High-temperature Breakaway Oxidation of Fuel Claddings in LOCA Temperature Range

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1. Introduction

The structural integrity of a fuel cladding under the normal and abnormal operation conditions should be considered to secure its safety in the design of a light water reactor. During a typical LOCA condition, the cladding is subjected to a high temperature oxidation which is finally quenched because of an emergency coolant reflooding into the core. In this LOCA situation, the equivalent cladding reacted (ECR) should not exceed the criterion of 17% and the peak cladding temperature (PCT) should also be below 1200^{o}C [1]. The susceptibility to a high-temperature breakaway oxidation is reported the key to understand the contrasting performance of the Russian alloys (E110, E635) and the Western alloys (M5, ZIRLO) [2]. It was found that the premature breakaway of the Russian alloys could have resulted from an excessive hydrogen pickup due to different fabrication routes from the raw materials to the final tube [2]. So, the validity of the breakaway oxidation at LOCA-like high temperatures remains to be confirmed by the symmetrical studies. In this study, the high-temperature breakaway oxidation phenomenon of the advanced Zr alloys was investigated.

2. Experimental

The test samples used in this study were HANA claddings including two reference claddings (Zircaloy-4 and A). As mentioned in the previous paper [3], the apparatus for the high-temperature testing was established by modifying the Shimadzu TGA (Thermo-gravimetric Analyzer, TGA 51H). The heating rate up to the desired temperature was set at 50^{o}C/min. The specimens were steam-oxidized at the temperatures of 950 to 1200^{o}C for 10 to 18000 s. After an oxidation testing at the desired temperatures for the exposure time, the oxidation rate constant was calculated in accordance with the parabolic rate law. Besides, the hydrogen content, pre-to-post transition time and microstructures were also determined for the oxidized specimens.

3. Results and Discussion

3.1 Breakaway Oxidation

In this study, the high-temperature oxidation tests were performed on the KAERI developed Zr alloys (hereinafter, which are called HANA^{TM}) and two reference claddings. The weight gain kinetics of all claddings followed a parabolic rate law when they were exposured less than 2000s at 1000^{o}C.

When the exposure time was prolonged for more than 3000s, as shown in Fig. 1, Zircaloy-4, A and HANA-4 claddings showed an oxidation kinetics transition. This transition is known to be caused by the 'breakaway oxidation' at high temperatures of the LOCA range. The oxidation kinetics at the breakaway was changed from a parabolic rate to a quasi- parabolic rate.

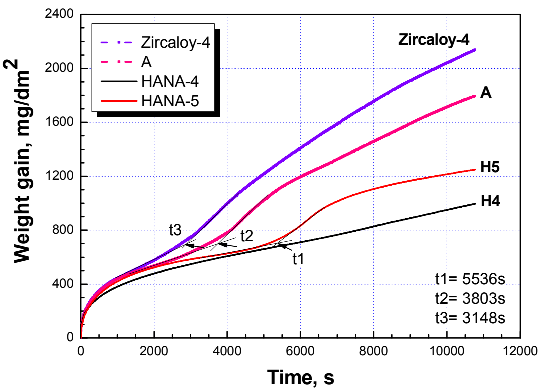

Fig. 1 Oxidation behaviors of Zr Claddings at 1000^{o}C

From our tests, the breakaway phenomenon was detected in some claddings exposured for more than ~3000s. Specifically, the breakaway time (pre-to-post transition time) of the Zircaloy-4 cladding was 3148s, the A cladding was 3803s, and the HANA-5 cladding was 5536s. One can easily assume that the breakaway time of the claddings could be related to the Sn contents within the claddings. As the Sn content within the claddings increased, the breakaway time increased. But the HANA-4 cladding containing 0.4% of Sn did not reveal the breakaway oxidation phenomenon up to the exposure time of 10800s. It was thought the critical Sn content could affect the breakaway oxidation of the Zr claddings.

3.2 Oxidation Rate Constant

As mentioned above, the kinetics of all the tested claddings was controlled by the parabolic rate and quasi-parabolic rate at the pre-breakaway and post-breakaway regimes, respectively. The parabolic rare constant could be calculated from the relationship between the weight gains and the oxidation time at an oxidation temperature. The rate constants of the HANA-4 and HANA-5 claddings at the pre-breakaway regime were lower than those of the reference claddings. The HANA-5 cladding in the post-breakaway regime also showed a lower rate constant than the reference claddings. The rate constants of the reference claddings in the post-breakaway regime were similar to the values by both Cathcart-Pawel and Baker-Just equations [4,5].

After the oxidation tests at 950~1200^{o}C for 18000 s, the oxidation rate constants of the claddings were calculated separately for two pre-breakaway and post-breakaway regimes. While the rate constants from the pre-breakaway regimes lay on a straight line in the rate constant versus reciprocal temperature plot, those from the post-breakaway regimes were on another straight line in same type plot. This means that the oxidation kinetics is different for the pre-breakaway and the post-breakaway regimes. And the line obtained from the post-breakaway regimes was plotted on a more part than that from the pre-breakaway regimes. And the rate constants from the post-breakaway regimes also well matched with the values by both the Cathcart-Pawel and Baker-Just equations.

Fig. 2 shows an example of the oxidation rate constants calculated separately for two pre-breakaway and post-breakaway regimes on the Zircaloy-4 cladding. As noted already, the rate constants from the post-breakaway regimes also well matched with the values by both the Cathcart-Pawel and Baker-Just equations. And the rate constants from the pre-breakaway regimes were also plotted on one line in spite of a small deviation. Compared with the rate constants at the post-breakaway regimes, the rate constants of the HANA-4 and HANA-5 claddings were lower than those of the reference claddings (Zircaloy-4 and A). Therefore, the oxidation resistance of the HANA claddings was superior to the commercial claddings at the LOCA temperatures and the susceptibility of the HANA claddings was less sensitive than the Zircaloy-4 and A claddings.

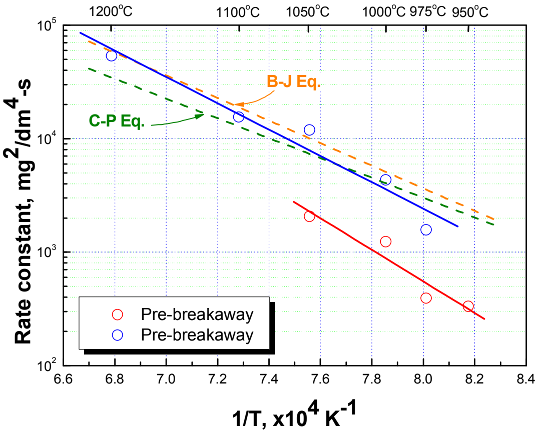

Fig. 2 Oxidation rate constant of Zircaloy-4

4. Conclusions

1. The breakaway time (pre-to-post transition time) of Zircaloy-4 cladding was 3148s, the A cladding was 3803s, and the HANA-5 cladding was 5536s.
2. The breakaway time of the Zr claddings could be related to the Sn contents within the claddings.
3. The rate constants from the post-breakaway regimes were well matched with the values by both the Cathcart-Pawel and Baker-Just equations
4. The oxidation resistance of the HANA claddings was superior to the reference claddings at the LOCA temperatures and the susceptibility of the HANA claddings was less sensitive than the Zircaloy-4 and A claddings.

ACKNOWLEDGEMENT

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