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Aluminum cladding oxidation of prefilmed in-pile fueled experiments

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HIGHLIGHTS

- New experimental data is presented on oxide layer thickness of irradiated aluminum fuel.
- Five oxide growth correlations and four convective heat transfer correlations are used to compute the oxide layer thickness.
- The oxide layer thickness distribution is predicted via correlation for each respective experiment.
- The measured experiment and predicted distributions correlate well, with few outliers.

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ABSTRACT

A series of fueled irradiation experiments were recently completed within the Advanced Test Reactor Full size plate In center flux trap Position (AFIP) and Gas Test Loop (GTL) campaigns. The conduct of the AFIP experiments supports ongoing efforts within the global threat reduction initiative (GTRI) to qualify a new ultra-high loading density low enriched uranium-molybdenum fuel. This study details the characterization of oxide growth on the fueled AFIP experiments and cross-correlates the empirically measured oxide thickness values to existing oxide growth correlations and convective heat transfer correlations that have traditionally been utilized for such an application. This study adds new and valuable empirical data to the scientific community with respect to oxide growth measurements of highly irradiated experiments, of which there is presently very limited data. Additionally, the predicted oxide thickness values are reconstructed to produce an oxide thickness distribution across the length of each fueled experiment (a new application and presentation of information that has not previously been obtainable in open literature); the predicted distributions are compared against experimental data and in general agree well with the exception of select outliers.

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1. Introduction

Recently, a series of in-pile fueled experiments were irradiated in the Advanced Test Reactor (ATR). These experiments, referred to as the Advanced Test Reactor Full size plate In center flux trap Position (AFIP) configuration, were placed in the ATR for the purpose of supporting the qualification of a prototypic ultra-high loading density low enriched uranium-molybdenum alloy fuel (U–10Mo). The qualification of said fuel supports ongoing efforts within the Global Threat Reduction Initiative (GTRI) to convert all civilian Research and Test Reactors (RTRs) from highly enriched uranium to low enriched uranium fuel. The outcome of this experimental study has provided insight regarding the fuel's microstructural stability under extreme conditions to complement existing work that has been completed within the Reduced Enrichment for Research and Test Reactors (RERTR) irradiation campaigns. In addition to the fuel's microstructural state, characterization of the cladding has been performed on samples removed from the ATR. This study details the characterization of cladding oxide growth on the fueled experiments and cross-correlates the empirically measured oxide thickness values to existing oxide growth correlations and convective heat transfer correlations that have traditionally been utilized for such application. This study adds new and valuable empirical data to the scientific community with respect to oxide growth measurements of highly irradiated experiments, of which there is presently very limited data. The outcome of this study provides context toward the limitations of each respective oxide growth correlation and its partner convective

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heat transfer correlation. In general the correlated thickness levels match well with respect to the experimental data with the exception of a few outliers. A summary of the correlations and irradiation experiments (reference tests) that are considered herein is presented in Table 1.

2. Theory

The study of metal corrosion directed toward nuclear reactor applications has become a canonical problem in recent years. Metal corrosion studies have recently centered focus on accident tolerant nuclear reactor cladding and in-core components [1–3] to expand upon the existing fuel and cladding technologies within the nuclear power industry [4]. Although there has been a resurgence of scientific interest in this area, corrosion within the RTR community has been a topic of interest since operation of the earliest reactors [5]. Traditionally, aluminum (of a variety of alloys) has been a common material for RTR cladding. The use of aluminum cladding in a RTR has many benefits including its efficient heat conductance, relatively low specific activity, low thermal neutron absorption, and ease of manufacturability. In contrast, the use of aluminum over other alloys often used as cladding (such as zirconium or stainless steel) delivers a lower melting temperature, and higher rate of corrosion (oxidation). When considering these factors, most low power RTRs need not consider the negative aspects of aluminum, however for those reactors which operate under high power densities such as the U.S. High Performance Research Reactors (HPRRs), the rate of corrosion of aluminum clad relative to that of an operational cycle has potential to impact operations and safety [6]. The oxide layer that forms between the aluminum clad and coolant acts as an insulator due to its relatively low thermal conductivity (e.g. ~2.25 W/m-K [7]). Furthermore, if the Bayerite phase (α-$Al(OH)_3$) – a particularly ‘aggressive’ phase of aluminum oxide – begins to grow on the clad surface versus that of the anticipated Boehmite phase (γ-$AlO(OH)_3$) which is pre-filmed on in-pile aluminum hardware, the cladding surface has a higher likelihood of cyclic corrosion-spalling, leading to further safety related concerns [8].

