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Embrittlement of pre-hydrided Zircaloy-4 by steam oxidation under simulated LOCA transients

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ABSTRACT

During a Loss Of Coolant Accident (LOCA), the mechanical behavior of high temperature steam oxidized fuel rods is an important issue. In this study, as-received and pre-hydrided axial tensile samples were steam oxidized in a vertical furnace and water quenched in order to simulate a LOCA transient. The samples were then subjected to a mechanical test to determine the failure conditions. Two different rupture modes were evidenced; the first one associated to linear elastic fracture mechanics and the second one is associated to sample failure without applied load. The oxidized cladding fracture toughness was determined relying on intensive metallographic analysis. The sample failure conditions were then back predicted confirming that the main rupture parameters are well captured.

1. Introduction

During a Loss of Coolant Accident (LOCA), initiated by a break of a pipe of the reactor primary coolant loop, the core can be uncovered. The fuel cladding exposed to high temperature under steam environment oxidizes. Rods are then quenched by the water of the safety injection system. An important requirement is the preclusion of rod ruptures. The fuel rod failure is often considered as resulting from axial shrinkage of the cladding tube impeded by the guide tubes during the reactor core reflooding [1]. This loading might induce a cladding fracture with two or more separate cladding fragments corresponding to the so-called guillotine fracture. In the following, this situation is considered as cladding rupture. It is thus of primary interest to determine the maximum axial load that a steam-oxidized rod can withstand during the quench.

JAEA developed integral thermal shock test with axial restraint [1–6]. Both pre-hydrided and irradiated cladding rod strength during the quench was evaluated using this test. In the recent papers [3–6], many tested samples were machined on 17x17 type cladding tube geometry. Assuming that in-reactor corrosion main effects are hydrogen up-take and metal cladding thickness reduction, cladding tube were pre-hydrided up to about 1300 wppm and their thickness was reduced from 0.57 mm to 0.51 mm, to account for the influence of corrosion. This assumption was rather confirmed based on a limited number of tests performed on irradiated claddings [6] but other authors [7] rather indicated a complex influence of the corrosion layer, on the ballooning phase and on the subsequent steam oxidation embrittlement. This 60 μm metal thickness reduction corresponds to about 90 μm cumulated (at inner diameter and outer diameter) oxide layer thickness. The tested rodlets were internally pressurized, had inserted alumina pellets and were tightly sealed before the tests. The JAEA test consists in applying a thermo-mechanical transient to the rodlet, heated from outside using a four lamps infra-red furnace. In a first phase, the rodlet deforms inducing ballooning and burst of the cladding. The test sample is exposed to steam, oxidizing the rodlet, mainly at the maximum target temperature and is slowly cooled after this period. When the cooling reaches a temperature, usually comprised between 700 and 800 °C, the sample is then water quenched under applied axial load. After the test, the rod is examined to determine if it failed or not. The post-test examinations confirmed the presence of strong secondary hydriding surrounding the ballooned region. The ballooned region is rather subjected to two sided oxidation during steam exposure, affecting both, inner and outer surfaces. The secondary hydriding region, rather away from the ballooning opening, is subjected to single sided oxidation at the outer surface whereas the hydrogen

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adsorption takes place at the inner side [8,9]. Between these two regions, the oxidation is intermediate between single and two-sided oxidation with maximum zirconia layer thickness at the outer diameter. Two important conclusions were further drawn from the tests on pre-hydrided materials [3,4]:

- increasing axial load promotes fracture in the balloon region rather than in the secondary hydriding region,
- post-test local hydrogen content in the balloon region remains close to its initial value.

The cladding wall thinning in the ballooned region and the double sided oxidation imply a modified calculation of the Baker–Just ECR taking into account the average cladding thickness obtained from metallography at the maximum ballooning location.

As a consequence of the two above mentioned conclusions, Nagase [4,5] decided to discriminate the failure and non-failure conditions using mainly two tests parameters: the calculated local ECR in the balloon region and the nominal hydrogen content. In this mapping, the boundary discriminating rupture and non-rupture data is considered as limiting the safe region. The local ECR is determined with the Baker–Just correlation [10], using the deformed cladding geometry in the ballooned area. This procedure provided a direct understanding of each integral test using a minimum number of parameters.

