

Article

The Limitations of an Air-Oxidation Breakaway Model to Predict a Zirconium Fire in a Spent Nuclear Fuel Pool Accident

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Abstract: The OECD/NEA Spent Fuel Pool (SFP) project was conducted to investigate consequences of spent nuclear fuel pool accident scenarios. From the project, it was observed that cladding temperature could abruptly increase at a certain point and the cladding was completely oxidized. This phenomenon was called a “zirconium fire”. This zirconium fire is one of the crucial concerns for spent fuel pool safety under a postulated loss of coolant accident scenario, since it would lead to an uncontrolled mass release of fission products into the environment. To capture this critical phenomenon, an air-oxidation breakaway model has been implemented in the MELCOR code. This study examines this air-oxidation breakaway model by comparing the SFP project test data with a series of MELCOR code sensitivity calculation results. The air-oxidation model parameters are slightly altered to investigate their sensitivities on the occurrence of the zirconium fire. Through such sensitivity analysis, limitations of the air-oxidation breakaway model are identified, and needs for model improvement is recommended.

Keywords: zirconium fire; air-oxidation; breakaway; spent fuel pool; loss of coolant accident

1. Introduction

Recently a major experimental program to investigate consequences of a complete loss of coolant accident (LOCA) in spent nuclear fuel pools was conducted from 2009 to 2013 in the frame of the OECD/NEA Spent Fuel Pool (SFP) project [1,2]. Three years after the completion of the SFP project, the experimental data were made public and hence they become available for this study. The SFP experiments were performed using a 17×17 pressurized water reactor (PWR) fuel assembly in an air atmosphere. The phase-1 test was conducted in a 1×1 configuration of fuel assembly, and the phase-2 test was performed in a 1×4 configuration. During both the phase-1 and -2 experiments, zirconium cladding was oxidized by air and the cladding surface temperature was measured by two thermocouples. In addition, a video was recorded in real time to observe phenomena. It was observed that the zirconium cladding surface temperature suddenly increased strongly and a flame clearly appeared. This phenomenon of abrupt zirconium cladding temperature escalation was named a “zirconium fire”. After the onset of the zirconium fire, the whole fuel assembly was completely oxidized and broken into fine powder. The pre- and post-test photographs of the fuel assembly from the phase-1 experiment are illustrated in Figure 1. The post-test fuel assembly was severely degraded after the zirconium fire. After experiencing the zirconium fire, the whole fuel assembly was completely broken down into powder. It is noted that this observed zirconium fire phenomenon would occur at elevated temperatures when the coolant in the spent fuel pool is sufficiently drained out as assumed in the OECD/NEA SFP project.

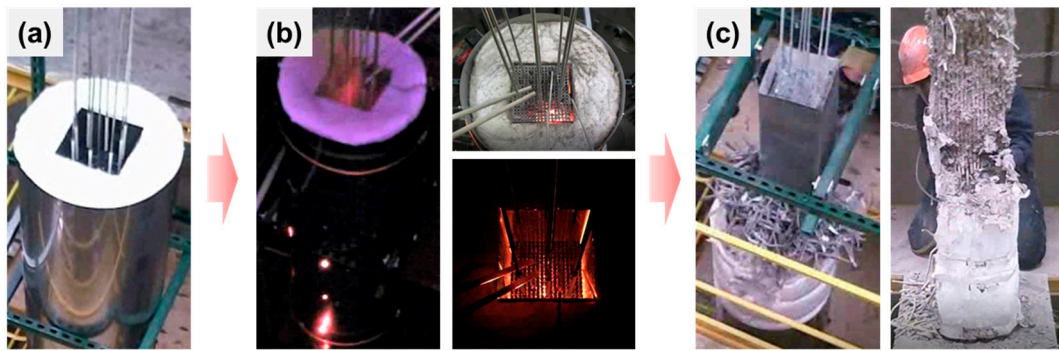


Figure 1. Pre- and post-test photographs of fuel assembly of the Spent Fuel Pool (SFP) project phase-1 test: (a) pre-test fuel assembly, (b) zirconium fire phenomenon, and (c) post-test fuel assembly (taken from the SFP project phase-1 test data [1]).

