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Uncertainty of the Oxide Layer Thickness Formed on the Aluminum Cladding of U–Mo/Al Plate-Type Dispersion Fuel

Faris B. Sweidan[a], Young-Wook Tahk[b], Jeong-Sik Yim[b], Ho Jin Ryu[a*]
[a] *Department of Nuclear and Quantum Engineering, KAIST, Yuseong-gu, Daejeon 34141, Republic of Korea*
[b] *Korea Atomic Energy Research Institute, Yuseong-gu, Daejeon 34057, Republic of Korea*
**corresponding author: hojinryu@kaist.ac.kr*

1. Introduction

As the development of low-enriched uranium (LEU) fuels has been pursued for research reactors to replace the use of highly-enriched uranium (HEU) for the improvement of proliferation resistance of fuels and fuel cycles [1], U-Mo particles dispersed in an Al matrix (U-Mo/Al) is a promising fuel for conversion of the research reactors that currently use HEU fuels to LEU-fueled reactors due to its high density and good irradiation stability [2].

Uncertainties of the key parameters may influence a significant impact on the fuel temperature since the fuel performance, represented by swelling, fission gas release, and interaction layers formation is affected by the fuel temperature and vice versa.

In the authors' previous work [3], the effect of the uncertainties of some of the important parameters affecting the operating temperature of the fuel, including reactor operation conditions, fuel fabrication, fuel properties, and the dynamic changes of fuel during irradiation, such as the thermal conductivity of irradiated fuel, oxide layer thickness and pH value uncertainties, are used to determine the probable fuel temperature ranges. However, the oxide layer thickness uncertainty seemed to have more complications because of the effect of the pH value uncertainty since it adds a non-linear effect to the oxide layer thickness uncertainty as it is considered as a major parameter affecting the oxide layer thickness forming on the cladding.

In this study, a deeper look into the uncertainty of the oxide layer thickness is evaluated, taking into account all the important parameters affecting the deviation of the oxide layer formation and their effects on the value of the formed oxide thickness. Multiple models are studied for the sake of comparison, including each model's potential parameters that are a part of the effect occurring to the oxide layer thickness. These parameters include the heat flux, the heat transfer coefficient, the coolant temperature, the coolant flow rate, and most importantly, the pH value. The pH is a major potential parameter in some of the chosen models, but some models do not include it in the calculations. Gaussian distributed random number generation is used as a type of Monte Carlo Simulation to evaluate the uncertainty of the oxide layer thickness as the potential parameters affecting it change randomly within their uncertainty ranges.

This study aims to evaluate the oxide layer thickness uncertainty using different models, which is an important parameter in the operating temperature evaluation of the U-Mo/Al fuel. This study will be added to the authors' previous work to cover all the parameters affecting the temperature evaluation.

2. Methodology and Model Choice

As mentioned previously, aluminum alloy cladding experiences oxidation layer growth on the surface during the reactor operation. The oxide growth model developed by Kim et al. [4] that uses a variable rate-law power in a function of irradiation time, temperature, surface heat flux, water pH, and coolant flow rate, was used to estimate the oxide film thickness as a function of burnup. The predicted oxide thickness is sensitive to water pH, as the chosen reference reactor for this study is ATR, it is reported that water pH is in the range of 5.0 ~ 5.7 [5].

The model by Kim et al. [4] consists of multiple equations for all the parameters needed to calculate the oxide layer thickness precisely. All these equations are as follows [4]:

$$x = [x_o^{p+1} + (p+1)kt]^{\frac{1}{p+1}} \quad (1)$$

$$p = 0.12 + 9.22\exp\left(-\frac{C_s}{6.82 \times 10^{-9}}\right) \quad (2)$$

$$lnC_s = -\left(-13.79 - \frac{1211.16}{T_{x/w}}\right)(0.041H^2 - 0.41H - 0.07) \quad (3)$$

$$k = 3.9 \times 10^5 \exp\left(\frac{-6071}{T_{x/w} + AB^{\frac{qx}{k_T}}}\right) \quad (4)$$

$$A = 0.43 + \frac{3.21}{1 + \exp\left(-\frac{v_c - 13.39}{3.60}\right)} \quad (5)$$

$$k_T = 2.25, \quad for\ x \le 25$$
$$k_T = 2.25 - 0.016(x - 25), \quad for\ 25 \le x \le 100 \quad (6)$$

where:

- x: the oxide thickness in μm
- t: time
- k: reaction constant
- p: rate-law power
- C_s: the oxide solubility in g/g H_2O
- $T_{x/w}$: The oxide-water interface temperature in K

- H: the pH value.
- A: the augmentation factor
- v_c: the coolant flow rate in m/s
- q: the heat flux (surface) in MW/m^2
- k_T: the oxide thermal conductivity in W/mK
- B: a correction factor (B=0.37)

For this study, the uncertainties of the heat flux, heat transfer coefficient, the coolant flow rate, the coolant temperature, and the pH value have been used to evaluate the uncertainty of the oxide layer thickness.

