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Modeling the Integrated Performance of Dispersion and Monolithic U-Mo Based Fuels

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ABSTRACT

The evaluation and prediction of integrated fuel performance is a critical component of the Reduced Enrichment for Research and Test Reactors (RERTR) program. The PLATE code is the primary tool being developed and used to perform these functions. Recent code developments and the development direction are discussed here. The code was validated for U_3Si_2 based dispersion fuels. The code is being modified to incorporate the most recent fuel/matrix interaction correlations as they become available for both aluminum and aluminum/silicon matrices. The code is also being adapted to treat cylindrical and square pin geometries to enhance the validation database by including the results gathered from various international partners. Additional modeling work has been initiated to evaluate the thermal and mechanical performance requirements unique to monolithic fuels during irradiation.

1. Introduction

Integrated fuel performance modeling is a critical component of any fuel development program. It can provide a useful context in which to interpret experimental data during the early stages of a program and can provide a predictive tool that demonstrates understanding at the end a program. A wide spectrum of modeling approaches can be applied to any given system but integrated fuel

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performance models are typically born with empirical foundations that gradually evolve into increasingly mechanistic models over time.

It was evident very early in the RERTR fuel development program that a vast majority of the critical fuel performance behaviors were strongly temperature dependent and that the dependencies was often non-linear and strongly coupled. A robust, reliable technique for evaluating the temperature history of various experiments was required in order to interpret any of the data. The PLATE (Plate Lifetime Accurate Thermal Evaluator) code was subsequently developed by Hayes, et al. [1] to supply this information. The code has gradually been upgraded to incorporate as many critical phenomena as possible including fuel phase swelling due to solid and gaseous fission products, density and thermal conductivity changes due to fuel/matrix chemical interaction, fuel particle shape and so on. As additional data continues to come available the PLATE code will be updated and validated.

2. PLATE Code Validation for U_3Si_2 Based Dispersions

The PLATE code was originally designed to model the behavior of plate-type dispersion fuels and is equally well suited to both U-Mo and U_3Si_2 based dispersions. The irradiation behavior of U_3Si_2 dispersions is well known and is therefore a reliable system to benchmark the code against. Correlations available in the literature for fuel-matrix interaction rate between U_3Si_2 and aluminum and the fuel swelling rate of U_3Si_2 were inserted into the code. The following fuel/matrix interaction layer growth rate model is a function of temperature and fission rate and was proposed by Hofman, et al. [2]

$$\Delta y^2 = (0.23)\left(1.286\times10^{-20}\right)\dot{f}_f'''^{0.75} e^{-\frac{7000}{RT}}\Delta t$$

where Δy, $\dot{f}_f'''$, R, T, and Δt are the change in interaction layer thickness, the fuel phase fission density rate (f/cm^3/s), universal gas constant, fuel temperature (K), and time step (s). The fuel phase and reaction product swelling are treated as linearly increasing functions of the fission density [2] as described below

$$\frac{\Delta V_f}{V_{f_i}} = 1.0\times10^{-22} f_f''' \qquad \frac{\Delta V_{rp}}{V_{rp_i}} = 1.0\times10^{-21} f_{rp}'''$$

where ΔV_f, V_{fi}, ΔV_{rp}, V_{rpi}, f_f''', and f_{rp}''' are the change in fuel volume, initial fuel volume, change in reaction product volume, initial reaction product volume, fuel phase fission density (f/cm^3), and the reaction product fission density (f/cm^3).

The analytical results from the PLATE code were compared with measurements from several U_3Si_2 plates irradiated during the RERTR-3, RERTR-4, and RERTR-5 experiments. These experiments covered a wide range of conditions and are summarized in Tables 1 and 2.