During the SFP project phase-1 test, however, the oxygen concentration drastically decreased at the initiation of zirconium fire. As shown in Figure 2, as oxygen is consumed dramatically, the cladding temperature increased very sharply up to almost 1850 K from around 1100 K within less than 10 min. After that time, the thermocouples failed, so temperature records became scattered. Interesting behavior was seen concerning the oxygen concentration profile. Firstly, it was consumed significantly, but after the cladding temperature reached the maximum, the oxygen concentration increased to 21%, then decreased again. It seems that the zirconium alloy cladding was fully oxidized by oxygen within a very short period. After the zirconium alloy cladding was fully oxidized, the oxygen was no longer consumed by the cladding. For this reason, integrity of cladding was completely lost, and it no longer functioned as a safety barrier against the fission products liberated from the fuel. Therefore, the possible consequence of zirconium fire would be an uncontrolled mass release of source term to the spent fuel pool building and even to the environment. The severe risk of a zirconium fire to the environment was raised by Hippel and Schoeppner [3–5]. In addition, a number of spent fuel pool accident analyses were performed by several safety analyses codes such as MELCOR, MAAP, ASTEC, ATHLET-CD, ICARE/CATHARE, RELAP/SCDAPSIM, MAAP, and SPECTRA [6–12]. More recently, spent fuel pool accident scenarios were analyzed with most of the other mentioned safety analyses codes, and the trends of cladding temperature escalation due to the zirconium fire were not comparable with the various code analyses [12].

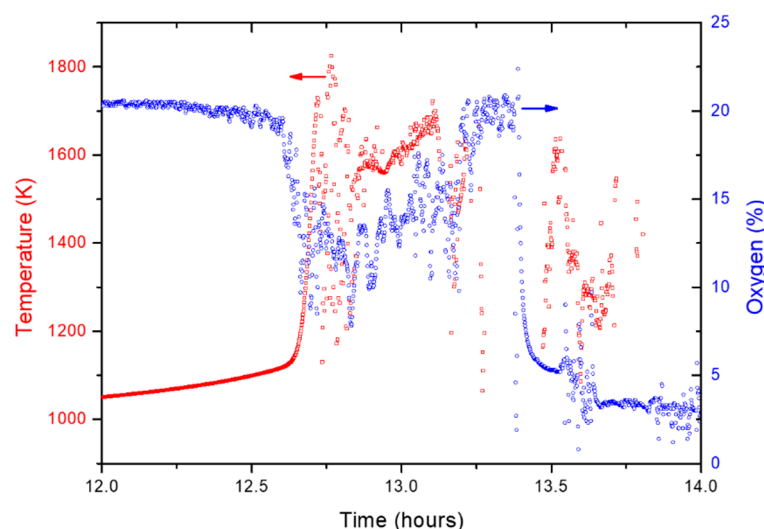


Figure 2. Comparison of fuel assembly temperature (red) and oxygen concentration (blue) in the SFP project phase-1 experiment.

Several institutes also participated in the benchmark of zirconium fire that was observed in the SFP phase-1 experiments using the MELCOR code, and they captured the cladding temperature escalation in the zirconium fire relatively well [13]. However, no discussion was made on the characteristics of the model which best captured the zirconium fire. In this paper, we carefully assess the model in the MELCOR code which can calculate the zirconium fire in the case of a spent fuel pool accident. A detailed review of the model is provided in Section 2 and the series of sensitivity analyses are described in Section 3.

2. Review of the Air-Oxidation Breakaway Model in the MELCOR Code

In order to capture the zirconium fire phenomenon accompanied by very severe zirconium heat-up and simultaneous dramatic oxygen consumption, an air-oxidation breakaway model was developed in the MELCOR code [1,2,14]. The breakaway phenomenon refers to the kinetic transition from parabolic kinetic rate law (pre-breakaway kinetics) to a linear kinetics rate law (post-breakaway kinetics). At the onset of breakaway, the air would be consumed by the zirconium alloy cladding significantly higher in comparison with the air consumption before the breakaway. Due to this sudden severe oxidation, zirconium ignition would occur. Therefore, the current model assumes air-oxidation breakaway as the direct cause of the zirconium ignition phenomenon.

During air-oxidation, the breakaway (i.e., kinetic transition) is initiated when the value of “lifetime” becomes 1. The lifetime function (LF) is given in the following

$$LF(T) = \frac{\int_0^t dt'}{\tau(T)} \quad (1)$$

$$\tau(T) = 10^{-12.528 \log_{10} T + 42.038} = T^{-12.528} \cdot 10^{42.038} \quad (2)$$

where T is the cladding temperature in K and $\tau(T)$ is the breakaway transition time in seconds. As shown in Equations (1) and (2), the breakaway transition is only dependent on the cladding temperature. At a given cladding temperature, the transition time is given by Equation (2) and the breakaway is initiated at this transition time. The model parameters (−12.528, 42.038) in equation (2) were determined by fitting to Argonne National Laboratory (ANL) Zry-4 air-oxidation test data at 773–1173 K with only 8 data points [15].

