

MODELING OF HIGH-DENSITY U-MO DISPERSION FUEL PLATE PERFORMANCE

S. L. Hayes^a, G. L. Hofman^b, M. K. Meyer^a, J. Rest^b and J. L. Snelgrove^b

^aArgonne National Laboratory
P. O. Box 2528
Idaho Falls, ID 83403-2528 USA

^bArgonne National Laboratory
9700 South Cass Avenue
Chicago, IL 60439-4803 USA

To be presented at the 2002 International Meeting
on Reduced Enrichment for Research and Test Reactors

November 3-8, 2002
San Carlos de Bariloche, Argentina

The submitted manuscript has been created by the University of Chicago as Operator of Argonne National Laboratory ("Argonne") under Contract No. W-31-109-ENG-38 with the U.S. Department of Energy. The U.S. Government retains for itself, and others acting on its behalf, a paid-up, nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.

Argonne National Laboratory's work was supported by the U.S. Department of Energy, Office of International Policy and Analysis (NA-241) and National Nuclear Security Administration, under contract W-31-109-Eng-38.

MODELING OF HIGH-DENSITY U-MO DISPERSION FUEL PLATE PERFORMANCE

S. L. Hayes^a, G. L. Hofman^b, M. K. Meyer^a, J. Rest^b and J. L. Snelgrove^b

^aArgonne National Laboratory
P. O. Box 2528
Idaho Falls, ID 83403-2528 USA

^bArgonne National Laboratory
9700 South Cass Avenue
Chicago, IL 60439-4803 USA

ABSTRACT

Results from postirradiation examinations (PIE) of highly loaded U-Mo/Al dispersion fuel plates over the past several years have shown that the interaction between the metallic fuel particles and the matrix aluminum can be extensive, reducing the volume of the high-conductivity matrix phase and producing a significant volume of low-conductivity reaction-product phase. This phenomenon results in a significant decrease in fuel meat thermal conductivity during irradiation. PIE has further shown that the fuel-matrix interaction rate is a sensitive function of irradiation temperature. The interplay between fuel temperature and fuel-matrix interaction makes the development of a simple empirical correlation between the two difficult. For this reason a comprehensive thermal model has been developed to calculate temperatures throughout the fuel plate over its lifetime, taking into account the changing volume fractions of fuel, matrix and reaction-product phases within the fuel meat owing to fuel-matrix interaction; this thermal model has been incorporated into the dispersion fuel performance code designated PLATE. Other phenomena important to fuel thermal performance that are also treated in PLATE include: gas generation and swelling in the fuel and reaction-product phases, incorporation of matrix aluminum into solid solution with the unreacted metallic fuel particles, matrix extrusion resulting from fuel swelling, and cladding corrosion. The phenomena modeled also make possible a prediction of fuel plate swelling. This paper presents a description of the models and empirical correlations employed within PLATE as well as validation of code predictions against fuel performance data for U-Mo experimental fuel plates from the RERTR-3 irradiation test.

INTRODUCTION

Results from the postirradiation examination of U-Mo/Al dispersion fuels indicate that the interaction between the U-Mo alloy fuel and Al matrix phases occurs readily during irradiation

and is a sensitive function of temperature [1], as is apparent in Figure 1. As the interaction proceeds, a low-conductivity reaction-product phase builds up, with a corresponding depletion of the high-conductivity Al matrix phase. This leads to a substantial degradation of fuel meat thermal conductivity with time, and fuel centerline temperatures can increase with burnup even if plate power decreases. This strong interrelationship between fuel temperature and fuel-matrix interaction makes the development of a simple empirical correlation between the two difficult, since it is unclear what temperature to employ, and without a correlation for interaction thickness it is impossible to calculate fuel temperatures during irradiation. For this reason, a complex thermal model has been developed to calculate fuel temperatures, taking into account the changing volume fractions of fuel meat constituents, including fuel, matrix, and reaction-product phases within the fuel meat, as well as gas generation/swelling in the fuel and reaction-product phases. Within the context of this best-estimate temperature calculation, an empirical fuel-matrix reaction-rate equation has been developed in an integral way. The resulting interaction rate correlation and other associated behavior correlations and models have been implemented within a computer code designated PLATE, which is now in use to evaluate U-Mo/Al dispersion fuel plate irradiation performance.

