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The Influence of Steam Pressure on the High Temperature Oxidation of Low-Sn Zircaloy-4

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1. Introduction

Zirconium alloys have been used as a cladding material for nuclear fuel in PWRs(Pressurized Water Reactors). Therefore, it is important to maintain their integrity in an accident because they are the first barrier against the release of radioactive fission products. Many researchers have studied to evaluate the oxidation behavior between the high temperature steam and the cladding materials. Finally, U.S. NRC (Nuclear Regulatory Commission) established limits and requirements including (1) peak cladding temperature, (2) maximum cladding oxidation, (3) maximum hydrogen generation, (4) coolable geometry, and (5) long-term cooling.

The amount of cladding oxidation during an accident is generally calculated by the Baker-Just correlation[1], but this correlation considers only the oxidation temperature and time, regardless of the steam pressure. Some researchers have investigated the effect of steam pressure on high temperature oxidation. Cox observed the effect of steam pressure below 700℃[2]. Pawel compared the oxidation amount at different oxidation temperatures, between 900℃ and 1100℃[3]. He found that the acceleration effect caused by the steam pressure exists at 900℃, but not at 1100℃. Bramwell observed the steam pressure effect from 700℃ to 950℃[4]. But most of these studies generated only the qualitative data, without quantitative analysis or detailed observation. And in contrast with previous studies using original Zircaloy-4, Low-Sn Zircaloy-4 which has low Sn content is used to cladding material, recently.

This study investigated in detail the oxidation behavior of Low-Sn Zircaloy-4 under steam pressure.

2. Experimental

Table 1 shows the chemical composition of the Low-Sn Zircaloy-4 used in this study. The apparatus for high temperature oxidation under high steam pressure was devised and manufactured as shown Fig. 1. The test apparatus is composed of a high pressure vessel and two heaters. The outside heater enhances the steam pressure by boiling, and the inside heater directly controls the specimen's temperature.

Table 1. Chemical composition of Low-Sn Zircaloy-4

Specimen	Sn (wt%)	Fe (wt%)	Cr (wt%)	Zr (wt%)
Low-Sn Zry4	1.35	0.2	0.1	base

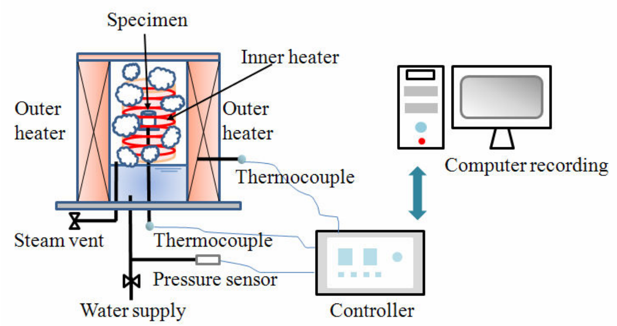

Fig. 1. Apparatus for high temperature oxidation under high steam pressure

After the pickled specimen was located in the test position, the pressure vessel was dropped and sealed. Distilled water was put into the test apparatus, and the steam pressure was enhanced by heating. Initial interior gas (air) was discharged before the test because air could affect the oxidation amount. The specimen's temperature was held below 400℃ until reaching the desired steam pressure. After the steam pressure had reached the target steam pressure, the specimen's temperature was increased to the target temperature and the specimen was oxidized at the target time. The specimen then was cooled to room temperature and pulled out, and its weight gain was measured. Optical Microscopy (OM) and Scanning Electron Microscopy (SEM) were used to observe the specimen's oxide layer and microscopic structure.

The target temperature range was from 700-900℃, and the steam pressure was from 0.1MPa-10.0MPa. The oxidation time was controlled at 1000-2000sec.

3. Results and Discussion

Fig. 2 and 3 show the oxidation amount and oxide layer thickness, along with the steam pressure. Compared to the results at 0.1MPa(atmosphere), the conditions of 700℃, 2000sec and 10.0MPa doubled the oxidation amount. At 800℃, 1500sec and 10.0MPa, the oxidation was about 2-3 times that seen at 0.1MPa; and at 900℃, 1000sec and 10.0MPa, it was about 3-4 times. That is, between 700℃ and 900℃, as the temperature increased, the oxidation amount was greatly affected by the steam pressure. After measuring its weight gain, the specimen was molded, and the oxide thickness was measured. The measured oxide thickness shows results similar to the weight gain.