Fuel Plate	Fuel Volume Fraction (%)		Reaction Product Volume Fraction (%)		Matrix Volume Fraction (%)	
	Measured	**Calculated**	**Measured**	**Calculated**	**Measured**	**Calculated**
W001	25.2	26.8	2.3	2.6	72.5	70.6
W003	27.8	28.2	4.3	4.3	67.7	66.6
W004	27.5	28.0	4.3	4.0	68.2	67.6
U005	33.9	28.2	6.5	6.1	59.5	64.2
U04	55.1	55.3	19.5	23.3	12.3	11.1
U6008J	48.1	52.3	29.1	27.5	22.5	20.3

Table 1. Measured and Calculated Constituent Volume Fractions

Fuel Plate	Reaction Product Thickness (μm)	
	Measured	Calculated
U005	1.58	U005
U04	3.55	3.71
U6008J	4.74	4.87

Table 2. Measured and Calculated Reaction Product Thicknesses

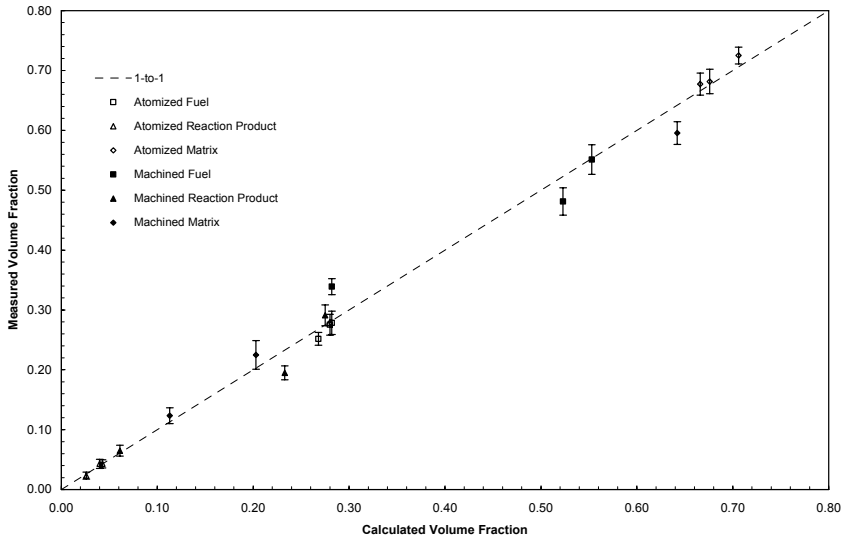

Figure 1. Comparison of measured and calculated volume fractions

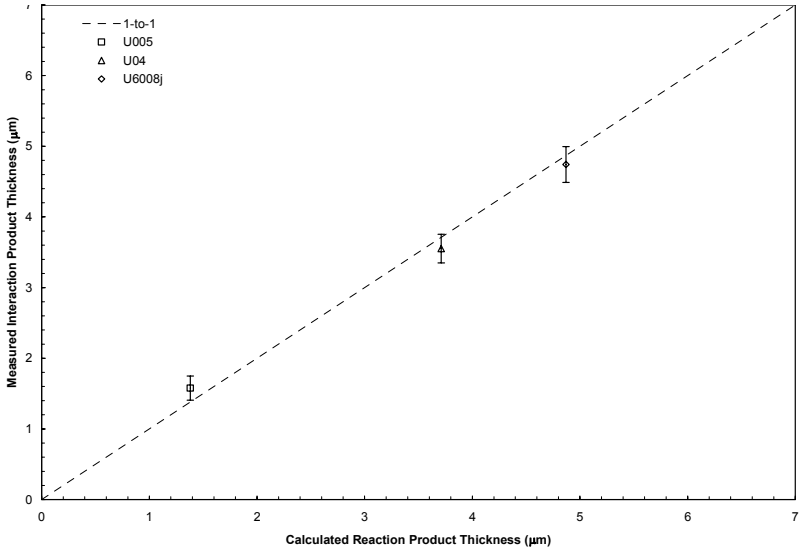

Figure 2. Comparison of measured and calculated reaction product thickness

As seen in Figure 1, the calculated reaction product volume fractions showed excellent agreement with the measured reaction product volume fractions for both atomized and machined fuel plates. The majority of the calculate data points lie within the 95% confidence uncertainty bands for the measured fuel and matrix volume fractions. It appears that atomized fuel particles are in better agreement than the machined fuel particles for volume fraction calculations. Shape factors used by the PLATE code to account for irregularly shape machined fuel geometries contribute to this disagreement. The shape factors are used to enhance the calculated reaction product volume for non-spherical shaped machined fuel particles but a considerable amount of uncertainty is inevitable due the wide variation in geometry from particle to particle. Although the data set is small, good agreement between the measured and calculated reaction product thickness can be seen in Figure 2.

