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Investigation of steam velocity effect on cladding oxidation phenomena

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1. Introduction

Ensuring safety is paramount when it comes to utilizing nuclear power plants. Regardless of how promising an energy source may be, if it isn't used safely, it could lead to disastrous consequences. Consequently, extensive safety research is conducted within nuclear power plants, with the aim of preventing the release of radioactive materials to the public. This is achieved through the implementation of multiple barriers, employing the Defense-in-Depth (DiD) concept. Despite these efforts, core melting severe accidents such as TMI and Chernobyl accident had been experienced, and the demand for nuclear safety has increased each time [1]. In addition, the accident that occurred in Fukushima in 2011 reminded us that severe accidents might be occurred in our surroundings. As a result, research on various phenomena threatening the multiple barriers which prevent release of radioactive materials to the public in the severe accident had been accelerated, and global research cooperation had been conducted through programs such as SARNET [2].

Among the various phenomena, nuclear fuel cladding oxidation is considered significant because it can damage the fuel cladding, which serves as a physical barrier to prevent the release of radioactive substances. To improve safety of the physical barrier, numerous separate effect tests (SET) and integral effect tests (IET) such as LOFT-FP, Quench, and Phebus [3-10] for nuclear fuel cladding oxidation phenomenon had been performed. All research about cladding oxidation cannot be treated in this paper because tremendous number of studies have been conducted on various cladding materials (Zircaloy, DX-D4, M5, E110, ZIRLO, ATFs, etc.), various oxidants (steam, air, nitrogen, oxygen, etc.), and wide range of temperature and pressure. However, if we focus on the development of the oxidation model, the various models had been developed for the steam oxidation phenomenon of zircaloy cladding. And the existing oxidation models are only a function of temperature. Since the oxidation is a chemical reaction, it is reasonable to have a dominant effect on temperature according to Arrhenius eq., and it has been clearly revealed through studies so far.

However, as the oxidation reaction occurs, the amount of steam decreases and the fraction of hydrogen gas will be increased. As a result, the density of the oxidant, in this case steam, changes, which will cause changes in the Re and Nu number. Therefore, it is also clear that the steam velocity affects the cladding oxidation phenomenon. In other words, there may be a situation in which heat generation due to oxidation is dominant and another situation in which the convective cooling effect due to fast steam velocity is dominant. Notwithstanding, steam velocity effect on the cladding oxidation phenomenon is not investigated quantitatively until now, it is not reflected in the existing oxidation models. The problem is that if the steam convection cooling effect is much larger than expectation, the safety analysis using the current models may contain significant uncertainties. Therefore, an experimental apparatus for research the steam velocity effect on the cladding oxidation had designed and developed, and experimental methodology had established in this paper. And some isothermal steam oxidation experiments at 900°C were carried out and compared with existing model to show the steam velocity effect.

2. Methodology for High-steam Velocity Oxidation Experiment

In this study, an experimental apparatus capable for observing the effect of steam velocity on fuel cladding oxidation was constructed. The designing an experimental apparatus was reflecting the limitation of TGA experimental facility to observe such phenomena. The first parameter is the range of steam velocity being applied. Core steam velocity in large LOCA condition is approximately between 0 m/sec and 20 m/sec based on the SPACE code simulation. To realize actual steam velocity condition in large LOCA, this experimental apparatus was designed to supply steam into a test section at maximum 20 m/sec. This steam velocity was realized by connecting two metering pumps in parallel.

The second parameter is the cladding temperature. In TGA, the cladding surface temperature cannot be measured because the cladding specimen is connected to a very sensitive balance. However, cladding temperature, especially PCT (Peak Cladding Temperature), is a very important variable in the safety study of nuclear power plants. And since it is a variable directly affected by oxidation reaction, the cladding temperature must also be measured in the new experimental apparatus. Therefore, the test section was designed that thermocouples are mounted on inner and outer wall surface of cladding specimen. And real time mass gain was designed to measure by real time hydrogen concentration measurement. The most important design requirements of this experimental apparatus are related to above parameters, i.e., high steam velocity and cladding wall surface temperature measurement. Schematic drawing of the experimental apparatus is shown in Figure 1.