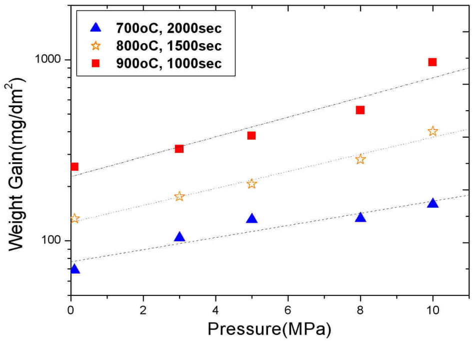

Fig. 2. Measured weight gain of Low-Sn Zircaloy-4

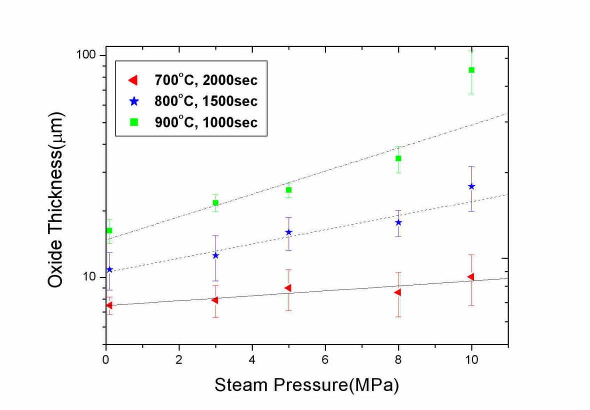

Fig. 3. Measured oxide thickness of Low-Sn Zircaloy-4

After the oxidation test, the oxidized specimen's surface appearance was observed. The surface was black under all steam pressures at 700℃, and a little change was detected under high steam pressure at 800℃. But as the steam pressure increased, a remarkable change was observed at 900℃, as shown Fig. 4. The surface was black at 0.1MPa, gray at 0.5MPa and yellow-gray at 10.0MPa. As the steam pressure increased, cracks in the oxide layer increased considerably. This shows that the cracks are closely related to the oxidation amount.

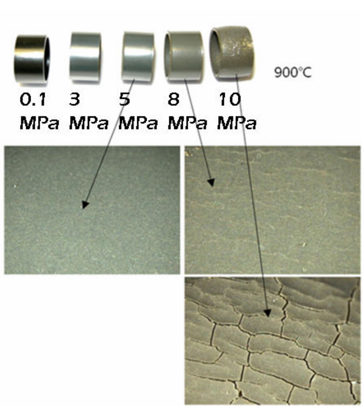

Fig. 4. The specimen's surface appearance after oxidation at 900℃

Fig. 5 represents the oxide layer image as seen by SEM. Generally, a uniform oxide layer was observed under an atmosphere of pressure. But the oxide layer formed at 800℃ under 10.0MPa was shown to be uneven. Many cracks were detected in the oxide layer formed at 900℃ under 10.0MPa. These oxide cracks accelerate the oxygen inflow.

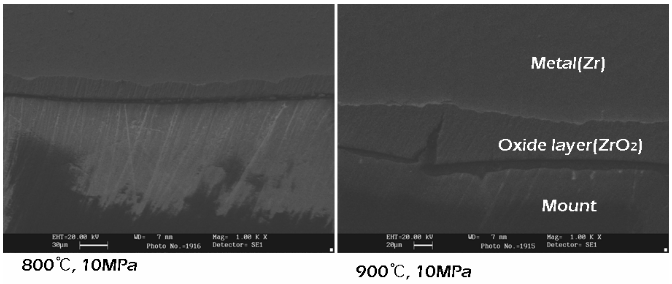

Fig. 5. SEM image of oxide layer

4. Conclusion

An effect of steam pressure on the high temperature oxidation of Low-Sn Zircaloy-4 was detected. This effect was shown to increase as the temperature increased. Many cracks in the oxide layer were detected at high temperature under high steam pressure. These cracks in the oxide layer accelerate the oxidation of Low-Sn Zircaloy-4.

Acknowledgment

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