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Investigation of Zircaloy-2 oxidation model for SFP accident analysis

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HIGHLIGHTS

- An oxidation model of Zircaloy-2 in air environment was developed.
- The oxidation model was validated by the comparison with oxidation tests using long cladding tubes in hypothetical spent fuel pool accident condition.
- The oxidation model successfully reproduced the typical oxidation behavior in air.

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ABSTRACT

The authors previously conducted thermogravimetric analyses on Zircaloy-2 in air. By using the thermogravimetric data, an oxidation model was constructed in this study so that it can be applied for the modeling of cladding degradation in spent fuel pool (SFP) severe accident condition. For its validation, oxidation tests of long cladding tube were conducted, and computational fluid dynamics analyses using the constructed oxidation model were proceeded to simulate the experiments. In the oxidation tests, high temperature thermal gradient along the cladding axis was applied and air flow rates in testing chamber were controlled to simulate hypothetical SFP accidents. The analytical outputs successfully reproduced the growth of oxide film and porous oxide layer on the claddings in oxidation tests, and validity of the oxidation model was proved. Influence of air flow rate for the oxidation behavior was thought negligible in the conditions investigated in this study.

1. Introduction

After the severe accidents in Fukushima Daiichi nuclear power plants (NPP) in 2011, concern for loss-of-cooling or lost-of-coolant severe accident in spent fuel pool (SFP) has been spotlighted [1,2]. The potentiality of a SFP severe accident drawn by aircraft attack on NPP had been noticed after the terrorism in US on September 11, 2001, and research programs on the SFP accident with experiments using mock-up fuel bundles of boiling water reactor (BWR) and pressurized water reactor (PWR) were proceeded under international collaboration. The heating experiments in air environment using mock-up bundles showed that the fuel claddings were drastically oxidized, and in this case the fuel bundles were collapsed [3–5]. Other international programs have been organized in Europe as well. Mock-up fuel bundle tests were conducted in assumption of air inlet for reactor pressure vessel which would occur in occasion of piping rupture in NPP during a severe accident. They reported that the cladding oxidation behavior was much more intense in steam/air mixture or in dry air compared to the oxidation in steam [6–8].

Isothermal oxidation tests such as thermogravimetric (TG) analyses using short samples of cladding have provided the knowledge that oxidation rate increases in air compared to steam or in pure oxygen (O_2) [9–14]. Cross sectional observation of the specimens after oxidation test in air revealed that zirconium nitride (ZrN) was precipitated in porous oxide layers which were grown beneath fractures of dense oxide film on the sample surface [10–15]. From this result, catalytic action of nitrogen (N_2) in air for the oxidation of zirconium (Zr) alloy was suggested, and it was thought that it occurred in connection with O_2 starvation in the narrow space beneath oxide film fracture at the meatal-oxide interface [10].

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There have been a limited number of reports on cladding oxidation in air from Japan [16–18]. These previous studies were not conducted for the purpose of SFP accident research, and the published experimental data are thought to be not applicable for the SFP accident evaluation. Detailed investigation on hypothetical SFP accidents is required for the development of safety measures, therefore improvement of severe accident codes by embedding a cladding oxidation model in SFP accident conditions is thought to be important.

The Argonne National Laboratory (ANL) in US and the Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in France have proposed oxidation models for cladding in air environment [9,19]. In addition, simulations using severe accident codes which were modified by introducing these oxidation models have been conducted to calculate the oxidation behavior of mock-up fuel bundles in heating tests in air environment [3,8,20,21]. These oxidation models were constructed for claddings made of alloys such as Zircaloy-4 (Zry4), ZIRLO™, and M5®, while there were only few reports on the oxidation behavior of Zircaloy-2 (Zry2) in air, even though this alloy is widely utilized in BWRs [22,23].

The authors have previously conducted isothermal oxidation tests in air on short samples of Zry2 cladding by using TG analysis apparatus, and reported in detail the oxidation behavior [24–26]. In this work, a model for Zry2 oxidation in air environment was constructed by using these basic data. The constructed oxidation model was applied for simulation of oxidation tests using long cladding tubes of Zry2 in hypothetical SFP accident conditions. The agreement between the calculations and the results of experiments was evaluated to validate the oxidation model. In addition, the influence of air flow rate on oxidation behavior was investigated.

