Study on the high temperature oxidation and mechanical properties of hydrided Zircaloy-4

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1. Introduction

Loss of the coolant accident (LOCA) is one of the most important design-based accidents concerning the behavior of a fuel cladding. During a typical LOCA condition, the fuel cladding balloons due to increasing oxidation temperature and internal pressure [1-3]. And high temperature oxidation, rapid quenching, rupture were occurred sequentially. Nowadays, a nuclear power plants work high burn up operation for the purpose of the economy [4]. To maintain the fuel integrity under postulated LOCA conditions, the Nuclear Regulatory Commission (NRC) established the fuel safety criteria about a LOCA, where the peak fuel temperature and total oxidation cannot exceed 1204 ℃and a 17 % level , respectively [5].

The hydrogen charged in the Zr-based alloys is important element which affects on embrittlement of the fuel claddings. The effect of hydrogen plays a role in decrease of a safety of the fuel claddings under the LOCA condition. This study investigated the effects [6] of hydride on the high temperature oxidation and mechanical properties of Zircaloy-4 in a LOCA condition.

2. Experimental procedure

The test material was hydrogen charged Zircaloy-4 cladding (Zr-1.35Sn-0.2Fe-0.1Cr). The pre-hydrided Zircaloy-4 samples were prepared by the hydriding at 400℃ with hydrogen partial pressure of 350 torr. Fig.1 shows the cross-section of typical hydrided sample. Cladding type samples were cut into 8mm in length and were ground carefully up to Grit No. 1200 of SiC paper and then pickled in a solution of 5 % HF, 45 %HNO_3 and 50 %H_2O and cleaned ultrasonically in an ethanol and acetone solution. Oxidation test in steam was conducted by using thermo-gravimetric analyzer (TGA) at 1000℃ [7]. The duration time was from 120 to 18000 sec. The microstructure and oxide layer were observed by using optical microscope (OM). In order to investigate the mechanical properties of the samples, Vickers micro-hardness tester was utilized for the specimens. Ring compression tests for high temperature oxidized cladding tubes were performed at room temperature.

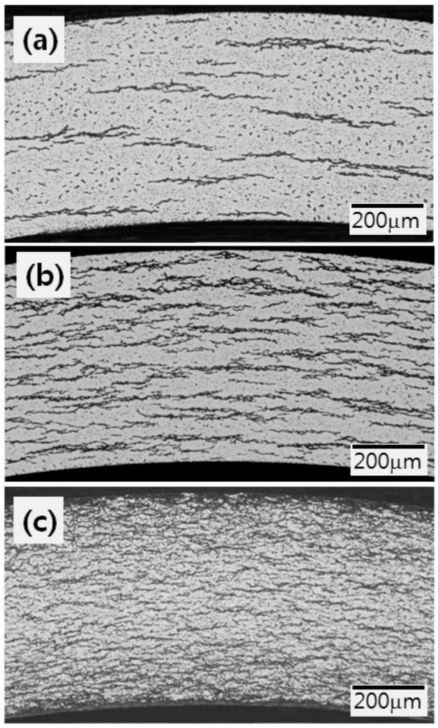

Fig. 1. Hydride morphology with hydrogen concentration of (a) 100 ppm, (b) 300 ppm and (c) 600 ppm.

3. Results and Discussion

3.1. Oxidation behavior

Fig.2 shows the oxidation behaviors of the hydrided Zircaloy-4 cladding for the steam oxidation experiment performed at 1000^{o}C. In Fig. 2(a), the oxidation kinetics for an exposure time of less than 1000 s obeyed the parabolic rate law, which is generally accepted for a high-temperature oxidation of Zr-based alloys under the steam atmosphere at that temperature. The weight gains of the hydrided Zircaloy-4 slightly increased with hydrogen concentration in early stage of high temperature oxidation. The long-term oxidation behaviors of the 300 ppm hydrogen charged specimen are shown in Fig. 2(b). Up to the exposure time of 12000s, the weight gain of the specimen increased monotonously. The breakaway oxidation of the 300 ppm hydrogen charged specimen occurred after exposure for about 12000s. The increase of an oxidation rate after breakaway was very small in the ranges of hydrogen concentration.

