OVERVIEW OF ALUMINUM CLADDING OXIDE PREDICTION MODELS FOR EUROPEAN HIGH POWER RESEARCH REACTORS

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ABSTRACT

UMo/Al dispersion fuel clad with aluminum alloy is a primary candidate fuel form that is being developed for the European high power research reactors (EUHPRR). In BR2 tests, water oxidation of cladding grew thick due to high powers. Excessive cladding oxidation is a cause for serious fuel performance degradation in a test while fuel performance itself may be sound. It is necessary for a fuel designer to have a reliable model to predict cladding oxidation for fuel licensing activities. Using oxide data from BR2 tests, this study reviews the prominent oxide prediction models and cladding-to-coolant heat transfer correlations. Because the oxide growth models were based upon different coolant flow conditions from those of the EUHPRR, the effect of the heat transfer correlations at the cladding-to-coolant interface on the oxide thickness prediction were also examined. The best combination of the oxide prediction model and the heat transfer correlation applicable for the EUHPRR is recommended. Since the existing oxide models were all developed for aluminum alloy types different from those currently considered for the EUHPRR, the effect of cladding alloy type is also discussed.

1. INTRODUCTION

For the conversion of the European high power research reactors (EUHPRR) such as BR2, RHF, and JHR from highly enriched uranium (HEU) to low enriched uranium (LEU), a high uranium-density fuel is being developed. The primary candidate is UMo alloy fuel kernel dispersion in an Aluminum matrix (UMo/Al) with a meat uranium-density of up to 8.5 g-U/cm³. In order for this fuel to be qualified, stable and predictable fuel behavior, and mechanical integrity and dimensional stability of the fuel plate must be demonstrated over the range of anticipated normal and off-normal operating conditions. In this regard, two traditionally used metrics to assess fuel performance are fuel meat swelling (or fuel plate thickness expansion) and cladding oxidation. The former performance topic has received extensive studies including experiment and modeling. The latter has had relatively less attention because no further development for cladding has been pursued from previously qualified fuels.

Aluminum alloy has well served as research reactor fuel cladding since its first use for the MTR in 1950s. When fuel temperatures were relatively low, cladding oxidation was not a concern. However, when LEU fuel is considered for high power applications, cladding
oxidation becomes a critical factor that elevates fuel temperature because its oxide, typically a Boehmite, has a thermal conductivity that is about two orders of magnitude lower than aluminum. Therefore, excessive oxidation can potentially degrade fuel performance that may be otherwise sound. In this sense, providing a reliable prediction model for cladding oxidation is crucial to help fuel design and improve modeling fuel performance behavior.

Because the existing oxide growth models were mostly developed based on the measured data obtained at lower temperatures and powers than the EUHPRR, a review of the existing models became necessary before applied to the EUHPRR. The frequently used oxide prediction models available in the literature were investigated in this study by comparing with the measured data from the tests at bounding power and burnup conditions in the BR2.

The accurate prediction of cladding surface temperature relies on an accurate model for the heat transfer coefficient at the cladding surface. The most frequently used models predicting the heat transfer coefficient at the cladding surface were also examined, incorporating detailed thermal-hydraulic properties.

In addition, the effect of alloy types on oxidation growth kinetics was examined between AG3NE and AlFeNi.

2. Basic data for oxide thickness prediction

2.1 Irradiation tests

The oxide data of the irradiation tests E-FUTURE [1] and SELENIUM [2], irradiated in the BR2 at SCK.CEN, were examined in this study. Three full-sized plates out of four of the E-FUTURE test (see Figure 1) and one plate from the SELENIUM test were picked. Two different cladding types, AG3NET and AlFeNi, were studied. Plate dimensions are shown in Figure 2.

Figure 1 Schematic of the cross section of the E-FUTURE basket with four fuel plates. The same basket was used for the SELENIUM test, with the fuel plates loaded in the bottom half of the basket.

Figure 2 Schematic of a fuel plate and meat used for E-FUTURE and SELENIUM.
Both tests had the same coolant speed of 12 m/s (downward flow), the coolant pH of 6.1, and the coolant inlet temperature of 38 °C, and the same test basket having the coolant channel gap of 6.4 mm.

Power histories of the plates are shown in Figure 3 [3][4]. Heat flux data along the axial line 41 mm from the lower-power plate-edge are given because the peak oxide thicknesses were measured along this line. Although the power was lower there than at the meat edge, the highest cladding surface temperature was predicted along this line due to lower cooling.

