Air Oxidation Behavior of Nuclear Fuel Cladding under Severe Accident Conditions in Spent Fuel Pools

Sun-Ki Kim[a], Seok Kim[b]

[a]*LWR Fuel Technology Research Division,* [b]*Reactor System Safety Research Division*
Korea Atomic Energy Research Institute
Daedeok-daero 989-111, Yuseong-gu, Daejeon, 34057, Korea
[a]*Corresponding author: kimsk@kaeri.re.kr*

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1. Introduction

Since the Fukushima Daiichi Nuclear Power Plant accident in 2011, the operational paradigm of nuclear power plants has shifted toward “Safety First.” Although severe accidents at nuclear power plants generally originate from accidents occurring within the reactor, severe accidents can also occur in wet spent fuel pools (SFPs). If the cooling function of a spent fuel pool is lost or damaged due to a large-scale earthquake, terrorism, or other external events, the spent fuel may become exposed above the coolant water surface. Under such conditions, the spent fuel cladding can undergo rapid oxidation with high-temperature air. The resulting embrittlement caused by oxidation, together with internal rod pressure, may lead to failure of the spent fuel cladding. Once the spent fuel cladding fails, the release of radioactive materials can be accelerated. Therefore, an accurate understanding of the oxidation behavior of the cladding under severe accident conditions is essential. Zircaloy-4 cladding has been widely used as a fuel cladding material in light water reactors for many years, and numerous studies have therefore been conducted on its oxidation behavior [1–5]. However, accurate information on the air oxidation behavior of Zircaloy-4 cladding remains limited, despite the fact that air oxidation can accelerate cladding degradation more rapidly than the steam oxidation that occurs during a loss-of-coolant accident in a reactor. Therefore, in this study, the air oxidation behavior and oxidation kinetics of Zircaloy-4 cladding under severe accident conditions in a spent fuel pool were evaluated.

2. Experiment

In this study, a thermogravimetric analyzer (TGA) was used to simulate the reaction of Zircaloy-4 cladding with air under spent fuel pool conditions following a loss-of-coolant accident and to measure the oxidation behavior in real time as a function of air exposure time. Air oxidation tests were conducted at temperatures ranging from 600°C to 1,400°C.

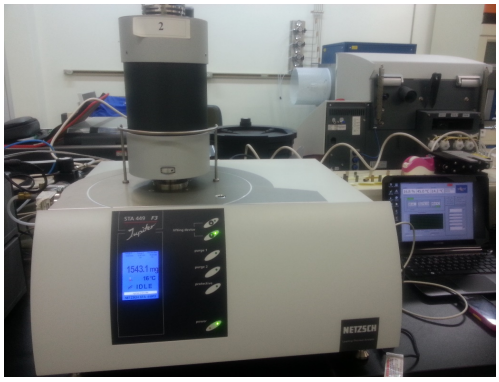


Figure 1. Thermogravimetric analyzer (TGA) for air oxidation

3. Results and Discussion

The results of the air oxidation experiments conducted under isothermal conditions are shown in figure 2.

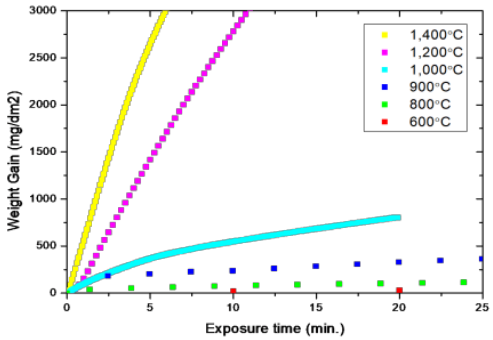


Figure 2. The results of the air oxidation experiments of Zircaloy-4

As shown in Figure 2, the changes during the initial oxidation period were analyzed to investigate the initial oxidation behavior over the total oxidation time. The results showed that the oxidation rate gradually decreased as the accident progression time increased. The initial air oxidation behavior at high temperatures was found to follow a parabolic kinetic law, with the

oxidation rate exponent being approximately 2. In other words, under spent fuel pool accident conditions, when Zircaloy-4 cladding is exposed to air, the rate of weight gain due to the oxidation reaction gradually decreases with increasing exposure time.

Meanwhile, photographs of the specimens after air oxidation at each test temperature are presented in Figure 3. As the extent of oxidation increased, the original metallic luster of the cladding gradually disappeared, and the surface color changed to gray and flesh-colored. With further increases in the extent of oxidation, the surface eventually changed to white.



Figure 3. Appearance of the Zircaloy-4 cladding specimen after oxidation

Furthermore, based on the air oxidation behavior observed at various temperatures in this study, the oxidation rate at each temperature was evaluated, and the results were compared with those of previous studies in Figure 4. The results confirmed that the oxidation reaction rate under air was higher than that under steam oxidation. Based on these results, kinetic model equations were developed for two distinct temperature regions to describe the oxidation behavior of Zircaloy-4 cladding.

$$k=97.51\exp(-109{,}524/RT)\ (873K<T<1373K)\ (kg/m^{2s1/2})$$

$$k=0.69\exp(-46{,}365/RT)\ (1373<T<1673K)\ (kg/m^{2s1/2})$$

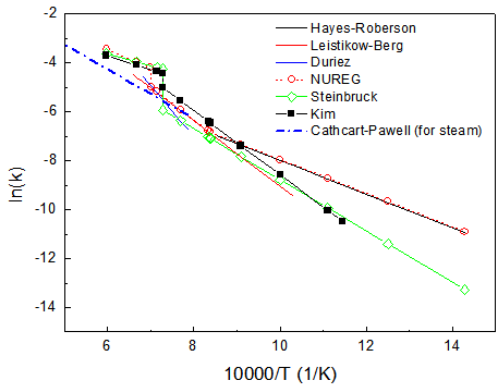


Figure 4. Oxidation reaction rates at various temperatures

4. Conclusions

This study evaluated the high-temperature air oxidation behavior of Zircaloy-4 cladding under temperatures ranging from 600 to 1,400°C, considering a loss-of-cooling accident in an SFP. The results showed that the oxidation rate gradually decreased as the oxidation time increased. In particular, the initial oxidation behavior followed a parabolic kinetic law, with an oxidation rate exponent of approximately 2. This behavior is considered to be associated with the formation of an oxide layer, which gradually limits further oxidation as oxidation progresses.

In addition, as the extent of oxidation increased, the metallic luster of the cladding surface disappeared and the color changed from gray and flesh-colored to white. Comparison of the oxidation rates with those reported in previous studies showed that air oxidation proceeded faster than steam oxidation. Based on the measured oxidation behavior, kinetic model equations for Zircaloy-4 oxidation were developed for two distinct temperature regions. These results can provide fundamental data for evaluating the oxidation and degradation behavior of Zircaloy-4 cladding and the potential release of radioactive materials during severe accidents in spent fuel pools.

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