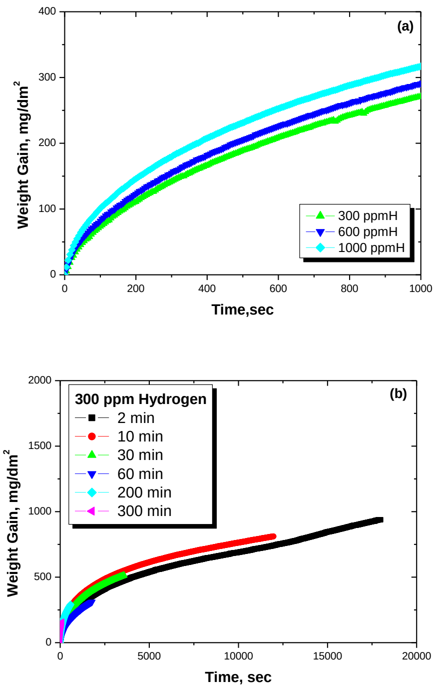

Fig. 2. Oxidation behaviors of hydrided Zry-4 at 1000 °C for (a) short-term (1000 s) and (b) long-term (18000 s) periods.

3.2 Mechanical properties

The as-received (non-oxidized) sample first deforms from circular to elliptical shape. And it just deforms into a peanut-shape. The oxidized with different times deforms into a peanut-shape with a crack at equatorial azimuths finally. It means that the effect of loads influence on the outer surface of the ring and propagate toward the inner surface. And buckling failures of inner oxide layer at the equatorial azimuths were occurred and then all tested specimens failed.

Fig. 3 shows seven load–displacement curves recorded during the ring compression tests of 300 ppm hydrided Zircaloy-4 oxidized at 1000°C. The deformation behavior of an as-received (non-oxidized) samples were completely plastic, it deformed without cracking in the ring compression test. On the basis of the load–displacement curves of the ring compression tests, the ductile or brittle character of the hydrogen charged specimens can be determined. One can distinguish between two types of curves: Ductile curves are characterized by a horizontal ductile plateau after the elastic deformation (AR and 2min oxidized one in Fig. 3). In the case of brittle curves, the ductile plateau is not formed, and the elastic deformation is followed by a quick breakdown indicating the formation of cracks and the rupture of the sample (10-300 min oxidized one in Fig. 3).

As increased oxidation time, the failures were occurred at early stage. It was anticipated that the claddings became brittle because of rapid oxidation by oxygen diffused easily through the oxide as oxidation time increased.

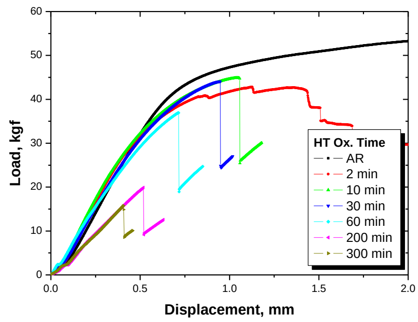

Fig. 3. Load-displacement diagrams recorded during radial ring compression testing of 300 ppm hydrogen charged Zircaloy-4 oxidized in steam at 1000°C.

4. Conclusions

The high temperature oxidation and mechanical properties of hydrided Zircaloy-4 cladding were investigated. The influence of pre-hydriding on the high-temperature oxidation kinetics was found to be very small in the ranges of hydrogen concentration and oxidation times examined. The effect of hydride on ductility, however, was very severe. The ductility of the hydrided Zircaloy-4 claddings was evaluated on the basis of the load–displacement curves. The ductile samples were characterized by a horizontal ductile plateau after the elastic deformation, while in the case of brittle samples the ductile plateau was missing.

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