3. The Oxide Layer Thickness Parameters and their Uncertainties

The oxide layer thickness calculations require the value and the uncertainty of the five major parameters: the heat flux, the heat transfer coefficient, the coolant temperature, the coolant flow rate, and the pH value. The cladding outer temperature is calculated from the heat flux, heat transfer coefficient, and the temperature of the coolant, accordingly.

3.1 Heat Flux Profiles and their Uncertainty

A realistic fitted heat flux profile has been used from the beginning of life (BOL), maximum, and end of life (EOL) heat flux values of the Advanced Test Reactor (ATR) for low and high heat flux profiles [5]. The high heat flux position represents the hot region (midplane is located 4.5 cm from the bottom of the fuel plate) and the low heat flux position represents the cold region (midplane is located 23 cm from the top of the fuel plate).

The uncertainty of the surface heat flux is a result of the uncertainty of two major parameters, the variation of uranium density (homogeneity) in the fuel meat (uranium density uncertainty) and the neutron flux uncertainty.

The uncertainties of uranium density and neutron heat flux are ± 16% [6] and ± 10% [7], respectively. To combine the uncertainties, the equation to calculate the uncertainty of a parameter resulting from the multiplication/division of parameters is used [8] as follows:

$$\frac{\Delta y}{y} = \sqrt{\left(\frac{\Delta x_1}{x_1}\right)^2 + \left(\frac{\Delta x_2}{x_2}\right)^2 + \left(\frac{\Delta x_3}{x_3}\right)^2 + \dots} \quad \dots (7)$$

Combining the uncertainties of the two parameters results in the uncertainty of the surface heat flux of about ±19%.

3.2 Heat Transfer Coefficient and its Uncertainty

The heat transfer coefficient was used as 3.03 W/cm^2-K and was assumed to be constant. This value has been used by Medvedev [9] for the temperature calculation of U-Mo/Al dispersion fuel as a part of the RERTR program and by Burkes [1]. According to a reference by W.L Woodruff [10], the heat transfer coefficient uncertainty fits within a band of ±20% for any of the single phase correlations commonly used.

3.3 The pH Value and its Uncertainty

The predicted oxide thickness is sensitive to water pH and it is a reactor dependent. It is reported that water pH is in the range of 5.0 ~ 5.7 for ATR [5].

3.4 The Coolant Flow Rate and its Uncertainty

Kim et al. [4] reported that the range of the coolant velocity during the oxide layer thickness formation experiment was 3-28 m/s. This range indicates a large deviation in the coolant flow rate resulting of an uncertainty of around ±80%.

3.5 The Coolant Temperature and its Uncertainty

It is reported by Burkes [1] that the coolant temperature was relatively consistent throughout the irradiation experiment, ranging from 52 – 55 ºC for the low heat flux position and 55 – 61 ºC for the high heat flux position. These ranges indicate that the uncertainties of the coolant temperatures ranges from ±3% to ±5% for the low and high heat flux positions, respectively.

Table 1 shows the parameters used for the oxide layer thickness calculation and their uncertainties.

Table 1: Parameters and their uncertainties.

Parameter	Uncertainty
Surface heat flux profile	±19%
Heat transfer coefficient	±20%
pH value	5.0 - 5.7
Coolant temperature	High Flux: ±5% Low Flux: ±3%
Coolant flow rate	3-28 m/s (~±80%)

4. Results and Discussion

A set of random numbers was generated for each parameter. The set is distributed following the Gaussian distribution and the numbers were generated for each burnup step consisting of 1000 numbers. These numbers were used to calculate the oxide layer thickness resulting of the calculated oxide layer thickness for each burnup step. The uncertainty of the oxide layer thickness based on the Gaussian random number generation was obtained applying an upper and lower bound of the calculated values of a confidence level of 95% (~2σ).

The Gaussian distributed random number generation shows an average oxide layer thickness uncertainty of ±29% and ±44% for the low and high heat flux profiles, respectively. The uncertainty increases from the low heat flux position to the high heat flux position as the uncertainty of the parameters becomes more impactful on the oxide layer thickness as the heat flux increases, this leads to a higher deviation in the oxide layer thickness calculations.

Comparing these uncertainties with the uncertainty resulting from the scatter of data stated by Kim et al. [4], which was ±10%, shows that the uncertainties of the parameters that are used to calculate the oxide layer thickness contribute more to the overall uncertainty of the oxide layer thickness resulting of a significantly higher uncertainty.

5. Conclusions

In this study, the uncertainty of the oxide layer thickness formed on the aluminum cladding of U-Mo/Al plate-type dispersion fuel was calculated. The resulting uncertainty was calculated using random number generation method (Monte Carlo Simulation) utilizing the uncertainty of the heat flux, the heat transfer coefficient, the coolant flow rate, the coolant temperature and the pH value.

The chosen model to calculate the uncertainty in this part of the study is the model by Kim et al. [4]. The resulting uncertainty from the calculations is considerably higher when compared to the uncertainty value available in the literature resulting from the data scatter. However, additional work must be added to compare the uncertainty of the oxide layer thickness when calculated using different models, such as Griess model. This uncertainty value will be added to the authors' work that analyzes the effect of the potential parameters and their uncertainties on the operating temperature of the U-Mo/Al plate-type dispersion fuel.

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