The breakaway data at 773–1173 K (time of the kinetic transition) were plotted against temperature using a log–linear scale. However, as shown in Figure 3, the fitting of these data was performed using a log–log scale. The model parameters that were determined from the log–log data fitting are likely to include unexpected large errors even within their confidence intervals. This unexpected large error may significantly affect the uncertainties in the calculated sequence progression in the SFP accident scenarios. In addition, using this method of empirical model fitting may have several limitations, as follows:

- ANL air-oxidation test data showed no “zirconium fire phenomenon” during air-oxidation tests at 773–1173 K [15]. In other words, air-oxidation breakaway did not trigger a zirconium fire. It seems a strong assumption that the air-oxidation breakaway is the direct cause of the zirconium fire. However, only separate-effect tests with very small Zry-4 samples were performed in the ANL air-oxidation tests. For this reason, it seems that an abrupt high heat release in the fuel assembly scale might induce the zirconium ignition in the integral effect tests like the SFP project experiments [1,2,14].
- The relationship between air-oxidation breakaway time and temperature was considered as linearly correlated on a log–log scale. However, no sound physical meaning was supported by this model fitting. Only 8 data points were fitted, and also each data point was scattered to some extent.

The fitted breakaway time is given as a function of temperature with the exponent coefficient as follows: Time to breakaway (T) = $T^{-12.528} \cdot 10^{42.038}$. If the model parameters are slightly altered, the calculated breakaway timing will be significantly shifted. Let one model parameter (−12.528) be called A_0 and the other (42.038) be called B_0 . For example, at a fixed temperature of 1100 K with these A_0 and B_0 , the time to breakaway is 8619 sec. If the value of A_0 is altered from −1% to 1% with the fixed B_0 at the fixed temperature of 1100 K, the time to breakaway changes from 3591 to 20,683 s. The variation in the breakaway time is −58% to 140% when the value of model parameter A_0 varies from −1% to 1%. The model parameter A_0 was from the fitting of the experimental data, and this fitted value includes its own error from the curve fitting. In this respect, the model concept (i.e., curve-fitting model in log–log scale) introduces a high uncertainty to the prediction of the target value. Another example is the variation of the other parameter B_0 from −1 to 1% with the fixed A_0 at the fixed temperature of 1100 K. In this case, the variation of the breakaway time is 3277 to 22,669 s and its percentage is −62% to 163%. From these simple calculations at a fixed cladding temperature with a very small variation of model parameters, the predictions of air-oxidation breakaway were varied. However, the actual cladding temperature in a postulated SFP complete LOCA is not fixed but varied according to the accident progression.

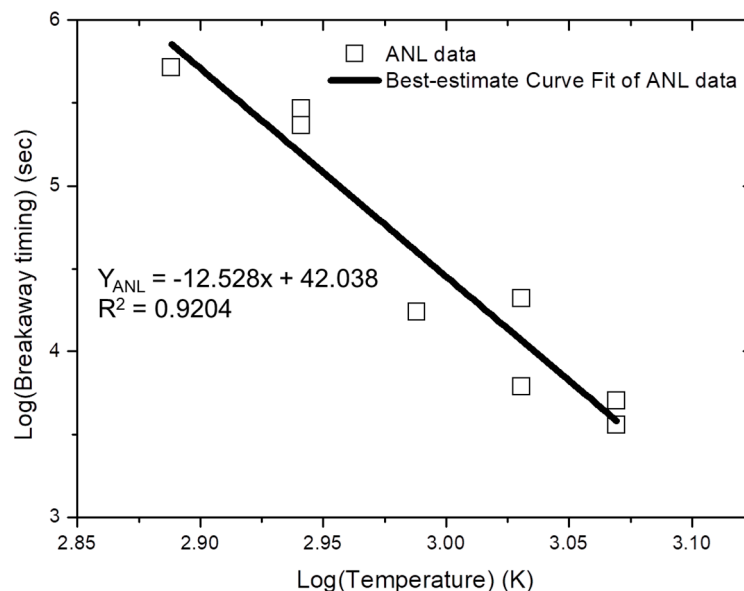


Figure 3. Air-oxidation breakaway model parameter fitting (data were taken from [15]).

