

Air Oxidation Behaviors of Zircaloy-4 Cladding During a LOCA In Spent Fuel Pool

Je Geon Bang, Tae Hyun Chun, Sun Ki Kim, Yang Hyun Koo

Korea Atomic Energy Research Institute, Daedeok-daero 989, Yuseong-gu, Daejeon, Korea, 305-353

*Corresponding author: jbbang@kaeri.re.kr

1. Introduction

Air oxidation behaviors of spent fuel in a spent fuel pool during a LOCA have been studied since a severe accident in Fukushima-Daiichi Nuclear complex. As seen in the accident in Fukushima-Daiichi Nuclear complex, it may be possible that exposure of spent fuel cladding to air during a hypothetical severe accidents such as loss of cooling and/or loss of coolant in the spent fuel pool. It is well known that air oxidation induces a serious degradation of the Zircaloy cladding material, compared with steam oxidation[1-3].

From the oxidant point of view, in comparison with steam, chemical heat release during oxidation in air is higher by 80%, which may lead to a more rapid degradation of the Zircaloy cladding, and further evolution of the accident[4-6]. Additionally, the oxidation kinetics in air is much faster than in steam due to the formation of non-protective oxide layer[1]. From the safety point of view, the barrier effect of the cladding against release of fission products is lost much earlier in air compared to steam.

The objective of this study is to investigate the oxidation behaviors of fuel cladding in two different conditions such as isothermal and transient condition and to generate its kinetic data under an accident condition in the spent fuel pool. In this study, the oxidation behaviors and its kinetics of the Zircaloy-4 were investigated in air environment for various air ingress scenarios in the temperature range 600 °C ~ 1,400 °C by thermo-gravimetric analysis.

2. Experimental

2.1. Specimens

2 cm long Zircaloy-4 tube segments (9.50 mm outer diameter, 0.570 mm wall thickness) were cut from longer tubes, deburred, ground at both ends, and cleaned in an ultrasonic bath of acetone.

2.2. Test procedure

For isothermal tests, the specimens were heated to the target temperature at a rate of 30 K/min in argon atmosphere after maintaining the chamber in vacuum condition with a rotary vacuum pump, and thermally equilibrated for 10 min. Then, the air was injected with a flow rate of 50 cc/min. The test was terminated by switching off the oxidizing gases(air) and cooling down the furnace as fast as possible with an argon atmosphere.

3. Results

Oxidation behaviors and rates of Zircaloy-4 in air were compared for both isothermal condition and transient condition at 1,200 °C and 1,400 °C. The result was shown in Fig.1. As shown in Fig.1, it was found that the rate of weight gain due to oxidation was decreases with time, and the weight gain behavior follows parabolic kinetic law (pink and red line).

For transient oxidation, pre-oxidation during heating the chamber occurs because the chamber would be heated in air environment without maintaining the chamber in vacuum condition. The oxidation rate in transient condition is slightly lower than the oxidation rate in isothermal condition (green and blue line).

In comparison with isothermal condition, the retardation of oxidation rate in transient condition was observed at both 1,200 °C and 1,400 °C. This seems to be ascribed to the effect of thin oxide formed during a heating.

It was also revealed that two lines with difference slope exist in Arrhenius plot. This means that two mechanisms are involved in the oxidation in air environment in the temperature ranges of this study (600 °C ~ 1,400 °C). Accordingly, two rate constants are needed in the given temperature ranges. In this study, two parabolic rate constants were withdrawn as follow (873K < T < 1673K):

$$k(\text{kg/m}^2\text{s}^{1/2})=97.51\exp(109,524/RT):(873\text{K}<T<1373\text{K})$$

$$k(\text{kg/m}^2\text{s}^{1/2})=0.69\exp(-46,365/RT):(1373<T<1673\text{K}).$$

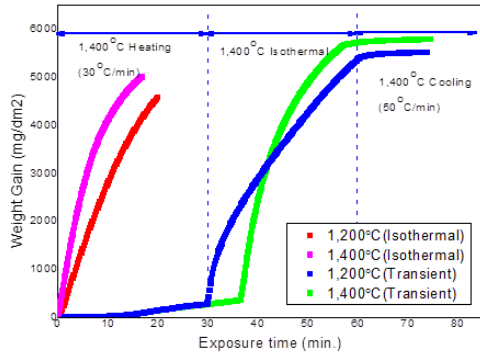


Fig. 1 Comparison of air oxidation rate between isothermal condition and transient

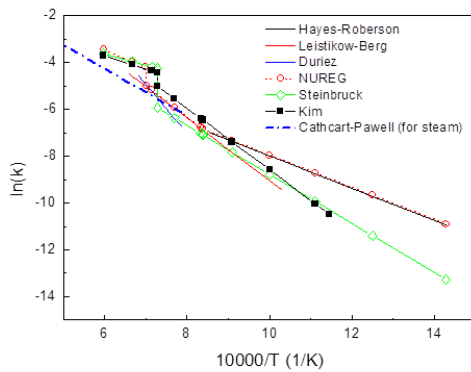


Fig. 2 Arrhenius plot for air oxidation rate of Zircaloy-4

4. Conclusions

In this study, the oxidation behaviors of the Zircaloy-4 for both isothermal condition and transient condition were investigated in air environment. In comparison with isothermal condition, the retardation of oxidation rate in transient condition was observed at both 1,200 °C and 1,400 °C. This seems to be ascribed to the effect of thin oxide formed during a heating.

From the kinetic study in various temperatures, the parabolic oxidation rate constants were withdrawn as follows:

$$k(\text{kg/m}^2\text{s}^{1/2})=97.51\exp(109,524/RT):(873\text{K}<T<1373\text{K})$$

$$k(\text{kg/m}^2\text{s}^{1/2})=0.69\exp(-46,365/RT):(1373<T<1673\text{K}).$$

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