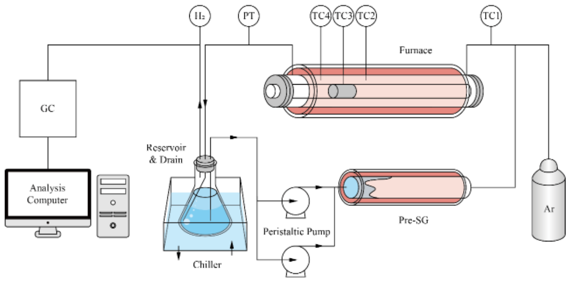

Figure 1. Schematic drawing of experimental apparatus for steam velocity effect of the cladding oxidation phenomenon

2.1 Water supply pumps

As mentioned above, the most important design requirement in this experimental apparatus is the steam flow rate reflecting the actual LOCA condition. To realize this, two peristaltic pumps were designed to supply water by connecting them in parallel, and performance evaluation was performed. The peristaltic pump used in this experiment is Longer Pump's WT3000-1JA. The performance curve obtained through performance test is shown in Figure 2. In this paper, isothermal experiment with 900℃ steams will be conducted, so it can be confirmed that the maximum steam velocity is approximately 15 m/sec.

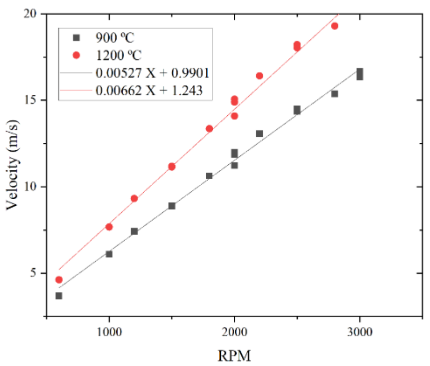

Figure 3. Water supply system performance test result

2.2 Test section

An isothermal oxidation experiments at 900℃ will be conducted in this study. However, the test section was made of high temperature material (alumina) since much higher temperature condition should be applied in further study. The alumina test section design can be seen in Figure 3. In this experimental apparatus, the test section consists of external and internal test sections. The inner test section is an alumina tube with the same outer diameter as the cladding specimen and serves as a fuel rod in this experiment. The inner diameter of the outer test section was determined to have the same flow area as the flow area considering the pitch of the APR1400 fuel assembly. In the outer test section, thermocouple insertion tubes were connected to measure the steam temperature up and downstream of the cladding specimen and the temperature inside and outside wall of the cladding specimen.

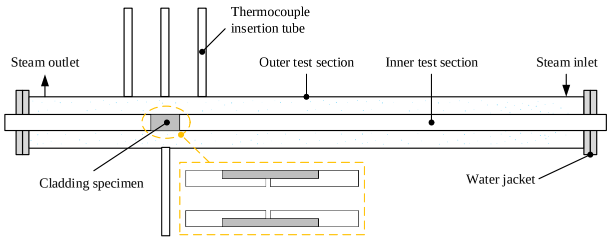

Figure 3. Test section drawing

Since the saturated steam injected into the test section and must be heated to the target temperature (900℃) within the test section, the cladding specimen should be located at the rear part of the test section. The length of heating part of the test section is 700 mm, and the cladding specimen is located 550 mm from the steam inlet. Water jackets were designed at both ends of the test section so that the inner test section could be located concentrically with the outer test section. The water jacket not only fixes the position of the inner test section and the outer test section, but also provides a seal to prevent the inflow of high-temperature steam from leaking out of the test section. The material of the cladding specimen installed inside the test section is Zircaloy-4. In addition, the inner and outer diameters of the cladding specimen are 8.25 mm and 9.5 mm, respectively, which are the same as those of actual nuclear fuel cladding. Alumina inner test section has same outer diameter to Zircaloy-4 specimen. And alumina inner test section and specimen is connected like orange box in Figure 4 to maintain fuel rod shape.

2.3 Test Procedure and Matrix

The test procedure can be separated into three sections namely preparation stage, reaction stage, and shutdown stage. The preparation stage was designed to heat the furnace at target temperature before the steam supply while ensuring there was no oxidation reaction.

To achieve this objective, argon gas was supplied at 5ml/min to create an inert environment in the test section while the furnace was heated up. To heat up the alumina test section safely until designated temperature, heating time was controlled less than 20 °C/min of heating rate. This is to protect the integrity of alumina test section from too high thermal stress.