This analysis indicates that the PLATE code is an acceptable tool for prediction the performance of U_3Si_2 dispersion fuels with fuel loadings between 3.1 g/cm^3 to 6.0 g/cm^3, fuel burnups up to 70%, and fission densities as high as 2.4 x 10^{21} fiss/cm^3.

3. Interaction Rate Correlation Development

The presence of an interaction layer between the fuel and matrix in U-Mo based dispersion fuels has a significant effect on the integrated fuel performance. It has been observed in numerous irradiation experiments that the growth of this interaction layer is strongly dependent on the temperature and fission rate. Initial efforts to correlate the experimental data led to the following equation [4] that shows the square of the interaction layer growth is an exponential function of temperature and is linearly related to the fission rate,

$$\Delta y^2 = 5.57 \times 10^{-23}(1.625 - 6.25 \cdot W_{Mo})\exp(-10000/RT) \cdot \dot{f} \cdot \Delta t \ (\text{cm}^2)$$

where Y is the interaction layer growth, R is 1.987 cal/mole-K, T in K, $\dot{f}$ is fission rate (f/cm^3-sec), and Δt is time (sec).

It was recently proposed by Kim, et al. [3] that the fission rate dependence should have square root dependence and should be corrected for the effects of fission damage. It was also noted that the

activation energy in the exponential term is dependent on the composition of the interaction product. Microchemical analysis of a highly reacted plate irradiated by the French [4] indicated that at higher temperature the aluminum content in the interaction product increased as a function of local temperature. The effect appears to be evident when the measured interaction layer thickness Y for a number of experiments is plotted on an Arrehnious plot where one axis is the $\ln\left(Y^2/t\sqrt{\dot{f}Y}\right)$, as shown in Figure 3.

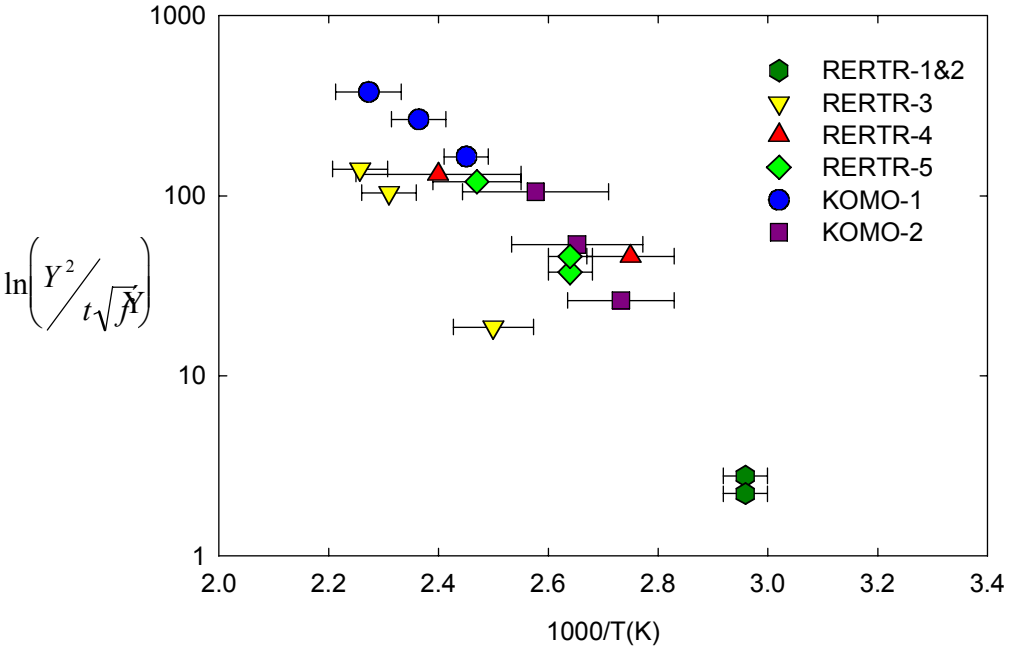

Figure 3. The modified interaction layer thickness measured from several irradiation tests as a function of temperature [5].