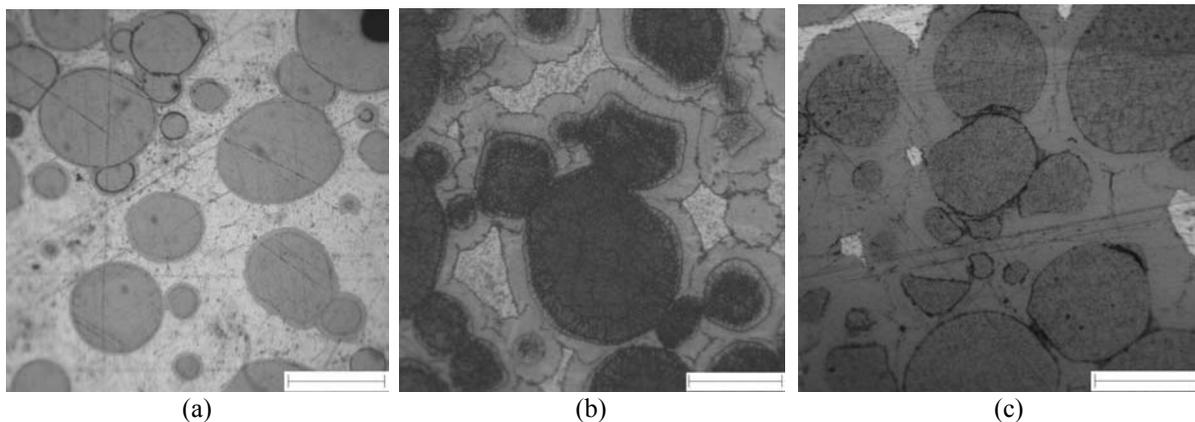


Figure 1. U-Mo/Al fuel-matrix interaction at (a) 145°C, (b) 200°C, and (c) 215°C (scale=100 μ m).

CODE FRAMEWORK

The thermal model within PLATE is based on a steady-state, three-dimensional, control volume-based finite-difference temperature calculation implemented within a FORTRAN computer code. Although the calculation is a steady-state calculation, a series of such calculations are made while marching along in time to simulate irradiation, allowing fuel-matrix interaction to take place based upon an empirical reaction-rate equation. As the reaction-product phase increases and the matrix phase depletes, the effective fuel-meat thermal conductivity is continually modified through the use of an analytical multiphase conductivity model. Combining the changing fuel thermal conductivity and a detailed plate-power history results in a best-estimate fuel temperature calculation made on time-step intervals of typically one day of irradiation on computational nodes throughout the fuel meat. Knowledge of the temperature history within the fuel plate also allows other fuel behavior phenomena to be estimated, including fuel and reaction-product swelling, fission-product gas production and the development of porosity in fuel phases, and the change in fuel plate dimensions. The major elements that contribute to the

PLATE thermal and behavior evaluation are described in the sections that follow. The paper concludes with an initial validation of PLATE calculations versus measured data from the postirradiation examination of the RERTR-3 irradiation test.

THERMAL CONDUCTIVITY

The most important element of the calculations made in PLATE is the evaluation of the fuel meat thermal conductivity. As fuel-matrix interaction proceeds, volume fractions of the meat constituents change significantly. The volume fraction of the low-conductivity reaction product phase is initially zero, but increases during irradiation. Conversely, the high-conductivity matrix phase in the meat decreases from its as-fabricated value as it is consumed by fuel-matrix interaction, by its incorporation into solution with the unreacted fuel alloy, and by extrusion out of the fuel meat region owing to the growth and swelling of the other phases. The fuel-phase mass is also consumed by fuel-matrix interaction; however, the volume of unreacted fuel can increase owing to decreases in density resulting from fission-product swelling and from the incorporation of aluminum into solid solution with the fuel alloy. Also considered are fabrication porosity and fission-product porosity generated during irradiation in the fuel and reaction product phases. Keeping track of constituent masses and densities for each meat control volume allows the change in constituent volume fractions with time to be calculated. This in turn leads to a degradation of the effective fuel-meat thermal conductivity with time/burnup. The effective fuel-meat thermal conductivity can be evaluated at any particular point in time if the constituent volume fractions are known; this is done by using an analytical model for the thermal conductivity of a multiphase material where conductivities of the constituent phases are assumed known.