After the furnace showed a stable temperature at target value, the reaction stage was started by opening the steam supply valve and closing the argon gas injection valve simultaneously. A sudden pressure drop detected by pressure transmitter at this stage conversion. This difference in pressure will ensure an accurate timing of the oxidation reaction starting. After certain times, the reaction stage was ended by closing the steam supply valve and opening back the argon gas injection valve. Argon gas flowing can ensure no additional oxidation reaction after the reaction stage.

An instant pressure increasing was detected at this stage conversion which was useful for marking the end of reaction stage. The furnace was shut down under the appropriate cooling rate to protect alumina test section. And the argon was kept flowing as the cooling process of the furnace took place. After the cooling down of the furnace is over, the argon flow was stopped, and furnace was disassembled to take out the test specimen. The specimen then measured in a weight scale (OHAUS, PAG214) to obtained final mass gain. The above test procedure can be expressed as Figure 3.

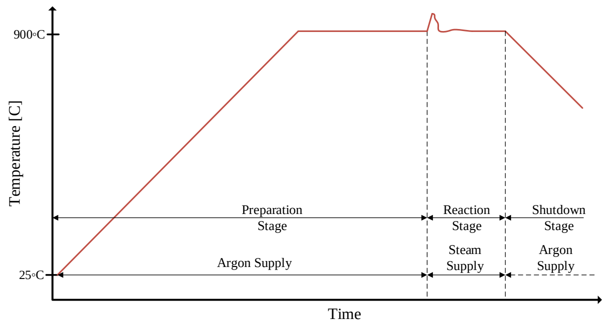

Figure 3. Test Procedure of the steam oxidation experiments at 900℃

Table 1. Test matrix for steam oxidation experiments at 900℃

Test No.	Steam velocity (m/sec)	Re (-)	The number of tests
T900-v01	1.02	43.9	1
T900-v02	2.6	110.1	1
T900-v03	8.7	377.2	1
T900-v04	16.5	702.8	1

The effect of steam velocity was measured through experiments on nine different flow rates as the main variable. Velocity and Reynolds number of steam velocity were calculated based on steam flow rate and temperature. The test matrix in this experimental study is shown in Table 1. According to the LOCA analysis results of APR1400, the maximum and average steam velocity were approximately 20 m/sec and 10 m/sec, respectively. Therefore, this experiment was designed to supply steam at a velocity of up to 16.5 m/sec, and the Re number is in the laminar flow region.

3. Experimental Results and Discussion

This chapter presents the results of a high-temperature steam oxidation experiment. Through experiments, it is confirmed the behavior of PCT and oxidation coefficient according to changes in steam velocity. And compared them with existing oxidation models to show the steam convection effect on the cladding oxidation phenomenon.

3.1 PCT (Peak Cladding Temperature) Behavior

To confirm the effect of steam convection on PCT behavior, the change in PCT according to the various Re number is shown as shown in Figure 4. Even if it was a 900℃ experiment, it is hard to control the initial temperature accurately at 900℃ in all experiments depending on the performance of the temperature controller and environmental conditions. Therefore, the difference between the initial temperature and the maximum temperature during each experiment was expressed in the y-axis of Figure 4.

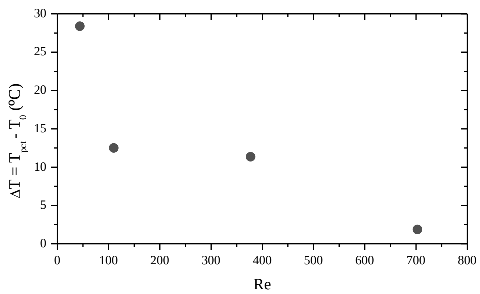

Figure 4. Maximum Cladding temperature difference behavior varying Reynolds number

Looking at the PCT behavior of the high-temperature steam convection oxidation experiment at 900℃, there is a clear tendency to decrease as the Re number increases.