Based on these observations a new 2-stage correlation [3] was developed by curve fitting a correlation to low (KOMO-2 494Ni) and high (KOMO-1 428L4) temperature cylindrical fuel pins irradiated in the HANARO reactor by KAERI. The proposed correlations are, for high temperature (> 120°C).

$$\Delta y^2 = 1.46\times10^{-14}\exp(-11000/RT)\cdot(f_D\cdot\dot{f}_Y)^{1/2}\cdot\Delta t \ \ (\mathrm{cm}^2)$$

and for low temperature (< 120°C),

$$\Delta y^2 = 1.94\times10^{-10}\exp(-18000/RT)\cdot(f_D\cdot\dot{f}_Y)^{1/2}\cdot\Delta t \ \ (\mathrm{cm}^2)$$

where Δy is the interaction layer growth, R is 1.987 cal/mole-K, T in K, f_D is fission damage factor, $\dot{f}_Y$ is fission rate (f/cm^3-sec), and Δt is time (sec). This correlation was subsequently incorporated into the PLATE code to compare with the existing RERTR program database for U-Mo/Al based dispersion fuels. Nine plates were analyzed from three different mini-plate experiments. The mini-plate data is summarized in Table 3. The interaction layer thickness predicted using the correlation developed from the KOMO tests is reasonably effective in the low temperature regime, but significantly overpredicts at higher temperatures.

Experiment	Plate ID	Composition	Measured Interaction Layer Thickness (μm)	Predicted Interaction Layer Thickness (μm)	Predicted Peak Fuel Temperature (°C)
RERTR-4	V6001M	U-7Mo/Al	12	11.1	103
	V6022M	U-7Mo/Al	17	33.4	179
	V6015G	U-7Mo/Al	19	34.0	180
RERTR-5	V6018G	U-7Mo/Al	7	8.6	102
	S6007C	U-7Mo/Al	8.4	9.0	99
	S6010D	U-7Mo/Al	12.7	26.5	161
	R6007F	U-7Mo/Al	14	22.3	155
	V6019G	U-7Mo/Al	15	17.1	134
RERTR-6	R5R020	U-7Mo/Al-6061	11.3	16.6	123

Table 3. Interaction product thickness from irradiated RERTR mini-plates and data predicted using the correlation fit to KOMO-1 and -2 data.

Although it appears that the activation energy would be best modeled as a continuous function of temperature, a two stage model should be able to bound the range of activation energies required. An improved fit to the high temperature data was developed by performing a parametric study that varied the pre-exponential term and the activation energy until the results consistently matched the experimental data. The best fit was obtained with the following high temperature (> 120°C) correlation

$$\Delta y^2 = 5.00 \times 10^{-17} \exp(-7000/RT) \cdot (f_D \cdot f^X)^{1/2} \cdot \Delta t \,(\mathrm{cm}^2)$$

The predicted data is listed in Table 4 and compared with the measured data in Figure 4.

Experiment	Plate ID	Composition	Measured Interaction Layer Thickness (μm)	Predicted Interaction Layer Thickness (μm)
RERTR-4	V6001M	U-7Mo/Al	12	11.1
	V6022M	U-7Mo/Al	17	18.2
	V6015G	U-7Mo/Al	19	18.8
RERTR-5	V6018G	U-7Mo/Al	7	8.6
	S6007C	U-7Mo/Al	8.4	9.0
	S6010D	U-7Mo/Al	12.7	14.1
	R6007F	U-7Mo/Al	14	12.9
	V6019G	U-7Mo/Al	15	11.3
RERTR-6	R5R020	U-7Mo/Al-6061	11.3	11.3

Table 4. Comparison of measured interaction product thickness data and the modified correlation.