2. Experimental

2.1. Specimen

Zry2 cladding typically utilized in BWRs was applied for oxidation tests in this work [27]. Table 1 shows the chemical compositions of the material. The Zry2 cladding is a tube of 11.2 mm in outer diameter and 0.7 mm in thickness. The cladding was cut into 500 mm in length, and a plug was welded to close the one end of a portion of the cladding. The plug was 5 mm in length and 11.2 mm in diameter. The Zr liner on inner surface of the Zry2 cladding was removed before TG analyses in previous works [24–26], but on the contrary, it was not removed in this work because the cladding sample was closed by the plug and thus the inner surface was not oxidized in the oxidation tests.

2.2. Oxidation tests

Oxidation tests on long cladding samples by applying high temperature thermal gradient along the axial direction were conducted by using the system illustrated in Fig. 1. Dry air was heated by using a gas heater up to 340 °C before it was introduced into the quartz chamber at a constant flow rate of 1840 mℓ/min and 310 mℓ/min, respectively. Air was exhausted from the upper end of the quartz chamber. The upper part of the cladding sample was heated by using an infrared gold image furnace, and the lower part was heated by using two heating mantles to give an aimed temperature distribution. Temperature was measured by using five thermocouples which were spot-welded on the surface of the sample at 10, 90, 170, 270, 450 mm from the upper end of cladding sample. The power control of the infrared gold image furnace is based on temperature data measured by using the thermocouple welded at 90 mm from the upper end of cladding sample. The power of the heating mantles was constantly set maximum through the experiment to keep the temperature of the lower part of cladding at ca. 400 °C. A typical temperature profile during an oxidation test is given in Fig. 2. The temperature profile was controlled in the same way in the oxidation tests at high and low air flow rate condition. In the pre-heating regime, the whole sample was heated up to about 400 °C. After this regime, the power of the infrared gold image furnace was increased to give a high temperature gradient, in which the highest temperature was set to 900 °C or 1000 °C at the position of 90 mm from the upper end, and the lowest temperature was set to 400 °C at the position of 450 mm from the upper end of the sample. The temperature gradient was kept the same for 3600 s, then all heaters were turned off and spontaneously cooled down to the room temperature. The air flow rate was kept constant throughout one oxidation test procedure.

The authors previously conducted steady state analyses on hypothetical SFP accident, in which all cooling water was assumed

Table 1
Chemical compositions of the Zircaloy 2 (wt.%).

Sn	Fe	Cr	Ni	Zr
1.20–1.70	0.07–0.20	0.05–0.15	0.03–0.08	bal.

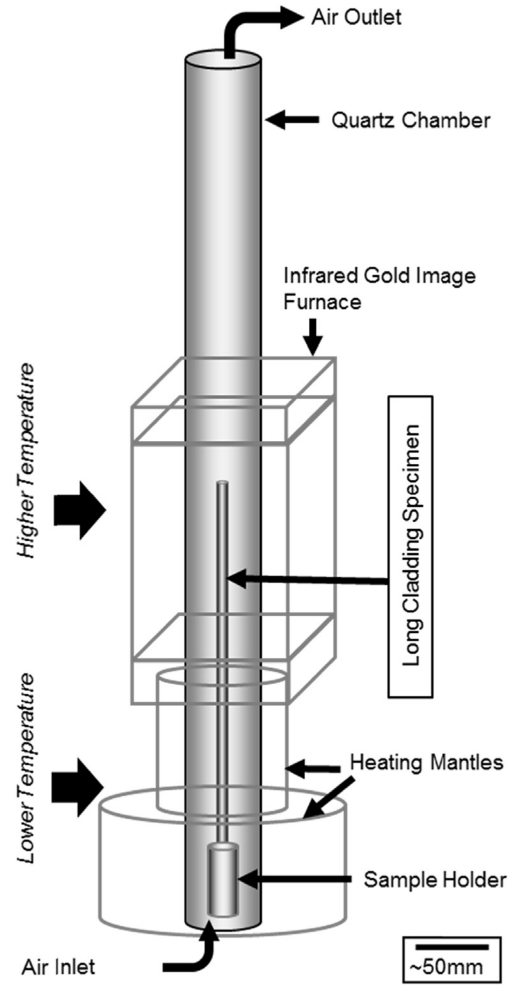

Fig. 1. High temperature oxidation test apparatus designed for long cladding specimens.