The Effect of Protective Coating on the LOCA Simulation of Zircaloy-4 Cladding

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1. Introduction

After the severe accident of Fukushima in Japan in 2011, the research and development of improving the cladding materials which may tolerate accident situations have been initiated. The alternative cladding materials should possess the tolerance of loss of active cooling in the core for a considerably longer time period while maintaining the fuel performance during normal operations.

These are three major potential approaches for the development of accident tolerant fuel designs [1]:

- Improved fuel properties
- Improved cladding properties to maintain core coolability and retain fission products
- Improved reaction kinetics with steam to minimize enthalpy input and hydrogen generation

And these are some constraints on new cladding designs and coatings, such as [1]:

- Backward compatibility (must be qualified in an existing reactor)
- Economics
- Minimal impact on the fuel cycle
- Minimal impact on the plant operations
- Significant impact (improvement) on plant safety (for the entire spectrum of design basis [DB] accidents and beyond design basis [BDB] accidents)

In this study, a transient fuel performance code has been used to study the impact of coating the Zircaloy-4 cladding by Silicon Carbide (SiC) on the fuel performance under design basis accident conditions, particularly a loss of coolant accident (LOCA).

2. Method

2.1 Simulation Tool

FRAPTRAN [Fuel Rod Analysis Program Transient] is a FORTRAN language computer code which calculates the transient performance of light-water reactor fuel rods during reactor transients and hypothetical accidents such as loss-of-coolant accidents (LOCA), anticipated transients without scram (ATWS), and reactivity-initiated accidents (RIA).

FRAPTRAN calculates the temperature and deformation history of a fuel rod as a function of time-dependent fuel rod power and coolant boundary conditions, which is a useful tool for the study on the effects of changing the cladding material.

For transient analyses at other than beginning-of-life conditions, FRAPTRAN needs input parameters that account for the effect of burnup (e.g., radial dimensions that account for fuel swelling and cladding creepdown). These values may be obtained from a steady-state fuel performance code such as FRAPCON-3. Due to this, FRAPCON-3 has initially been used to generate these values (in the "Restart" file) to be used as inputs for FRAPTRAN.

2.2. The Coating Material

The candidate coating material considered in this study is silicon carbide (SiC). Unlike Zircaloy-4, SiC has a low oxidation rate at high temperatures when the liquid water turns to steam during accident scenarios.

The properties which make SiC a very powerful candidate material are:

- Improved irradiation stability
- Effective barrier to fission product release
- Greater reliability under the anticipated operating and long-term storage conditions

But it is observed that the thermal conductivity of SiC is reduced by >30% after irradiation [1].

2.3 The Accident Scenario

LOCA is a design basis accident which can be defined as a break in the primary coolant piping of a reactor at full power with subsequent scram/trip. Those postulated accidents that result in a loss of reactor coolant at a rate in excess of the capability of the reactor makeup system from breaks in the reactor coolant pressure boundary, up to and including a break equivalent in size to the double-ended rupture of the largest pipe of the reactor coolant system.

In this study, a LOCA scenario, which affects the coolability of the reactor and induces the oxidation of the cladding at high temperatures, has been studied to see the effects of SiC coating on the oxidation behavior of the coated Zircaloy-4 cladding.

2.4 Pre-Simulation Requirements

Since the properties of silicon carbide are not available in FRAPCON and FRAPTRAN when choosing the cladding type, modifying the source code is necessary. But since the oxidation model of the coating is only important for high temperature steam oxidation in transient analyses, we need to modify the source code for FRAPTRAN only.

By modifying the oxidation behavior formula in source code of FRAPTRAN, in particular the subroutine "COBILD" and running the LOCA input file, the oxidation behavior results of SiC coating on the Zircolay-4 cladding can be obtained.

Starting by FRAPCON-3 which models nuclear fuel pin performance under normal operating conditions to get the required burnup-dependent parameters for the run of FRAPTRAN in the "Restart" file.

After running FRAPCON-3 and get the restart file, the parameters for FRAPTRAN are obtained.

3. Oxidation Models

3.1 The Oxidation Formulas of Zircaloy-4 and Silicon Carbide

Cathcart-Pawel (C-P) oxidation model calculates oxide thickness, weight gain, and energy generation once cladding average temperature exceeds 1073K (800°C). This model is considered a best-estimate model and is based on oxidation data collected at temperatures greater than 1273K (1000°C).

