High Temperature Oxidation Behavior of Zircaloy-4 under Applied Load

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1. Introduction

Zirconium-alloy cladding is the most important barrier against releasing radioactive fission products in the fuel. At the accidental situation, the inner pressure of the fuel might be higher than the reactor pressure. Once the cladding is under the tension state at high temperatures (i.e., accident cases), the cladding starts to be oxidized with undergoing creep. Hence, creepenhanced oxidation is expected during accidents of nuclear reactor.

Nuclear Regulatory Commission (NRC) regulation set LOCA accident criteria. The oxidation amount of cladding during an accident is generally calculated by the Baker-Just (B-J) correlation [1]. But, B-J correlation only considers oxidation temperatures, but, does not considered the effect of creep. Generally it is known that the oxidation rate is accelerated under loaded condition, however; no detail measurements on high temperature oxidation with load have been done so far. Therefore, the goal of this study is to investigate the oxidation behaviors of claddings at high temperature under load. In this study, specifically load is uniaxial.

2. Experimental

2.1 Apparatus

Fig.1 shows the equipment for the oxidation at high temperature under load. It consists of an electrical resistance heater, an alumina tube, kanthal wires, and cap. Temperature can be raised up to 1200 $^{\circ}$ C, the heat up rate is about 3 $^{\circ}$ C/sec. Specimen is oxidized in the alumina tube in air.



Figure 1. Experimental setup for loaded oxidation in air.

2.2 Specimen

Commercial Zry-4 tubes were used this study. The tubes were cut to specimens of about 30mm. Each specimen had 4 drilled holes – two at the top and two at the bottom (fig.1). Measured hot zone size of the heater was about 1cm. The specimen was degreased and pickled in aqueous HF/HNO3 solution, then cleansed in hot and cold water.

2.3 Analysis

When the temperature was reached at a set point, the specimen was put in the hot zone of the alumina tube. After oxidation, the specimen was pulled out from the heater. Then it was cooled down to the room temperature by air.

After the oxidation test, we measured the length and the weight of the specimen, and calculated the weight gain and creep strain. The specimen was also molded, grinded and polished for the optical microscopic (OM) observation. And scanning electron microscope (SEM) and X-ray diffractometer (XRD) was used for the examination of the specimens.

3. Result and discussion

The results of the experiments are shown in Fig. 2. Creep-enhancement of oxidation was clearly noticed for all specimens under load. When a specimen was oxidized too much, spalling of oxide layer was observed. There is a possibility that some specimens may reveal lower weight gain due to spalling. The more load was given to the specimen, the more oxidation was occured. Creep certainly accelerated the oxidation [2].



Figure 2. Weight gain of Zry-4 at 1000 ℃ under load.

Fig.3 shows the creep strains of specimens under uniaxial load during oxidation. Creep rate increases with oxidation time due to thining of the metal layer.



Figure 3. Creep strain of Zry-4 at 1000 °C by load.

Fig.4 shows cross-sectional views of specimens. Unload specimen shows uniform oxide and metal layers, but loaded specimens have non-uniform and irregular interface layers. Cracks or broken oxide layers are observed in the specimens experiencing creep-enhanced oxidation [3].



Figure 4. Cross-sectional view of Zry-4 specimen. (a) no load (b) 1.53kg/cm² (c) 4.60kg/cm²

Fig.5 shows surfaces of the specimens. There are many cracks in the loaded-specimen with spallation. These cracks seem to be the reason for acceleration of oxidation.



Figure 5. Surface of Zry-4 specimen. (a) 4.60kg/cm² (b) No load

4. Conclusion

High temperature oxidation behaviors of Zry-4 claddings under creep were studied. Clearly creepenhanced oxidation was noticed. The creep rate increased with oxidation time due to thinner metal layer after oxidation. The higher the creep rate it has, the higher oxidation rate the cladding undergoes. Oxidemetal interface was irregular in the specimen under creep-enhanced oxidation. It is believed that the tension in the oxide layer induces cracks at the surface, and this non-protective oxide results in enhanced oxidation.

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