![Figure 3](image)

Figure 3 Power histories of E-FUTURE plates and SELENIUM plate along the line 41 mm from the lower-power plate-edge

### 2.2 Heat transfer coefficient at cladding surface

Due to the high coolant speed, the coolant flow condition of the experiments in this study was in the turbulent flow regime. Convection is, therefore, the dominant mode of heat transfer. Applying Newton’s law of cooling, the cladding surface temperature can be calculated by

\[ T_{cw} = T_b + \frac{q''}{h_a} \]  (1)

where \( T_b \) is the bulk fluid temperature, \( q'' \) is the heat flux, and \( h_a \) is the heat transfer coefficient (HTC).

Four HTC correlations applicable to plate type geometry, including the Dittus-Boelter
correlation [5], the Sieder-Tate correlation [6], the Colburn correlation [7], and the recently developed KAERI correlation [8], were selected for the study. The Dittus-Boelter correlation is recommended specifically for a situation in which the difference between cladding surface temperature and fluid temperature is small. On the other hand, the Sieder-Tate correlation is known to be more accurate for a condition that has a large temperature difference between cladding surface and fluid because it is capable of explicitly incorporating the viscosities of the fluid at the bulk fluid temperature and the cladding surface temperature. The Colburn correlation is similar to the Dittus-Boelter correlation while it is different in that it considers the fluid properties at the film temperature defined by the average of the bulk coolant temperature and cladding surface temperature. The KAERI correlation was recently developed based on experimental data measured for rectangular channels, simulating coolant channels set by fuel plates.

2.3 Cladding surface temperature

In order to compare the four HTC correlations discussed in subsect. 2.2, the calculated Nusselt number and the cladding surface temperature versus axial length for the E-FUTURE 6301 plate are plotted in Figure 4. Because the HTC is proportional to the Nusselt number, a higher Nusselt number yields a lower cladding surface temperature (see Eq. (1)).

The Dittus-Boelter correlation gave the highest cladding surface temperature, which may be attributed to the model’s inability to differentiate the coolant viscosity at the cladding surface and that of the bulk coolant. The difference in the cladding surface temperature between the predictions by the Dittus-Boelter and KAERI correlations was ~30 °C at axial length ~500 mm. The axial surface temperature was calculated using a program developed at KAERI (Thermal Hydraulic Margin Calculator for Plate-type Fueled Reactor Core for Windows) [9].

3. Oxide thickness prediction models

3.1 ANL model

Oxide growth on cladding surface follows the following equation [10]:

\[ x = x_0^{p+1} + (p + 1) k t^{p+1} \]  

(2)

where \( x_0 \) is the oxide thickness at time zero, \( p \) is the rate law power, \( k \) is the rate function and \( t \) is the time.
The rate law power $p$ is given by:

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

(3)

Here $C_s$ considers oxide dissolution in coolant and is expressed by:

$$C_s = \exp \left[ - \left( -13.79 - \frac{1211.16}{T_{x/w}} \right) 0.041H^2 - 0.41H - 0.07 \right]$$

(4)

where $T_{x/w}$ is the temperature at the oxide-water interface in K and $H$ is pH of the coolant. The applicable temperature range is $25 \leq T \leq 300 \, ^\circ C$ and pH not greater than 7.0.

The rate function $k$ is expressed by an empirical formula:

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

(5)

where $T_{x/w}$ is the oxide-water interface temperature in K, $q''$ is the surface heat flux in MW/m$^2$, $x$ is the oxide thickness in µm, $k_T$ is the thermal conductivity of the oxide in W/m-K, $A$ is the augmentation factor as described below.

$A$ is added to the equation as a multiplier to take into account the effect of coolant velocity. The augmentation factor increases as the coolant velocity increases because of the water ingress through the defective oxide. $A$ is correlated with the coolant velocity using the following sigmoidal function:

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

(6)

where $v_c$ is the coolant velocity in m/s. The applicable range of coolant velocity for this correlation is 3 - 28 m/s.

A correction constant, $B$, is needed to account for the reduction in the ‘oxide thickness’ caused by oxidant migration. $B = 0.37$ for AA 6061 and the ATR data. Since $x$ is included in Eq.(5), the time interval was set equal to the reactor cycle length.

The oxide thermal conductivity decreases as the oxide thickens. The oxide thermal conductivity was formulated as a function of the oxide thickness as follows:

$$k_T = 2.25, \quad \text{for } x \leq 25,$$

$$k_T = 2.25 - 0.016 (x - 25), \quad \text{for } 25 \leq x \leq 100$$

(7)

(8)

where $k_T$ is in W/m-K and $x$ is the oxide thickness in µm.