The Cathcart-Pawel model is stated in MATPRO to be [2]:

$$K_2^2 = K_1^2 + 2 * 1.126 * 10^{-6} \exp\left(\frac{-1.502 * 10^5}{8.314 * T}\right) \Delta t \ldots (1)$$

where:

- K = thickness of oxide layer
- T = temperature (K)
- t = time (s)

The kinetics of the oxidation of SiC (that produces a silica layer on the SiC surface) consists of two processes: oxidation and volatilization. These can be modeled by a parabolic oxidation rate constant, k_p and by linear volatilization constants k_l, respectively.

When oxidation and volatilization occur simultaneously, the oxidation rate is affected by the volatilization rate. And the overall kinetics are described by the more complex paralinear kinetics.

Paralinear oxide scale thickness kinetics has been mathematically described by Tedmon for Cr_2O_3-forming Fe-Cr alloys [3]. This expression is directly applicable to SiC and is as follows:

$$t = \frac{k_p}{2k_l}\left[-\frac{2k_l x}{k_p} - ln\left(1 - \frac{2k_l x}{k_p}\right)\right] \ldots (2)$$

where:

- t: is the oxidation time.
- x: is the oxidation thickness.
- k_p: is the parabolic rate constant for oxidation in units of thickness per unit time = 0.5 (μm^2/h) at temperatures between 1200° and 1400°C.
- k_l: is the linear rate constant for scale volatilization in units of thickness/time = 0.02 (μm/h).

k_p (μm^2/h) is the parabolic rate constant [4]. This rate can be described by the usual Arrhenius type relationship as follows:

$$k_p = k^o_{p,H_2O}\left(\frac{p_{H_2O}}{p^m_{H_2O}}\right) exp\left(\frac{-E_{a,H_2O}}{RT}\right) \ldots (3)$$

where:

- k^o_{p,H_2O}: is the pre-exponential constant (kg^2-SiO_2/m^4-s) = 6.915 * 10^{-9}
- R: is the universal gas constant (J/mol-K) = 8.3144621
- E_a: is the activation energy (J/mol) = 35 * 10^3
- p_{H_2O}: is the water pressure (Pa) with m representing the water pressure at which k^o was derived.

At long times, the scale thickness becomes a constant as the oxidation rate equals the volatilization rate. This limiting scale thickness, thickness is equal to:

$$x = \frac{k_p}{2k_l} \ldots (2^*)$$

In another reference by Terrani [5], Eq. (2) has been modified and solved for x (the oxide thickness) using the *Lambert W function*. Starting by the evolution in mass of the specimen solely due to oxygen pickup in the oxide layer can then be determined using:

$$t = \frac{\alpha^2 k_p}{2{k_1}^2}\left[\frac{-2k_1}{\alpha k_p}\Delta w_1 - ln\left(1 - \frac{2k_1}{\alpha k_p}\Delta w_1\right)\right] \ldots (4)$$

Total mass evolution in the specimen is the sum of mass gain due to oxide layer formation (Δw_1) and mass loss owing to oxide layer volatilization (Δw_2). Solving for the former in Eq. (2) one arrives at:

$$\Delta w_1 = \frac{\alpha k_p}{2k_1}\left[1 + W\left(-exp\left(\frac{2k_1^2 t}{\alpha^2 k_p} - 1\right)\right)\right] \ldots (5)$$

where W is the Lambert W function. Mass loss due to oxide volatilization is expressed simply via:

$$\Delta w_2 = -k_1 t \ldots (6)$$

The net mass change in the specimen, Δw_{net} is then given by the sum of mass changes:

$$\Delta w_{net} = \Delta w_1 + \Delta w_2 \ldots (7)$$

α is determined based on the net mass gain according to the reaction and is defined as:

$$\alpha = \frac{MW_{SiO2}}{MW_{O2} - MW_{C}} \approx 3$$

Fig.1 shows a comparison between the oxidation behavior of Zircaloy-4 obtained by Eq. (1) and silicon carbide obtained by Eq. (2)

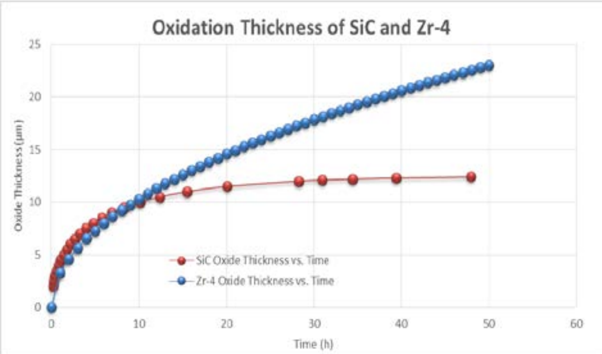


Fig.1: Comparison between Zr-4 and SiC Oxidation Behavior.