The multiphase conductivity model employed was derived for a two-phase material from purely theoretical considerations by Hashin and Shtrikman [2]. The matrix aluminum constitutes one phase, and the fuel (assumed to be spherical fuel particles surrounded by a uniform spherical shell of reaction-product) constitutes the other. The uniform reaction-product layer on the surface of the fuel particle produces a thermal resistance to radial heat flow out of the sphere that increases with time as the reaction-product thickness increases. This thermal resistance is calculated analytically and used to decrease the effective fuel-alloy thermal conductivity accordingly. The revised value for the fuel thermal conductivity, which represents both the fuel and interaction-product phases, is then used in the multiphase conductivity model to evaluate the effective meat thermal conductivity.

The analytical model of Hashin and Shtrikman calculates an upper and lower bound to the effective thermal conductivity of the multiphase material. PLATE actually makes use of a modified form of the Hashin and Shtrikman relation that allows for a smooth transition between the upper and lower bounds as a function of the fuel-phase fraction. This modified form was developed by the CEA-Cadarache [3] and is given as:

$$k_{meat} = -f + 3V + 2m - 3Vm + \frac{\sqrt{8fm + (f - 3Vf - 2m + 3Vm)^2}}{4}, \quad (1)$$

where k_{meat} is the effective thermal conductivity of the fuel meat, f is the composite thermal conductivity of the fuel and reaction-product phase, m is the thermal conductivity of the matrix phase, and V is the sum of the volume fractions of the fuel and reaction-product phases. The effective meat thermal conductivity of Eqn. 1 is labeled ‘autocoherent conductivity’ and shown plotted for a U-10Mo/Al dispersion fuel as a function of fuel volume loading in Figure 2. Also shown in the figure are measured thermal conductivities, showing that the model’s predictions are in excellent agreement with measured values.

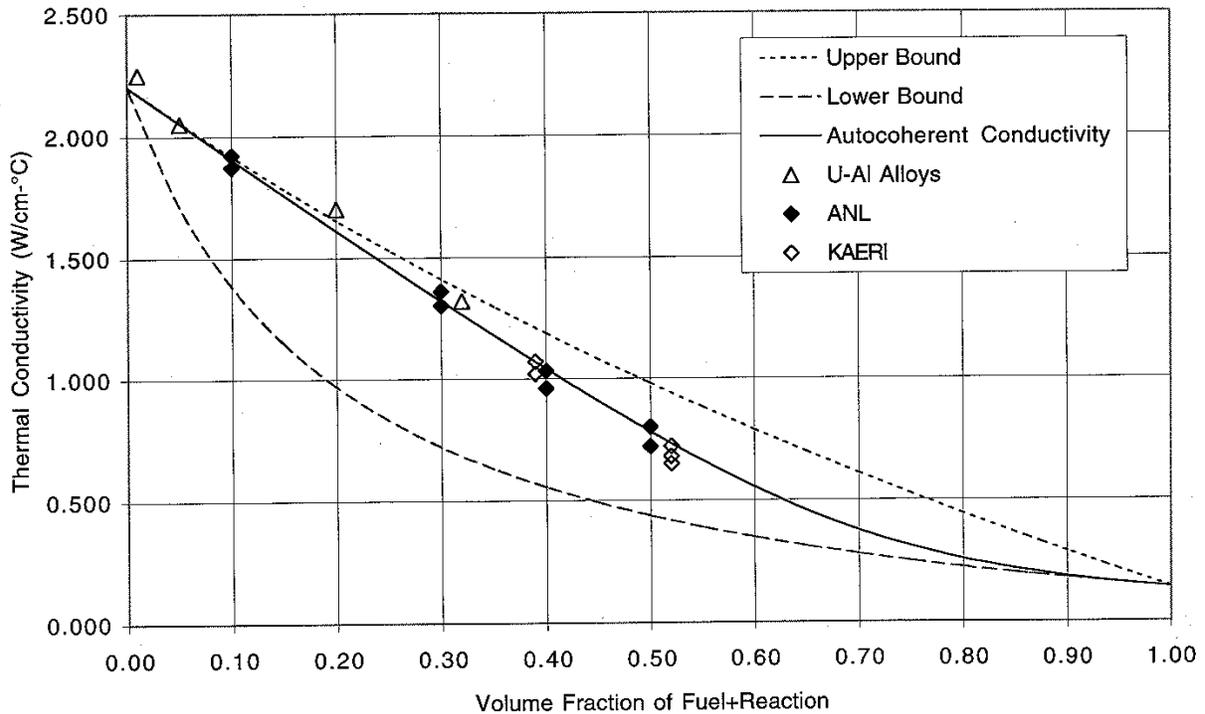


Figure 2. Dispersion fuel thermal conductivity model compared to measured data.