### 3.2 Griess Model

The Griess model uses the following correlations for two pH regimes [11]:

$$x = \begin{cases} 
11.252 \exp \left( - \frac{4.600}{T} \right)^{0.778}, & \text{for } pH = 5.0 \\
30.480 \exp \left( - \frac{4.600}{T} \right)^{0.778}, & \text{for } 5.7 \leq pH \leq 7.0
\end{cases}$$

(10)

where $x$ is the oxide layer thickness in µm, $t$ is the time in h, and $T$ is the cladding surface
temperature in K. The uniqueness of the Griess model is its ignorance of the effect of heat flux.

3.3 KAERI Model (Modified-Griess model)

The KAERI model is the product of an experimental study on the effect of the heat flux on oxide growth to modify the Griess model. The experiment study found that the Griess model over-predicted when the heat flux was below 3.18 MW/m². A modification factor was added to the Griess model as follows [9]:

\[
x = \begin{cases} 
11,252 \exp \left( -\frac{4,600}{T} \right) t^{0.778} f_q, & \text{for } pH = 5.0 \\
30,480 \exp \left( -\frac{4,600}{T} \right) t^{0.778} f_q, & \text{for } 5.7 \leq pH \leq 7.0 
\end{cases}
\]  

(9)

Here the factor \( f_q \) is a function of the heat flux given by

\[
f_q = \begin{cases} 
0.2, & \text{for } q < 2.16 \text{ MW/m}^2 \\
-0.20836 + 0.18915q, & \text{for } 2.16 \text{ MW/m}^2 < q
\end{cases}
\]  

(10)

where \( q \) is the heat flux in MW/m².

4. Oxide thickness prediction

The three oxide prediction models described in sect. 3 were employed to calculate oxide thickness for the test conditions at BR2. The details of this section can be found elsewhere [12].

The oxide thickness data measured along the axial line 41 mm from the cooler side of plate edge are compared with the predictions made by the models for four HTC correlations for E-FUTURE 6301 plate (Figure 5) and for SELENIUM 1221 (Figure 6). When coupled with the Colburn correlation or the Sieder-Tate correlation, the ANL model (Kim model) was consistent with the measured data.

(a) Dittus-Boelter

(b) Colburn

Figure 5 Comparison of HTC correlations for oxide thickness data of E-FUTURE 6301
Figure 5 Comparison of HTC correlations for oxide data of E-FUTURE 6301 (cont’d)

(a) Dittus-Boelter
(b) Colburn
(c) Sieder-Tate
(d) KAERI

Figure 6 Comparison of HTC correlations for oxide data of SELENIUM 1221

(c) Sieder-Tate
(d) KAERI
5. Effect of cladding type

Because the cladding materials of the BR2 test plates were AG3NE (close to AA5754) and AlFeNi, different from AA6061 that the models were based upon, one might expect an effect of cladding type on the oxide growth (see Table 1 of Ref. [13] for the nominal compositions of the cladding types).

The E-FUTURE test results with different cladding types were compared in Figure 7. The E-FUTURE 4111 plate made with AlFeNi showed, in general, lower oxide thickness than those of the E-FUTURE 4202 and 6301 plates made with AG3NE, implying that AlFeNi cladding appears to be slightly advantageous over AG3NE. This result is an outlier compared to the findings in the literature [14], in which no discernable effect was found. The possible reason may be attributed to the higher temperatures for the present test than those in the literature. Like a magnifying glass, the high temperature test magnifies the difference that was not obvious in the previous low temperature tests.

![Figure 7 Examination of the effect of cladding type on oxidation kinetics by using E-FUTURE test plates.](image)

4. Conclusions

The Griess model generally overpredicted for all heat transfer correlations. The ANL model was also found inapplicable to the EUHPRR test conditions at peak power locations when it is coupled with the Dittus-Boelter correlation because the Dittus-Boelter correlation resulted in high cladding temperatures. The ANL model, coupled with the Colburn correlation or the Sieder-Tate correlation, gave most consistent results with the measured oxide data.

The examination of the E-FUTURE test plates revealed that a noticeable difference, albeit small, exists between AG3NE and AlFeNi. AlFeNi appears to result in a slightly thinner oxide layer. However, it was thought that this difference only became significant because the high power test enhanced the oxide growth. For lower power test cases, however, the difference in alloy type was believed to have only a secondary effect, hard to differentiate from other uncertainties.
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