This study examines this air-oxidation breakaway model by comparing the SFP project test data and the MELCOR code calculation results by using this model. This study also aims to reveal the weaknesses of the model, and to propose model improvements to overcome the limitations identified. As previously mentioned, the current model was an empirical model with limited experimental data fitting. Furthermore, the formula of the empirical model was based on log–log model fitting of air-oxidation breakaway time and temperature data. The model parameters that were determined from the log–log data fitting are likely to include an unexpectedly large error even within their confidence intervals. This unexpected large error may significantly affect the uncertainties when estimating the sequence progression in the SFP accident scenarios. The air-oxidation model parameters are slightly altered to examine their sensitivities to the occurrence of zirconium ignition. Before performing this sensitivity analysis, the MELCOR code input model to simulate the SFP complete LOCA was prepared as described in the following section and is assessed using the SFP project phase-1 experimental data.

3. Sensitivity Analysis on the Occurrence of Zirconium Fire in the SFP Complete LOCA

Firstly, the MELCOR input model of SFP was developed to simulate the single fuel assembly that was used in the SFP project phase-1 experiments. The single fuel assembly contained a 17×17 PWR fuel bundle ~ 4 m in height; the detailed input model description is given in [1]. The peak cladding temperature from the calculation using the developed input and the corresponding SFP experimental data are given in Figure 4.

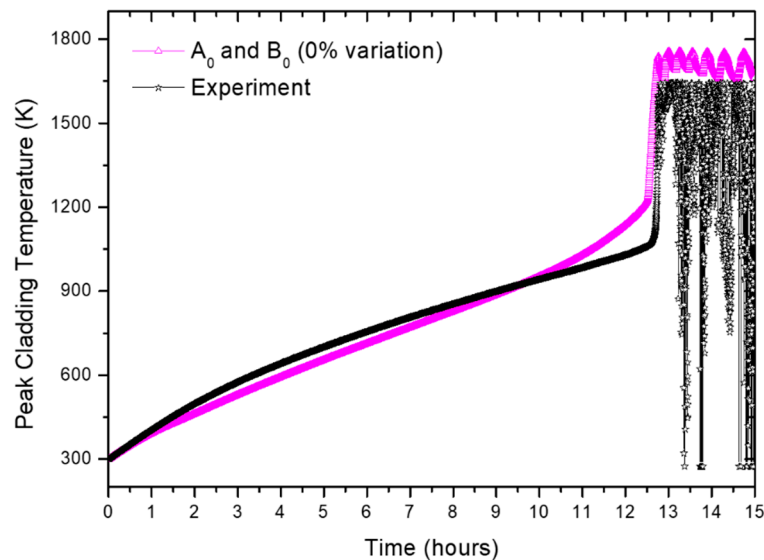


Figure 4. Peak cladding temperatures, code calculation and experiment for SFP project phase-1.

As shown in Figure 4, the time of the zirconium fire (i.e., the time immediately after the breakaway) calculated using the MELCOR model input is very comparable with the experimental data. The onset of the zirconium fire in the SFP project experiment was reported at 12.66 h and the cladding temperature increase rate at that time was ~ 490 K/h. In the code calculation, the time to ignition was 12.50 h, with an increase rate of ~ 490 K/h in cladding temperature. Based on the developed MELCOR input, various calculations were performed to investigate the sensitivity of model parameters on the time to ignition in the postulated SFP LOCA. In this paper, the criterion to determine zirconium ignition is an abrupt cladding temperature rise with increase rate of ~ 490 K/h. In this section, a systematic sensitivity calculation is performed by varying the model parameters A_0 and B_0 from -3% to 3% with an increment of 1% . The simulation matrix is given in Table 1.

Table 1. Simulation matrix of the sensitivity calculations. A_0 and B_0 are the variation of model parameters.

$A_0 \backslash B_0$	-3%	-2%	-1%	0%	1%	2%	3%
-3%	S1	S2	S3	S4	S5	S6	S7
-2%	S8	S9	S10	S11	S12	S13	S14
-1%	S15	S16	S17	S18	S19	S20	S21
0%	S22	S23	S24	S25	S26	S27	S28
1%	S29	S30	S31	S32	S33	S34	S35
2%	S36	S37	S38	S39	S40	S41	S42
3%	S43	S44	S45	S46	S47	S48	S49

In order to demonstrate the time-to-ignition variation with the change of A_0 from -3% to 3% at the fixed B_0 (i.e., simulations S22 to S28), the calculated peak cladding temperatures are plotted in Figure 5. Likewise for the variation of time to ignition in the model parameter A_0 , the variation by the model parameter B_0 is also plotted from a -3% to 3% range at the fixed A_0 (i.e., simulations S4, S11, S18, S25, S32, S39, and S46). As shown in Figure 5, the time to ignition is varies significantly with the variation of A_0 and B_0 from -3 to 0% . Its variation in comparison to the experimental value is approximately -20% to 2% in percentage.