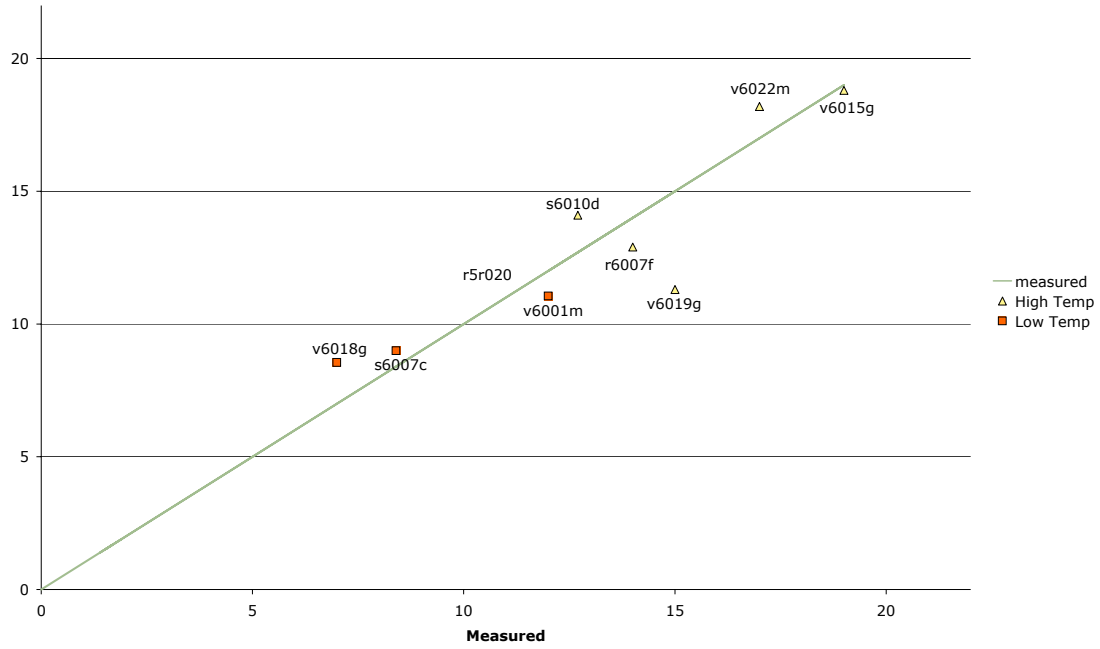

Figure 4. Comparison of measured (with a nominal uncertainty of +/- 1.5 μm) and the predicted interaction layer thicknesses.

This correlation can also be further utilized to evaluate the qualitative impact of adding silicon to the aluminum matrix. Five fuel plates with a silicon bearing matrix from the RERTR-6 experiment were evaluated using the PLATE code. The results are shown in Table 5 and Figure 5. It is clear that the relative growth of the interaction layer is significantly reduced by the presence of greater than 0.2% silicon in the matrix. The R5R020 plate (Al-0.2%Si) shows interaction layer thicknesses similar that of the pure aluminum matrix while the other plates show layers much thinner than predicted. A new correlation for the silicon bearing fuel plates will therefore need to be developed.

Experiment	Plate ID	Composition	Measured Interaction Layer Thickness (μm)	Predicted Interaction Layer Thickness (μm)
RERTR-6	R5R020	U-7Mo/Al-0.2%Si	11.3	11.3
	R3R030	U-7Mo/Al-4043	1.5	10.3
	R2R010	U-7Mo/Al-2.0%Si	1.7	20.3
	R2R020	U-7Mo/Al-2.0%Si	1.4	11.8
	R1R010-7	U-7Mo/Al-6061	1.9	7.8

Table 5. Measured and predicted interaction layer thickness for silicon bearing matrix experiments.