3.2 Alternative Oxidation Formula for Silicon Carbide

Converting Eq.(2) formula to give the oxide thickness as a function of time will result in the use of the Lambert W function which is complicated to be inserted in the subroutine COBILD. An alternative fitted correlation of the oxidation behavior can be obtained by using the literature data of SiC oxidation and comparing the oxidation kinetics of Zircaloy-4 and SiC which will result in a modified oxidation equation which can be used in COBILD.

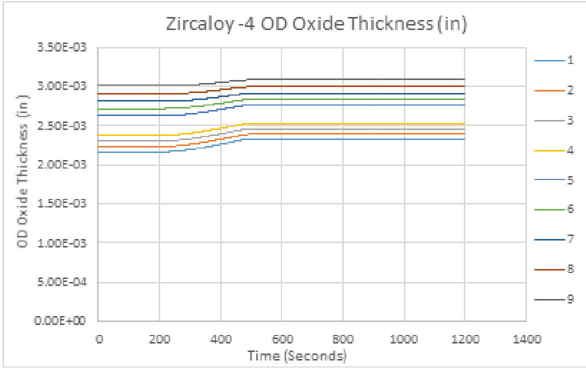
While modifying the source code, other interconnected equations such as the oxygen uptake and the oxygen stabilized α-phase have been adjusted to fit the oxidation behavior of SiC coated cladding.

4. FRAPTRAN Simulation and Results

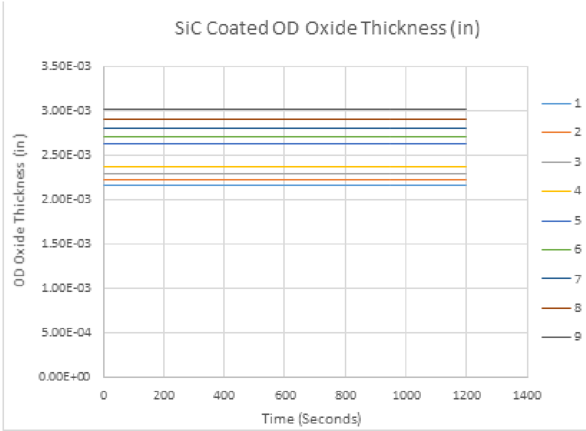
After modifying the source code, particularly the subroutine “COBILD”, a LOCA scenario input file has been prepared, under the name of “LOCA Test” considering a cladding type choice of Zircaloy-4 which has been modified through the source code to be coated by SiC and a typical power history for a PWR. and runs for both Zircaloy-4 cladding and the SiC coated cladding have been done and the outputs have been obtained.

Fig.2 shows the outer diameter oxide thickness as a function of time for the Zircaloy-4 cladding and the SiC coated cladding, respectively. These results show that the oxidation in the SiC coated cladding is minimal when compared with non-coated Zircaloy-4 under the same LOCA scenario.

The numbers 1-9 in the graphs represent the axial regions of the fuel where the oxidation thickness has been calculated.



(a)



(b)

Fig.2: The outer diameter oxidation layer thickness of (a) Zircaloy-4 cladding and (b) SiC Coated Zircaloy -4 cladding.

To evaluate the effectiveness of protective coating on normal and transient fuel performance, the material properties of protective coating under irradiation has to be considered. In addition to the oxidation behavior, further studies should cover the effects of the mechanical properties, corrosion, irradiation behavior, thermal expansion, fatigue and creep of candidate protective coating materials.

5. Conclusions

The preliminary transient analyses show that the protective coating on Zircaloy-4 cladding can lead to the minimization of LOCA consequences, because the steam oxidation rate of coated surface is reduced compared with that of bare Zircaloy-4 surface.

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