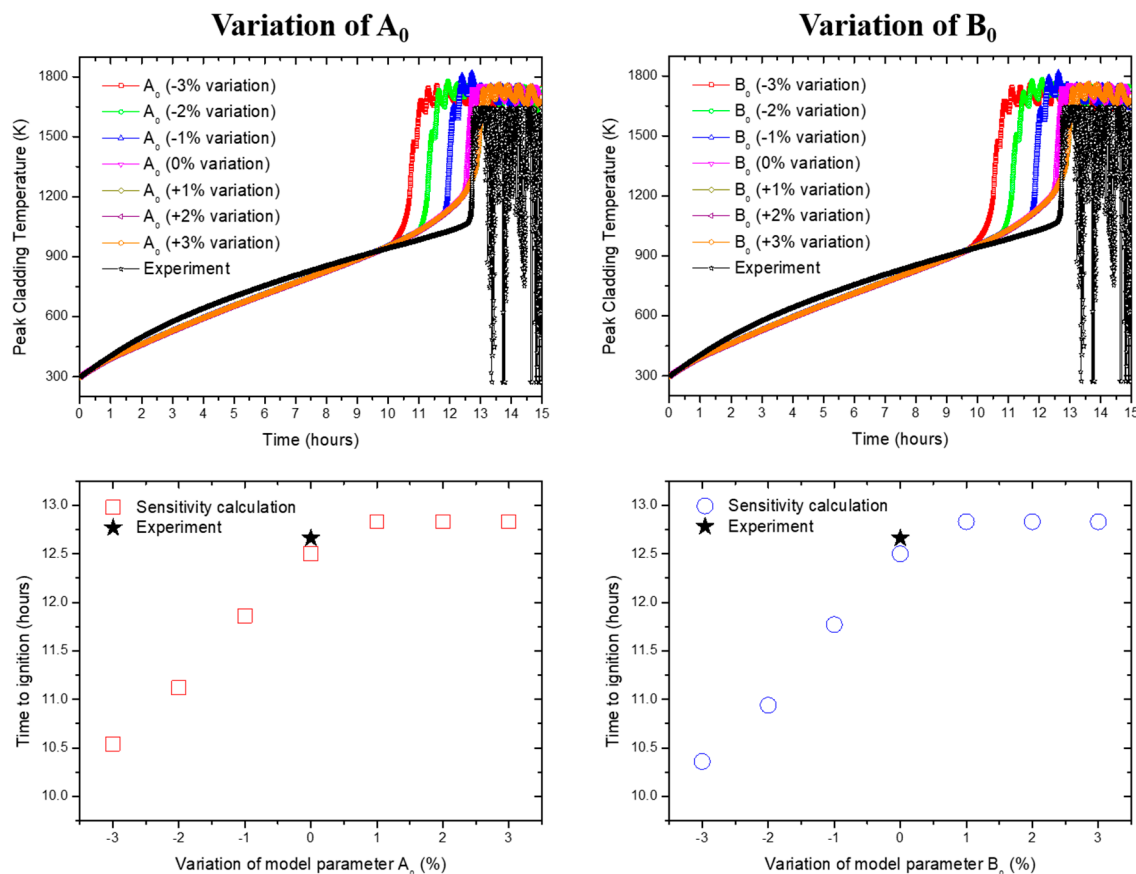


Figure 5. Calculated peak cladding temperature and variation of time to ignition.

A 3-dimensional plot to show the variation of time to ignition, with the variation of both A_0 and B_0 from -3% to 3% (i.e., simulations S1 to S49), is given in Figure 6. The highest time difference to ignition between the experiment and the calculation was ~ 2.76 h at the -3% variations of both A_0 and B_0 . This variation was not as extreme as expected by the simple calculations in the introduction. However, the variation in the predicted time of around 3 h may lead to an erroneous analysis of spent fuel accident sequences and the relevant accident management. As previously mentioned, the zirconium ignition would lead to a massive radioactive source term and hence the accurate prediction of time to ignition is very critical to prevent the large release of radioactive materials into the nuclear power plant and then to the neighboring environment. In order to prevent this disaster, timely accident management should be performed well before zirconium ignition. For this reason, improvement in the model is required to enable more accurate and reliable prediction of the time to ignition in comparison with the current model prediction.