2.1. Oxide growth

The influence and relation of oxide growth on aluminum has been correlated to a multitude of factors, including the heat flux passing through the oxide layer, fluid water chemistry, fluid flow rates (heat removal and shear forces), radiolysis of the water in the reactor environment, starting characteristics of the prefilmed layer, and initial aluminum surface conditions. Scientists have developed empirical and semi-empirical relations which attempt to predict said growth rate while including some or all of these factors into their study. The first prominently disseminated correlation developed was by Griess et al. through a series of out-of-pile flow tests. This empirical relation was originally developed to support the safety basis for the High Flux Isotope Reactor (HFIR) [9–11], and later expanded to include characteristics which supported design features specific to the ATR [12]. In Griess' study a relation was made through use of a power law

$$\frac{\partial x}{\partial t} = kx^{-p} \tag{1}$$

where x represents oxide thickness (μm), t is time (hr), k is the reaction constant, and p is the rate-law power. For the case of Griess' study, the rate-law power was 0.28535 and reaction constant was empirically found to be

$$k = 1.2539 \times 10^5 \exp\left(-\frac{5913}{T_x}\right) \tag{2}$$

where T_x is the surface temperature (K) of the oxide at the oxide-water interface. Integrating Equation (1) to time t with an assumed initial oxide thickness of x_o yields the generic solution [12].

$$x = \left(x_o^{(p+1)} + (p+1)kt\right)^{\left(\frac{1}{p+1}\right)} \tag{3}$$

or

$$x = \left(x_o^{1.28535} + 1.28535kt\right)^{\frac{1}{1.28535}} \tag{4}$$

for the explicit form of the Griess correlation. Expanding on the work of Griess et al., Hanson developed a relation which assumed that the growth rate of the oxide layer took on an exponential relation with a constant reaction constant, leading to the correlation [5].

$$x = 60.782t^{0.2578} \exp\left(-\frac{2412.5}{T_x}\right). \tag{5}$$

Following the relation developed by Hanson, Kritz identified a significant influence of the reaction constant on heat flux. He utilized the originally derived relation by Griess et al. in Equation (4), but developed a new rate constant relation [13,14].

$$k = 8.686q''^{(1+p)} \exp\left(-\frac{2416.5}{T_x}\right), \tag{6}$$

where q'' represents heat flux (MW/m^2), and p (the rate-law power) assumes the same value found by Griess et al. Following these efforts, design of the Advanced Neutron Source Reactor (ANSR) spurred the need to further expand upon the knowledge base of existing correlations. In the 1990s Pawel et al. published several reports which relate the previously developed relations to the conditions proposed for the ANSR. These relations were then tailored toward the conditions of the ANSR by changing the rate-law power to 0.351 and the reaction constant to [15,16].

Table 1
New empirical data irradiation experiment, heat transfer, and oxide growth correlations.

Irradiation experiment		Heat transfer		Oxide growth	
Reference test	Reference	Correlation	Reference	Correlation	Reference
AFIP-1	[23]	Dittus–Boelter	[18]	Griess	[9–12]
AFIP-2	[24,30]	Petukhov–Popov	[19]	Hanson	[5]
AFIP-3	[25]	Sieder–Tate	[20]	Kritz	[13,14]
AFIP-4	[27]	Hausen	[21]	Pawel	[15,16]
AFIP-6	[26]			Kim	[7,31]
GTL	[28]				