However, there are difficulties in analyzing some of the test results. Uetsuka and Nagase [1,3] indicate that the failure location was strongly influenced by the applied load: under very intense axial load, the rupture clearly happens in the balloon region, whereas rod fracture was rather located in the secondary hydriding region in the absence of axial loading. Grosse [8] and Stuckert [9], relying on detailed examinations, rather concluded that the failure location is affected by several complex parameters such as the presence of small cross cracks at the burst opening but also the location of the considered rod within a bundle. Grosse and Stuckert also confirmed that the level of secondary hydriding is influenced by the duration of high temperature exposure. The hydrogen distribution along the rodlet recently appeared to be influenced by the fuel pellet cladding gap [11] and the axial location of failure rather appears between the maximum ballooning section and the secondary hydriding region. Additionally, some influence of irradiation on secondary hydriding and oxidation was evidenced [6], the corrosion layer formed during irradiation delays the high temperature oxidation, but high temperature oxide then forms at the tip of cracks between oxide fragments. Moreover, Nagase considered that the corrosion layer influence disappears after a short period and has negligible impact on the oxidation rate. The measured maximum hydrogen content obtained at secondary hydriding regions on irradiated rodlets at JAEA [6] is rather lower than the one of comparable samples using pre-hydrided claddings. However, for long duration steam exposure, some of the tests showed higher hydrogen content in the balloon than before the test. A similar trend was observed in some of the ANL LOCA tests [12] performed on irradiated claddings without applied load. The analysis of the ANL tests also showed that for high burnup claddings, the secondary hydriding peak has the same order of magnitude than the one of comparable tests performed on fresh claddings.

As a consequence of the complexity of the many phenomena activated during the JAEA semi-integral tests, IRSN considers that there is a need to perform separate effect tests to provide a mechanistic understanding of the fracture conditions of these tests. Beyond the influence of the considered location on rod integrity, a failure or non-failure prediction for the entire rod is expected.

Considering the JAEA integral thermal shock test, the lower boundary of failure-non failure conditions in terms of applied load, hydrogen content and ECR consists in a limit below which the rod is not expected to fail. This limit corresponds to the worst possible embrittlement and consequently to rod ruptures at room temperature which is the lowest temperature achieved during these tests. As an illustration for that, a rodlet that failed during the quench would certainly fail at room temperature. A straightforward consequence of this is that the rodlet oxidation and mechanical resistance at room temperature can be sequentially tested to define the room temperature non failure limit. Consequently, the present study aims at determining the room temperature fracture mode of oxidized cladding under simulated LOCA conditions. The main phenomena affecting the sample rupture are described and analyzed. Several 70 mm long Zircaloy-4 (Zry-4) cladding tubes were thus pre-hydrided with hydrogen contents ranging between the as-received content (~11 wppm) up to about 4000 wppm. The samples were machined to provide pre-hydrided axial tensile samples which were steam oxidized at 1200 °C, and quenched in a water bath at room temperature. The 1200 °C temperature was chosen consistently with LOCA safety criteria. When possible, the high temperature oxidized sample was then submitted to a mechanical test in order to determine its tensile fracture load.

2. Experiments

2.1. Materials

Stress Relieved Annealed (SRA) low-tin Zry-4 with nominal chemical composition described in Table 1 is used in this study. The alloy was manufactured by CEZUS. The outer diameter of the tubes is 9.5 mm and the cladding thickness 0.57 mm.

2.2. Testing protocol

As-received Zry-4, with composition described in Table 1, tubular samples are first hydrogen charged and then machined into axial tensile samples. The sample hydrogen content is then assessed. The axial tensile samples are oxidized at 1200 °C under steam environment, and water quenched, without any cooling phase. The oxidized tensile samples are then mechanically tested at room temperature. After mechanical testing several post-test examination and measurements are performed. The details of this protocol are described in the following.

2.3. Hydrogen charging of the samples

The hydrogen charging is performed at the Ecole Centrale de Paris using 70-mm long cladding tubes with open ends. The Zry-4 sample is inserted in a tight quartz tube heated by a furnace. Pure hydrogen is injected in high vacuum at 520 °C for most of the test samples and 420 °C for two samples. This temperature is chosen below the α/β phase transition whatever the hydrogen content is.

2.4. Sample machining

After hydrogen charging, a 1 mm long ring is removed at each sample ends. Then, the approximately 70 mm long tube is spark machined to avoid residual stress deposition in the gage section. The sample geometry is schematically illustrated in Fig. 1. Scraps

Table 1
Measured chemical composition of the tested SRA Zry-4.

Sn (wt%)	Fe (wt%)	Cr (wt%)	O (wt%)	H (wppm)
1.30 ± 0.01	0.22 ± 0.01	0.11 ± 0.01	0.13 ± 0.01	7