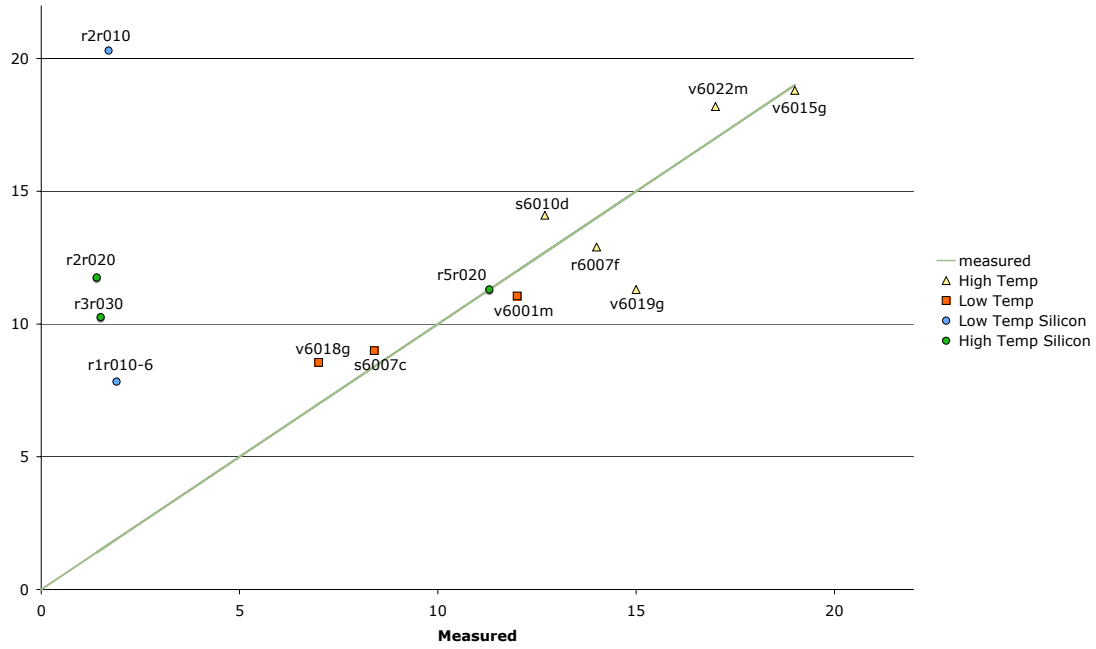

Figure 5. Comparison of predicted Al-Si binary matrix to predictions for pure Al matrix.

4. Modification of the PLATE Code for Pin Type U-Mo Based Fuels

The PLATE code has proven to be a valuable tool in the evaluation and correlation of data gathered from a diverse set of plate-type, U-Mo based fuel experiments. Although the US program is focused on developing a plate-type fuel, several other countries (Canada, South Korea, and Russia for example) are working on a U-Mo alloy based pin-type dispersion fuel. Modification to the PLATE code framework to allow analysis of pin geometries would substantially increase the database used to validate the fuel performance models that are the foundation of the code.

The PLATE code fundamentally solves a simple three-dimensional heat conduction problem. The standard geometry is a slab (i.e. x, y, z directions) within which the thermophysical properties change as a function of time and temperature. The time dependence varies slowly enough that the system can be treated as a string of successive pseudo-steady state problems. The PLATE code solves the problem by starting with an energy balance for a single three-dimensional element. The balance is written as

$$\Delta y \Delta z \left(k \frac{dT}{dx} \right)_{i-\frac{1}{2},j,k} - \Delta y \Delta z \left(k \frac{dT}{dx} \right)_{i+\frac{1}{2},j,k} + \Delta x \Delta z \left(k \frac{dT}{dy} \right)_{i,j-\frac{1}{2},k} - \Delta x \Delta z \left(k \frac{dT}{dy} \right)_{i,j+\frac{1}{2},k}$$
$$+ \Delta x \Delta y \left(k \frac{dT}{dz} \right)_{i,j,k-\frac{1}{2}} - \Delta x \Delta y \left(k \frac{dT}{dz} \right)_{i,j,k+\frac{1}{2}} + \Delta x \Delta y \Delta z \, g_{i,j,k} = 0$$

