxide instabilities grow, crack, interlink, delaminate, spall and provide easy pathways for hydrogen pickup (>4000 wppm at 1400-s test time). Ductility decrease for these samples correlates well with hydrogen pickup: nil ductility is observed for H > 400 wppm at >650 s. Instability initiation (i.e., <30-µm surface white spots), breakaway oxidation and high hydrogen pickup can be delayed significantly by polishing the E110 surfaces to ˜ 0.14-µm roughness prior to oxidation at 1000°C and 1100°C. Etching with HF-containing solutions tends to accelerate unstable oxide formation.

LOCA integral tests are being conducted with high-burnup fueled cladding segments from Limerick (9×9 Zry-2) and H.B. Robinson (15×15 Zry-4) reactors. Prior to testing, sibling samples are characterized with respect to fuel morphology, fuel-cladding bond, cladding oxide layer thickness, hydrogen content and high-temperature steam oxidation kinetics. Specimens that survive quench are subjected to four-point-bend tests, followed by local diametral compression tests. Companion tests are conducted with unirradiated cladding to generate baseline data for comparison with the high-burnup fuel results.

LOCA integral tests have the following sequential steps: stabilization of temperature, internal pressure and steam flow at 300°C, ramping of temperature (≈5°C/s) through ballooning and burst to ≈1204°C, hold at 1204°C for 1-5 minutes, slow-cooling (≈3°C/s) to 800°C, and water quenching at ≈800°C. As reported at NSRC-2002, two high-burnup tests were completed in 2002 with Limerick BWR rod segments: ramp-to-burst in Ar followed by slow cooling; and the LOCA test with 5-minute hold time at 1204°C, followed by slow cooling. With the exception of burst-opening shape, results for burst temperature, burst pressure, burst length, and ballooning strain profile are more similar to, than different from, results for unirradiated Zry-2 cladding exposed to the same time-temperature history. Due to hot-cell unavailability, the 3rd Limerick test with quench has been delayed until this fall. It will be followed by tests on high-burnup Robinson PWR fuel segments.

Extensive characterization has been performed on unirradiated companion-test specimens (measured ECR ˜ 18%) to determine axial profiles of hydrogen pickup, oxygen pickup and ECR. Transverse metallography and oxygen analysis have also been done at several axial locations to determine the circumferential variation of metal thickness prior to oxidation and of oxygen pickup. Both microstructure and oxygen concentration results suggest a cross-section that varies from very brittle near the burst edges to ductile 180° from the burst opening. The response of this cross-section to axial bending may appear brittle (based on bending moment vs. deflection data) if the burst area is subjected to axial tensile loads. Away from the burst region, the ECR and oxygen concentration decrease as expected with increasing wall thickness and decreasing inner-surface oxidation, but the hydrogen concentration increases dramatically to >3000 wppm near and just beyond the two neck regions. These high hydrogen regions are most certainly brittle. Characterization of the 2nd high-burnup LOCA integral test specimen is in process to determine the degree of inner-surface oxidation in the ballooned region and the amount of secondary hydriding at and beyond the neck regions.