The effect of porosity on thermal conductivity is treated by using a porosity correction factor applied to the base thermal conductivity of the phase that contains the porosity (i.e., the unreacted U-Mo alloy or the aluminide reaction-product). For meat porosity resulting from fabrication, the porosity correction factor is applied to the effective meat thermal conductivity. The porosity correction factor employed in PLATE is [4]:

$$k_p = k_{100} \cdot \exp(-2.14P) , \quad (2)$$

where k_p is the thermal conductivity of the porous materials, k_{100} is the thermal conductivity of the fully-dense material, and P is porosity. This porosity correction factor is valid for porosities below 0.30.

FUEL-MATRIX INTERACTION

The correlation for fuel-matrix interaction thickness used in PLATE takes the same form as the correlation developed by Rest [5] for silicide fuels. A dependence on molybdenum content in the fuel alloy has been added, the activation energy has been changed to a value appropriate for U-Mo fuels [6], and the pre-exponential constant has been changed based on a fit to the interaction thickness data available from postirradiation examination of fuel plates from the RERTR-3 irradiation test [7]. The resulting correlation is:

$$y^2 = 2.2443 \times 10^{-19} \cdot (1.625 - 6.25 \cdot w_{Mo}) \cdot (\dot{f}^{0.75}) \cdot \Delta t \cdot \exp\left\{\frac{-10,000}{RT}\right\}, \quad (3)$$

where y is the fuel-matrix interaction thickness (cm), w_{Mo} is the weight fraction of Mo in the fuel alloy, \dot{f} is the fission rate density in the fuel particle (fissions/cm³-sec), Δt is time (seconds), R is the ideal gas constant (1.987 cal/mole-K), and T is temperature (K).

Using the fuel-matrix interaction thickness calculated from Eqn. 3, reaction-product volumes are calculated. This calculation is made using an exact methodology for spherical fuel particles; for non-spherical fuel particles, the calculation of reaction-product volumes must be enhanced by the use of shape factors based on calculated fuel surface-area-to-volume ratios for various non-spherical fuel particle shapes. The fuel particle shapes must be defined and input to PLATE based on fabrication knowledge. After calculating the reaction-product volume, its mass is calculated via a density relation that is a function of the reaction-product stoichiometry. Masses of fuel and matrix that are consumed in the reaction can then be calculated. The stoichiometry of the reaction-product is not known with certainty, so the input to PLATE calculations allows it to be specified; by default the reaction product is assumed to be (U,Mo)Al₃.

FUEL SWELLING

Fission product swelling in the U-Mo fuel alloy is calculated using an empirical equation developed from postirradiation examination of U-Mo fuel plates from the RERTR-3, -4, and -5 irradiation tests. An increase in the swelling rate for U-Mo fuel has been noted above approximately 2.0×10^{21} fissions/cm³; below this fission density, U-Mo fission-product swelling is approximately 0.4 %/% burnup, and it essentially doubles above this fission density [1,9]; a slight dependence on composition has also been noted. Based on these observations, PLATE calculates the U-Mo fission-product swelling as:

$$\frac{\Delta V}{V} = 5.8336 \times 10^{-23} \cdot (1.25 - 2.5 \cdot w_{Mo}) \cdot f, \quad \text{for } f \leq 2.0 \times 10^{21} \quad (4)$$

$$\frac{\Delta V}{V} = (1.25 - 2.5 \cdot w_{Mo}) \cdot \left[0.1167 + 1.1667 \times 10^{-22} \cdot (f - 2.0 \times 10^{21})\right], \quad \text{for } f > 2.0 \times 10^{21} \quad (5)$$

where $\Delta V/V$ is the fuel swelling for the U-Mo alloy, w_{Mo} is the weight fraction of Mo in the fuel alloy, and f is the fission density in the fuel particle (fissions/cm³). Twenty-five percent of the fuel swelling is assumed to be due to gas bubbles, which are treated as porosity that develops within the alloy and leads to a degradation of the base U-Mo thermal conductivity.