In this experiment, the temperature of the cladding outer surface is measured. Accurate PCT measurement is possible when the thermocouple installed to measure the cladding temperature is in stable contact with the surface of the cladding during the experiment. It can be clearly seen that the temperature increase due to oxidation heat is decreased by steam convection effect. As the steam flow rate increases, the heat discharged to channel due to the oxidation reaction increases, but at the same time, the cooling effect due to convection increases. So, it is predicted that the temperature behavior of the cladding surface will be determined by the extent of heating and cooling phenomenon. According to Figure 8, isothermal condition at the 900℃, it is estimated that the convective cooling effect due to the steam velocity is larger than the heat generation due to the oxidation reaction. And this physical phenomenon is clearly shown in Figure 4.

3.2 Oxidation Coefficient Behavior

It was confirmed above that as the steam velocity increases, the cladding surface temperature decreases due to the steam convective cooling effect, the oxidation reaction decreases, and the resulting mass gain also decreases. Therefore, the oxidation coefficient (K_p) used in the oxidation model is also expected to decrease. Figure 5 shows the oxidation coefficient results from the high-temperature steam oxidation experiment.

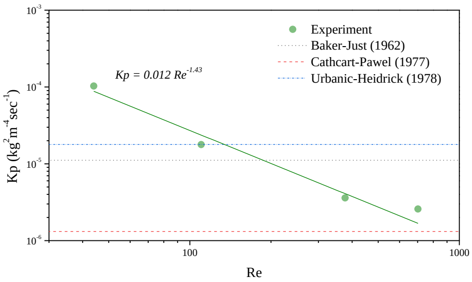

Figure 5. Oxidation coefficient behavior varying Reynolds number

It clearly shows a tendency for the oxidation coefficient to decrease as the Re number increases. All existing steam oxidation models are expressed as a function of only temperature. However, according to Figure 5, the steam velocity effect is clearly observed. Under 900℃ temperature conditions, the oxidation coefficient of the Baker-Just model is 1.111×10^{-5}, the oxidation coefficient of the Cathcart-Pawel model is 1.312×10^{-6}, and the oxidation coefficient of the Urbanic-Heidrick model is 1.786×10^{-5}. Existing oxidation models do not reflect the effect of steam velocity and are represented as a straight flat line in Figure 5 because it is only a function of temperature. On the other hand, the oxidation coefficient observed in this experimental study can be confirmed to vary depending on the Re number at a similar range to the existing model. Experimental data (green circle dot in Figure 5) were collected from a total of 4 Re numbers, and fitting line and formulas were derived using the fitting method. The derived correlation is presented in the equation (1) below.

$$K_p = 0.012\, Re^{-1.43} \qquad (1)$$

Based on the graph shown in figure 5, the equation (1) will help to predict more accurate value of cladding temperature and oxidation coefficient (K_p) by considering steam Reynolds number. However, this experimental study was done strictly only at 900℃, so the correlation can only be applied at particular temperature. And more data point is also required for more accurate results. This result, however, shows Reynolds number as an important parameter for predicting cladding temperature and oxidation coefficient in steam-zirconium oxidation which has not been studied before. Furthermore, this results also suggest the necessity of further study to develop new correlation for steam oxidation rate which take into account both temperature and steam Reynolds number.

4. Conclusion

In this study, an experimental investigation was conducted to identify the effect of steam convection on nuclear fuel cladding oxidation phenomenon. Existing oxidation models are only a function of temperature, but it was expected that the effect of steam velocity would also have a significant impact. However, in typical oxidation studies using TGA, the high steam velocity affects the mass measurements of the very sensitive balance, so the typical experimental methodology using TGA cannot be used. Therefore, through this study, an experimental apparatus and procedure were developed to perform a high-velocity, high-temperature steam oxidation experiment. Additionally, as a result of conducting the experiments using this experimental apparatus, the following findings were made.

- It was discovered that steam velocity effect is clearly exists.
- It was confirmed that there is also an effect of slowing

down the oxidation phenomenon due to the steam velocity increasing.

- It was experimentally shown that the oxidation coefficient decreased as the Re number of steam increased under 900°C conditions, and a correlation was also developed.

Through this study, it was confirmed that existing oxidation models should be improved for more accurate analysis and should be expressed as a function of steam velocity as well as temperature. Although this study is the first to clearly demonstrate experimentally the effect of steam convective cooling on cladding oxidation phenomenon, it is still an ongoing study and additional experimental research should be conducted further.

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