



Development of a semi-empirical cladding oxidation model and verification by BWR & VVER type CORA tests



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HIGHLIGHTS

- A non-parabolic and semi-empirical cladding oxidation model is proposed.
- The early failure of the cladding is taken into account.
- Three BWR type and two VVER type CORA tests are selected as the benchmarks.
- A judgment method of CORA bundle oxidation status is developed.
- A time-delay phenomenon of hydrogen generation rate is discussed and stressed.

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ABSTRACT

The high temperature oxidation with cladding damaged is complicated and the accurate simulation model needs to be developed. In this study, a non-parabolic and semi-empirical oxidation model is proposed for cladding high temperature oxidation with different damage regimes. Moreover, five past out-of-pile severe fuel damage experiments CORA-18, CORA-28, CORA-31, CORA-W1, and CORA-W2 (ISP36) are employed as cases for verification. Then, the hydrogen generation rates predicted by this semi-empirical model and several previous parabolic oxidation models are compared and analyzed. The total weight of hydrogen predicted by this new model can be in accordance with the related experiment values. This model considers the early cladding failure since 1473 K. The most conservative form of this model can be used as a replacer for Baker-Just model. Additionally, the suitable value of coefficients in this model may roughly represent the oxidation and damage status of the whole bundle.

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1. Introduction

Since the TMI-2 accident, a lot of research efforts have been focused on severe accidents. On the one hand, a large number of experiments and tests have been conducted for phenomenology and mechanism study. There are many important integrated experiment projects (Haste et al., 2015), such as CORAs, QUENCHs, PHEBUS-FPTs, PHEBUS-SFDs, etc. On the other hand, a lot of models and codes have been developed for severe accident simulation and analysis.

During severe accidents, the generation of hydrogen is a main hazard to the integrity of reactor pressure vessel, reactor building and containment. Hydrogen explosion leads to an early failure of these radiation barriers. The zirconium alloy oxidation is the most

important contributor for hydrogen source. One latest example is the Fukushima Daiichi accident, during which hydrogen explosions occurred and led to the collapse of reactor buildings (Fuchigami and Kasahara, 2015).

Steam oxidation of zirconium alloy is an important issue. Previous researchers (Baker and Just, 1962; Prater and Courtright, 1987; Urbanic and Heidrick, 1978; Leistikow and Schanz, 1987; Pawel et al., 1979) have laid the theoretical foundation for zirconium oxidation in steam. The zirconium oxidation kinetics is expressed in the parabolic oxidation law (Wang et al., 2015). The parabolic oxidation model is widely accepted in almost all famous severe accident analysis codes, including MELCOR (Gauntt et al., 2000), MAAP (Wang et al., 2014), SCDAP/RELAP5 (USNRC, 1997a, 1997b), ASTEC (Coindreau et al., 2010), and CATHARE/ICARE (Bandini et al., 2011). The parabolic law demonstrates that the rate-determining step of a metal-gas reaction is a solid-state diffusion process occurring within the barrier layer (Grosse, 2012; Tupin et al., 2003). Thus, the parabolic law is suitable for the solid-state processes of

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cladding oxidation, but not for the situations of oxide layer and/or clad damaged, such as oxide layer spalling (i.e. breakaway) (Grosse, 2012), cladding rupture and melting, etc. Different damage mechanisms conduct different kinetics of oxidation. For oxidation analysis of severe accident, both various damage regimes and the fractions of reaction area of different regimes should be taken into consideration. However, the previous clad oxidation models are only based on the assumptions of cladding solid-phase, intact oxide layer and unbroken cladding.

This work aims to develop a semi-empirical model to calculate the fuel cladding oxidation in steam under severe accident conditions, which expects to offer a more accurate post-analysis for integral weight and hydrogen generation rate. To verify this model, a new oxidation model analysis code (NOMAC) is developed, and then the rate or weight of hydrogen generated from three BWR and two VVER type CORA tests are conducted as benchmarks.

2. Theory model

2.1. Model framework

For the solid-state clad oxidation, the parabolic law has a similar form as Eq. (1) in severe accident analysis codes.

$$\frac{\Delta X^2}{\Delta t} = K(T) \quad (1)$$

X refers to the thickness of ZrO_2 , α -Zr(O) or ($ZrO_2 + \alpha$ -Zr(O)) layers, and the total weight of oxygen absorbed, zirconium reacted or ZrO_2 per unit surface area. T is cladding surface temperature.

When the oxides are removed completely, the reaction rate is determined by the transport rate of the oxygen ions in the gas phase, which can be assumed to be constant (Grosse, 2012). And then a linear dependence of parameters describing the reaction on the time t is the consequence (Grosse, 2012).

$$W = \sqrt{K(T)} \cdot t \quad (W(t=0) = 0) \quad (2)$$

W is the total weight of oxygen absorbed, zirconium reacted or ZrO_2 per unit surface area.