Fission-product swelling of the aluminide reaction-product is calculated as:

$$\frac{\Delta V}{V} = 4.0 \times 10^{-23} \cdot f . \quad (6)$$

Swelling of the U-Mo fuel alloy is also enhanced by the incorporation of matrix aluminum into solution with the unreacted fuel alloy. This phenomenon has been observed during postirradiation examination [7], and the effect this has on decreasing the fuel-phase density (and increasing its volume) appears to be an important phenomenon because of its effect on the calculation of the meat effective thermal conductivity.

Finally, by keeping track of volume changes in the fuel meat owing to the accumulation of the low-density reaction-product phase, depletion of the matrix phase, and fuel swelling, one can estimate the change in the final dimensions of the fuel plate. PLATE assumes that owing to constraint in the lateral and longitudinal dimensions of the fuel plate, volume increases result in increases in the fuel plate thickness only.

CONTACT RATIO

The use of a contact ratio for those phenomena that depend on the available surface area between the fuel and matrix phases is an important calculational detail. The surface area available for fuel-matrix interaction decreases as the matrix phase depletes. To account for this effect, the as-fabricated specific surface areas for fuel particles in each fuel control volume are updated at the beginning of each time-step by multiplication with a semi-empirical contact ratio correlation [8]:

$$R = 1 - 0.72 \cdot V + 1.50 \cdot V^2 - 1.78 \cdot V^3 , \quad (7)$$

where R is the contact ratio and V is the sum of the volume fractions of the unreacted fuel and reaction-product phases.

MATRIX EXTRUSION

During the development of the PLATE dispersion fuel performance code, an apparent bias toward over-predicting the end-of-life matrix phase volume fractions in the fuel meat was apparent. It has been postulated that this is due to extrusion of matrix aluminum out of the fuel meat region during irradiation, caused by the in-growth of the brittle reaction-product phase and fuel swelling. A simple model is used to calculate the extrusion of a contiguous matrix phase out of the center regions of the fuel plate and toward the fuel surface. The extrusion model is allowed to operate within any given meat control volume as long as the matrix phase is contiguous, currently defined as a matrix volume fraction of 0.25 or greater; once this limit on matrix volume fraction is reached, it is assumed that the remainder of the matrix phase is pinned and no further extrusion is allowed.

CODE VALIDATION

Results of the PLATE code validation versus measured data for U-Mo fuel plates from the RERTR-3 irradiation test [7] are shown in Figures 3 to 5. In these figures, measured quantities obtained from postirradiation examination are shown plotted versus the same quantities as calculated by PLATE. Calculated fuel-matrix interaction thicknesses are in excellent agreement with measured thicknesses; assuming the form of the interaction thickness correlation (Eqn. 3) is correct, this is not surprising since the pre-exponential term in that correlation was selected as a best-fit to this data. Figures 4 and 5 show that calculated fuel meat constituent volume fractions and fuel plate thickness increases are in good agreement with measured values.

Agreement between calculated and measured meat constituent volume fractions tends to be much better for fuel plates fabricated from spherical fuel particles. For this fuel particle shape, the geometrical calculations are performed using an exact methodology. The required use of shape factors for geometrical calculations associated with non-spherical fuel particles introduces considerable uncertainty into the resulting constituent phase fractions calculated by PLATE. This is further complicated by the difficulty in characterizing the fuel particle size and shape distributions for non-spherical fuel powder. Thus, it is no surprise that agreement between calculated and measured volume fractions is best for the plates fabricated using spherical fuel particles.

Figure 5 indicates an apparent bias toward over-prediction of the fuel plate thickness increase for plates using atomized (i.e., spherical) fuel particles. These fuel plates have almost no fabrication porosity in the fuel meat, so a very low value is used as input to PLATE calculations, typically 1 to 2 %. This low value of porosity is difficult to measure in finished fuel plates, and increasing the specified fabrication porosity used in the calculations by no more than one percent could bring the results into excellent agreement with the measurements. The calculations for the fuel plates fabricated from machined fuel powder (i.e., non-spherical) will always suffer from the inaccuracies introduced from the use of shape factors in the geometrical calculations.