The derivatives are then expressed in the discrete form and the equation is reduced to

$$A_{i,j,k} T_{i+1,j,k} + B_{i,j,k} T_{i,j+1,k} + C_{i,j,k} T_{i,j,k+1} + D_{i,j,k} T_{i,j,k} + E_{i,j,k} T_{i-1,j,k} + F_{i,j,k} T_{i,j-1,k} + H_{i,j,k} T_{i,j,k-1} + G_{i,j,k} = 0$$

where the coefficients are know values that are indexed after each time step.

To utilize the existing PLATE solver and code infrastructure, a similar equation is required for a cylindrical geometry. The power in a small pin does not vary significantly in the azimuthal direction so the problem can be solved in just two dimensions. The energy balance for an annular control volume is

$$2\pi r_{i+\frac{1}{2}}\Delta z\left(k\frac{dT}{dr}\right)_{i+\frac{1}{2},j}-2\pi r_{i-\frac{1}{2}}\Delta z\left(k\frac{dT}{dr}\right)_{i-\frac{1}{2},j}+\pi\left(r_{i+\frac{1}{2}}^2-r_{i-\frac{1}{2}}^2\right)\left(k\frac{dT}{dz}\right)_{i,j+\frac{1}{2}}$$
$$-\pi\left(r_{i+\frac{1}{2}}^2-r_{i-\frac{1}{2}}^2\right)\left(k\frac{dT}{dz}\right)_{i,j-\frac{1}{2}}+\pi\left(r_{i+\frac{1}{2}}^2-r_{i-\frac{1}{2}}^2\right)\Delta z\, g_{i,j}=0$$

Substituting in the discretized form of the derivatives and collecting on the temperatures leads to the following equation

$$A_{i,j,k}T_{i+1,j,k}+B_{i,j,k}T_{i,j+1,k}+C_{i,j,k}T_{i,j,k+1}+D_{i,j,k}T_{i,j,k}+E_{i,j,k}T_{i-1,j,k}+F_{i,j,k}T_{i,j-1,k}+H_{i,j,k}T_{i,j,k-1}+G_{i,j,k}=0$$

where the coefficients are

$$A_{i,j,k}=\left(\frac{r_i}{\Delta r}+\frac{1}{2}\right)\pi\Delta z\left(k_{i+1,j}+k_{i,j}\right) \qquad B_{i,j,k}=\frac{\pi r_i\Delta r}{\Delta z}\left(k_{i,j+1}+k_{i,j}\right) \qquad C_{i,j,k}=0$$
$$D_{i,j,k}=\left(\frac{r_i}{\Delta r}-\frac{1}{2}\right)\pi\Delta z\left(k_{i,j}+k_{i-1,j}\right) \qquad E_{i,j,k}=\frac{\pi r_i\Delta r}{\Delta z}\left(k_{i,j}+k_{i,j-1}\right) \qquad F_{i,j,k}=0$$
$$H_{i,j,k}=A_{i,j,k}+B_{i,j,k}+C_{i,j,k}+D_{i,j,k}+E_{i,j,k}+F_{i,j,k} \qquad G_{i,j,k}=2\pi r_i\Delta r\Delta z g_{i,j}$$

The *C* and *F* coefficients are always 0 since there is no variation in the aziumuthal (*k*) direction.

To complete the coefficient matrix required to solve the problem, the boundary conditions must be defined. Energy balances are therefore written for each boundary control volume. The cylinder centerline (i=1, 0<j<J), cylinder top, and cylinder bottom (0<i<J, j=1 and J) are adiabatic, the cylinder surface (i=I, 0<j<J) is cooled by convection, and the cylinder corners are exposed to a mixture of these boundary conditions. The coefficients generated from the discretized energy balances can be directly inserted into the PLATE code and will allow analysis of cylindrical pin elements. Complete integration of the two geometries will also require some modest modification to the input and output decks to allow geometry selection and appropriate display of results.

