Evaluation of a Ductility after High Temperature Oxidation with the Three-Point Bend Test in Zirconium Alloys

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1. Introduction

In a light water reactor, the fuel cladding play an important role of preventing leakage of radioactive materials into the coolant, and thus the mechanical integrity of the cladding should be guaranteed under the conditions of normal and transient operation [1]. In the case of a loss of coolant accident (LOCA), the cladding is subjected to a high temperature oxidation which is finally quenched because of an emergency coolant reflooding into the core. In this situation, the current LOCA criteria consist of five separate requirements: i) peak cladding temperature, ii) maximum cladding oxidation, iii) maximum hydrogen generation, iv) coolable geometry, and v) long-term cooling [2]. The claddings lose their ductility due to the microstructural phase transformation from beta to martensite alphaprime [3], and hydrogen up-take [4] after LOCA. Since the reduction in ductility can induce embrittlement of claddings, post-quench ductility is one of the major concerns in transient operation circumstances. For the analysis, usually ring compression test are performed on ring samples cut from the tube to examine the oxidized cladding ductility. However, the test would not be applicable to the platelet samples which are general form of a specimen for developing alloys.

As a high burn-up fuel cladding materials, Zircaloys are being replaced by modern zirconium alloys such as ZIRLO, and M5 [5,6]. Korea has also developed a new fuel cladding material HANA (high performance alloy for nuclear application) by the Korea Atomic Energy Research Institute [7,8]. Because of the different composition of the newer claddings in comparison with the conventional Zircaloy-4, the high temperature oxidation behavior and the ductility after the oxidation would be different, and the properties should be evaluated how much the newer claddings were improved.

2. Methods and Results

2.1 High Temperature Oxidation

The apparatus for the high-temperature testing was established by modifying the thermo-gravimetric analyzer (TGA; Shimadzu, TGA-51H) [1,9]. The specimens were hung on a microbalance above an electric furnace. The balance chamber was purged with an argon backflow to prevent the ingress of steam. A steam generator was attached to the apparatus to supply steam continuously into the furnace. Steam flow was maintained at a constant rate during the oxidation test. The weight change was measured by an in situ method to within ± 0.001 mg during the oxidation reaction. Other conditions and experimental details were referred to our previous papers [1,9].

2.2 Three-Point Bend Test

The oxidized samples with a dimension of 10 mm x 10 mm x 1 mm were loaded on the three-point bend jig (stand) with span length of 5 mm. Fig. 1 illustrates the schematic layout of the stand, and equations for the stress and strain during the three-point bend test. For the test, compressive stress with constant displacement rate of 0.05 mm/min was applied.



Fig. 1. Test stand and equations for the stress and strain during the three-point bend test.

2.3 Zircaloy-4

For the high temperature oxidation, Zircaloy-4 platelet samples gain weight 2615 mg/dm² for 1880 s. Fig. 2 shows the oxide weight gain of Zircaloy-4 during high temperature oxidation at 1100° C.



Fig. 2. High temperature oxidation of Zircaloy-4 at 1100°C

Fig. 3 shows flexural stress (σ) and strain (ϵ) during the three-point bend test. Oxide film on the Zircaloy-4 specimen was broken at the point of first load drop (~0.009 strain) and then fractured itself at the point of second load drop (~0.01 strain). The maximum stress was 279 MPa and maximum strain was about 1% in Zircaloy-4. The graph represented well the brittle fracture of the sample that underwent high temperature oxidation.



Fig. 3. Flexural stress and strain of Zircaloy-4 during a threepoint bend test.

2.4 HANA alloys

HANA-4 and HANA-6 alloy samples were also analyzed. The oxide weight gains were 1789 mg/dm² for HANA-4, and 1664 mg/dm² for HANA-6 during high temperature oxidation at 1100°C for 1900 s. The oxidation resistance of HANA alloys was higher than that of Zircaloy-4. The ductility was also obtained much higher than Zircaloy-4 as shown in Fig. 4. The maximum flexural stress was 427 MPa and maximum strain was about 1.6% in HANA-4, and 659 MPa and ~3.2% in HANA-6, respectively.



Fig. 4. Flexural stress and strain of HANA4 and HANA-6 during a three-point bend test.

For the comparison of the ductility, the area under the stress-strain curve was measured. The value was 1.6 MPa for Zircaloy-4. In the case of HANA, these were 3.6 MPa for HANA-4, and 10.9 MPa for HANA-6 alloys, respectively. This result is consistent with the post-quench ductility done by a ring compression test.

3. Conclusions

Three-point bend test was suggested as a method for the evaluation of ductility after a high temperature oxidation. The samples after high temperature oxidation were brittle enough to evaluate their ductility with this destructive test. Therefore, it is expected to replace the post-quench ductility evaluation – that performed by using a ring compression test. The test can be applicable to platelet samples which are generally manufactured during alloy development stage, and thus enable to evaluate (at the relatively early stage) how much the alloys would be safe at the accident circumstances. The developed alloys in KAERI, i.e. HANA, showed very high ductility after severe oxidation at $1100^{\circ}C$.

REFERENCES

[1] J.H. Baek, Y.H. Jeong, Steam oxidation of Zr-1.5Nb-0.4Sn-0.2Fe-0.1Cr and Zircaloy-4 at 900-1200C, J. Nucl. Mater., 361 (2007) 30–40.

[2] USNRC-SRP (Standard Review Plan), Sec. 4.2, NUREG-0800.

[3] J.H. Kim, M.H. Lee, B.K. Choi, Y.H. Jeong, Embrittlement behaviour of Zircaloy-4 cladding during oxidation and water quench, Nucl. Eng. Design, 235, (2005) 67–75.

[4] J.C. Brachet, L. Portier, T. Forgeron, J. Hivroz, D. Hamon, T. Guilbert et al., Influence of hydrogen content on the α/β phase transformation temperatures and on the thermalmechanical behaviour of Zy-4, M4(ZrSnFeV) and M5(ZrNbO) alloys during the first phase of LOCA transient. ASTM STP 1423, Edited by G.D. Moan, P. Rudling, American Society of Testing and Materials, (2002) 673–701. [5] G.P. Sabol, R.J. Comstock, R.A. Weiner, P. Larouere, R.N. Stanutz, In reactor corrosion performance of ZIRLO and

Zircaloy-4, ASTM STP 1245, Edited by A.M. Garde, E.R. Bradly, American Society of Testing and Materials, (1994) 724–524.

[6] J.-P. Mardon, D. Charquet, J. Senevat, Influence of composition and fabrication process on out-of-pile and in-pile properties of M5 alloy, ASTM STP 1354, Edited by G.P. Sabol, G.D. Moan, American Society of Testing and Materials, (2000) 505–524.

[7] Y.H. Jeong, S.-Y. Park, M.-H. Lee, B.-K. Choi, J.-H. Baek, J.-Y. Park, J.-H. Kim, H.-G. Kim, Out-of-pile and inpile performance of advanced zirconium alloys (HANA) for high-burn-up fuel, J. Nucl. Sci. Technol. 43 (2006) 977–983.

[8] K.W. Song, Y.H. Jeong, K.S. Kim, J.G. Bang, T.H. Chun, H.K. Kim, K.N. Song, High burnup fuel technology in Korea, Nucl. Eng. Tech. 40 (2008) 21–36.

[9] J.H. Baek, K.B. Park, Y.H. Jeong, Oxidation kinetics of Zircaloy-4 and Zr-1Nb-1Sn-0.1Fe at temperature 700-1200C, J. Nucl. Mater., 335 (2004) 443-456.