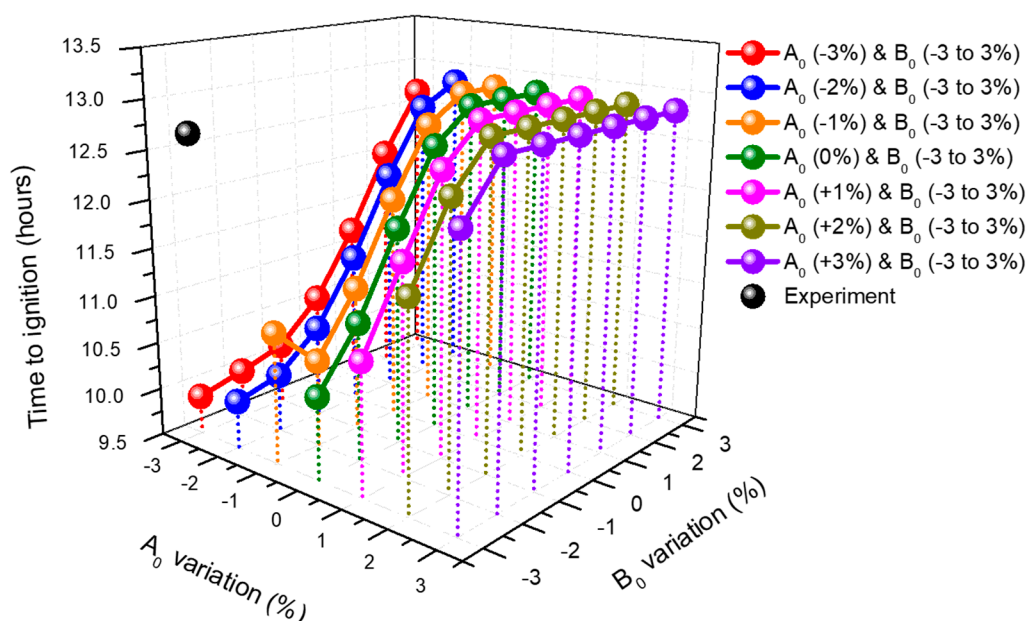


Figure 6. Variation of time to ignition according to the variation in model parameters A_0 and B_0 .

4. Conclusions

In this study, the zirconium fire phenomenon, which could occur during the postulated spent nuclear fuel pool loss-of-coolant accident scenario, was investigated. When the coolant is sufficiently drained out, the zirconium cladding is oxidized by air and develops the elevated temperature conditions where the zirconium fire would be initiated. Immediately after the initiation of a zirconium fire, oxygen was dramatically consumed and the cladding temperature very sharply increased. The air-oxidation kinetics changed from parabolic to linear, even to accelerated kinetics. This behavior is termed the air-oxidation breakaway. It causes a very rapid oxidation and a very high heat release. For this reason, the air-oxidation breakaway phenomenon was assumed to be the direct cause of zirconium ignition with a very abrupt cladding temperature escalation. To capture this phenomenon, a model was developed in the MELCOR code, namely the air-oxidation breakaway model. This current model was an empirical data-fitting model with limited air-oxidation test data. The empirical fitting of the current model was performed using a log–log scale to determine the model parameters between the air-oxidation breakaway timing and cladding temperature. It seems that the value of model parameters may include a non-negligible error from the data fitting on a log–log scale. Therefore, the sensitivity of a very small change (−3% to 3%) of model parameters on the air-oxidation breakaway timing was investigated through a number of simulations of the postulated SFP complete LOCA transient. It was found that a very small change of value in the model parameters induced a large difference in comparison to the experimental observation of the onset of a zirconium fire. From this sensitivity analysis, it is recommended that the current air-oxidation breakaway model should be improved by adopting a new model formula rather than using the current log–log curve-fitting model. In order to improve the weaknesses of the current model, the development of a new model is ongoing by considering the mechanism of air-oxidation breakaway phenomenon. It is known that the air-oxidation breakaway is initiated by the phase transformation from tetragonal to monoclinic ZrO_2 at the oxide–metal interface in the cladding. The newly developed model adopts this phase transformation mechanism and will be reported in the near future.

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