5. Modeling of Monolithic Fuels

The thermal-chemical behavior of a monolithic fuel plate should be very similar to that of the dispersion fuel plates. The PLATE code has already been modified to accommodate the monolithic fuel geometry and specific model development and validation will begin when the first irradiation performance data becomes available from monolithic mini-plates irradiated in the RERTR-6 and -7A experiments. However, the monolithic fuel form poses some additional unique fuel performance questions. The mechanical behavior of the fuel plates will likely be quite different than that of dispersion fuels and the integrity of the bond interface between the fuel and cladding could prove central to the overall fuel performance.

Understanding the bond quality in monolithic fuel is essential to the successful qualification of monolithic fuel plates. As such, the bond quality can be expressed in terms of strength so as to better understand the impact of fabrication processes and/or the variation in parameters that impacts the level

and quality of bonding. Development of a thermo-mechanical model will compliment current studies underway to determine bond strength. The model will provide a basis to understand the loads and corresponding stresses placed on the fuel-cladding interface due to thermal stresses and external loads on the fuel plate prior to irradiation. The model could potentially be expanded to investigate fuel performance during irradiation as a function of bond quality.

An unconstrained, flat plate model has been produced with thermal expansion stresses in order to evaluate shear forces and strains present at the interface. Expected initial and boundary conditions present during irradiation of the flat plates are being evaluated in the current model. Sequentially, the model will be constrained to evaluate the buckling stresses due to thermal expansion. This case will provide information that is more indicative of conditions present in-reactor. In addition, arced plates, with and without constraints, could be investigated to more accurately model the reactor conditions. Specific hydraulic loads experienced in ATR during steady state and transient irradiation could be furnished to the model.

Ultimately, a perfect plate, i.e. a plate with superior bond quality and no observed debonds[2], will be evaluated to determine the model baseline. A series of plates with introduced debonds of various geometrical size and dimension along with varying locations in the fuel plate will be investigated. This will provide initial information both on the thermal and mechanical impacts of debonds present in the fuel plate. The model will be validated using experimental results on bond strength and the impact of debonds under different fabrication processes and conditions.

In this regard, the maximum size of any debonds and minimum strength of the fuel-cladding interface can be determined. A more suitable criterion and specification as to what is acceptable bonding and what is not will be gleaned from the model cases and experimental findings. This specification will be coupled with ultrasonic testing currently used to validate bond quality of monolithic fuel plates after fabrication.

6. Conclusions

The PLATE code is consistently being developed to enable the interpretation of new and old fuel performance data. The code's ability to predict fuel/matrix interaction and net plate swelling was validated against the recent RERTR U_3Si_2 dispersion fuel plate experiment database. Recently proposed U-Mo/Al interaction layer growth rate correlations were evaluated against the RERTR U-Mo/Al database and improvements were suggested for the high temperature correlation. The performance of experimental plates with silicon bearing matrix material from RERTR-6 were predicted with the PLATE code. It was shown that the code predicts substantially higher growth rates for U-Mo/Al fuels than were observed for the Al-Si binary matrix fuels. A new silicon dependent interaction layer growth model is therefore required. A path toward enabling comparative analysis using the PLATE code of plate-type and pin-type U-Mo based fuels was outlined and will be implemented in the near future.

The unique challenges associated with the monolithic fuel form are just becoming evident. Although the PLATE code is appropriate for evaluating the thermal-chemical behavior of the fuel it is expected that thermal-mechanical phenomena will dominate the failure modes. A modeling effort is now underway that will complement experimental work being performed to characterize the

[2] For the purposes of this paper, a debond is defined as a gap or non-bound area between the outer surface of the monolithic fuel and the inner surface of the aluminum cladding

fuel/clad interface. It is anticipated that the progressively more detailed and prototypic analysis will yield the required fuel/clad interface bond strength necessary to prevent failure during irradiation.

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