The Eq. (2) is derived from a difference equation

$$\frac{\Delta W^2}{(\Delta t)^2} = K(T) \quad (3)$$

When the structure of the oxide layer is changed or partially damaged, the parabolic law will be inapplicable for cladding oxidation. Such as the formation of cracks in cladding surface can speed up the diffusion, because the transport rate in open cracks or along the surface of a closed crack is much higher than that through the oxide layer (Grosse, 2012). Instead, a flexible power function can be used to describe this situation.

$$W = \sqrt{K(T)} \cdot t^{1/x} \quad (1 < x < 2) \&(W(t=0) = 0) \quad (4)$$

The Eq. (4) is derived from a difference form as Eq. (5)

$$\frac{\Delta W^2}{(\Delta t)^{2/x}} = K(T) \quad (5)$$

Under severe accident conditions, the core temperature is very high and the fuel clad is oxidized severely. At temperature about 1773 K, there will be a phase transition from tetragonal ZrO_2 to cubic ZrO_2 . This phase transition leads to an enlargement of diffusion coefficient. Nevertheless, in the case of high temperature oxidation, the mechanisms of oxygen ions diffusion within the oxide layer which corresponds to the dense oxide layer, the porous oxide layer, and the cracked oxide layer, respectively. Meanwhile, there exists the processes of the internal clad exposed due to cladding

rupture and cladding meltdown. Moreover, these processes will result in an increase of the total reaction area relative to the original surface of the fuel clad. This phenomenon is named as the Surface Increase Effect by this work. Notably, there is the low temperature failure of the cladding due to the interactions between low melting point materials and zirconium alloy cladding in the realistic reactor core.

Considering the above mentioned, the oxidation models can be expressed in a general form as

$$\Delta W^2 = K(T) \cdot [b_1 \cdot \Delta t + b_2 \cdot (\Delta t)^{2/x_2} + b_3 \cdot (\Delta t)^{2/x_3} + b_4 (\Delta t)^{2/x_4} + b_5 \cdot (\Delta t)^{2/x_5} + b_6 \cdot (\Delta t)^2] \quad (6)$$

where the unit of W^2 is $kg^2 m^{-4} s^{-1}$, $K(T)$ is the essential reaction rate, $b_i = \frac{A_{i, reaction}}{A_{ext}}$, A_{ext} is the sum of external surface of the fuel clad, $A_{i, reaction}$ is the reaction area of regime i, b_i is the fraction of reaction area of regime i. b_i is a function depending on the oxidation time and temperature.

The first term is the dense oxide term. It follows the parabolic law.

The second term is the phase transition term. The third term is the porous oxide term. The fourth term is the cracked oxide term. The fifth term is the cladding rupture term. Their oxidation rates will be faster than the parabolic law rate but slower than the linear law rate. Where $1 < x_5 \leq x_4 \leq x_3 \leq x_2 < 2$, x_2 is relatively close to 2, and x_5 is approximately equal to 1. Hence, the rate of the phase transition term is close to the parabolic rate, and that of the cladding rupture term is very similar to the linear law. In this model the phase transition term and the dense oxide term are considered as two minor factors for high temperature oxidation ($T > 1773$ K).

The sixth term is the cladding melting term. This term follows the linear law.

For model simplification, the x_2 is set to 2, the x_3 is set as the same with x_4 , and the x_5 is set to 1, then the simplified model without considering the Surface Increase Effect can be written as:

$$\frac{\Delta W^2}{K(T)} = \begin{cases} [c_1 \cdot \Delta t + (c_3 + c_4) \cdot (\Delta t)^{2/x_4} + (c_5 + c_6) \cdot (\Delta t)^2] & \text{Tetragonal } ZrO_2 \\ [(c_1 + c_2)\Delta t + (c_3 + c_4) \cdot (\Delta t)^{2/x_4} + (c_5 + c_6) \cdot (\Delta t)^2] & \text{Tetragonal to Cubic } ZrO_2 \\ [c_2 \cdot \Delta t + (c_3 + c_4) \cdot (\Delta t)^{2/x_4} + (c_5 + c_6) \cdot (\Delta t)^2] & \text{Cubic } ZrO_2 \end{cases} \quad (7)$$

where $c_i = \frac{A_i}{A_{ext}}$, A_i is the radial nominal area of regime i, c_i is the fraction of radial nominal area of regime i. c_i is also a function depending on the oxidation time and temperature. $A_i \leq A_{i, reaction}$, $c_i \leq b_i$.

If the value of x_4 is assumed to be 1.5, then

$$\Delta W^2 = K(T) \cdot [c'_1 \cdot \Delta t + c'_2 \cdot (\Delta t)^{4/3} + c'_3 \cdot (\Delta t)^2] \quad (8)$$

and

$$[\Delta t] = c'_1 \cdot \Delta t + c'_2 \cdot (\Delta t)^{4/3} + c'_3 \cdot (\Delta t)^2 \quad (9)$$

where $c'_1 = c_1 + c_2$, $c'_2 = c_3 + c_4$, $c'_3 = c_5 + c_6$, $1.0 \leq c'_1 + c'_2 + c'_3 \leq 2.0$, $1.0 \leq c'_1 + c'_2 \leq 2.0$, $0.0 \leq c'_3 \leq 1.0$. A nominal time term $[\Delta t]$ is defined in this semi-empirical model, and the unit of $[\Delta t]$ is still defined as second.

However, when the Eq. (9) is used for oxidation calculation immediately, the results will be underestimated. This is because the total reaction area should be greater than the initial surface area under severe oxidation conditions. To solve this problem, an alternative form for the nominal time term is recommended as:

$$[\Delta t] = [c'_1 \cdot \sqrt{\Delta t} + c'_2 \cdot (\Delta t)^{2/3} + c'_3 \cdot (\Delta t)]^2 \quad (10)$$

With the expansion of Eq. (10), it has an equivalent form as follows

$$[\Delta t] = c_1^2 \cdot \Delta t + 2c'_1 c'_2 \cdot (\Delta t)^{7/6} + c_2^2 \cdot (\Delta t)^{8/6} + 2c'_1 c'_3 \cdot (\Delta t)^{9/6} + 2c'_2 c'_3 \cdot (\Delta t)^{10/6} + c_3^2 \cdot (\Delta t)^2 \quad (11)$$

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