As a representative example, Figure 6 shows the results of the PLATE calculation for one of the experimental fuel plates, V03 from the RERTR-3 irradiation test. V03 was a fuel plate fabricated using atomized U-10Mo fuel powder loaded to 8 g-U/cm^3 in an Al matrix and irradiated in one of the higher power positions in the RERTR-3 test vehicle. RERTR-3 underwent two irradiation cycles in the Advanced Test Reactor, lasting 34 and 14 effective full-power days, respectively. The behavior of the peak fuel-meat temperature shown in the figure mirrors the plate power history, which changes throughout the power cycles due to the near-continuous rotation of control drums located on the periphery of the reactor, near the RERTR-3 irradiation position. The substantial change in effective fuel-meat thermal conductivity with time/burnup as calculated by PLATE is also shown in the figure.

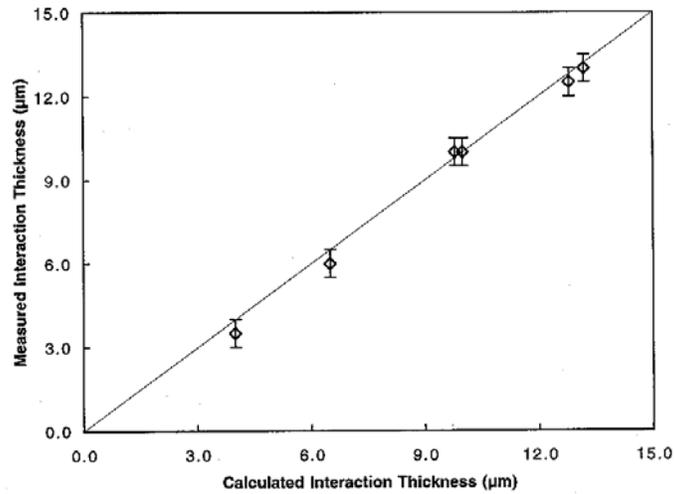


Figure 3. Comparison of measured and calculated fuel-matrix interaction thicknesses.

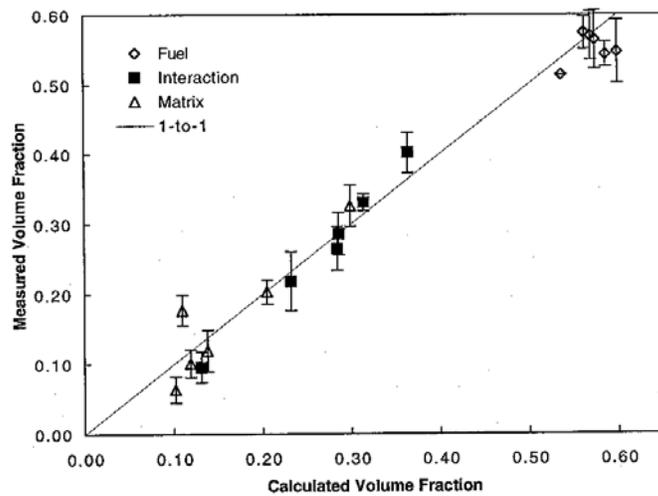


Figure 4. Comparison of measured and calculated fuel meat constituent volume fractions.

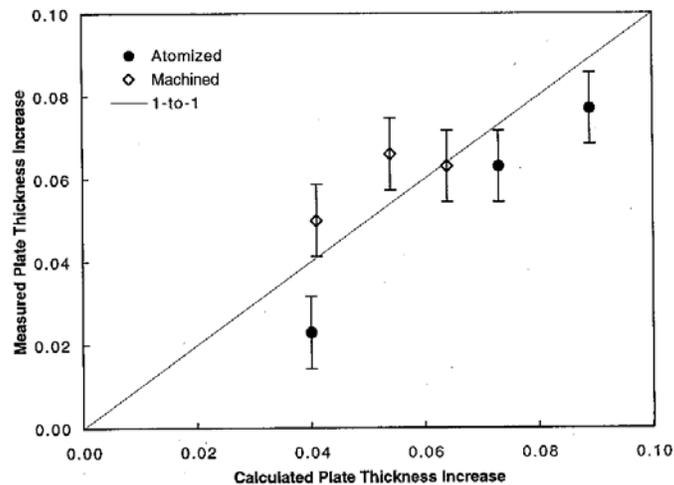


Figure 5. Comparison of measured and calculated fuel plate thickness increases.

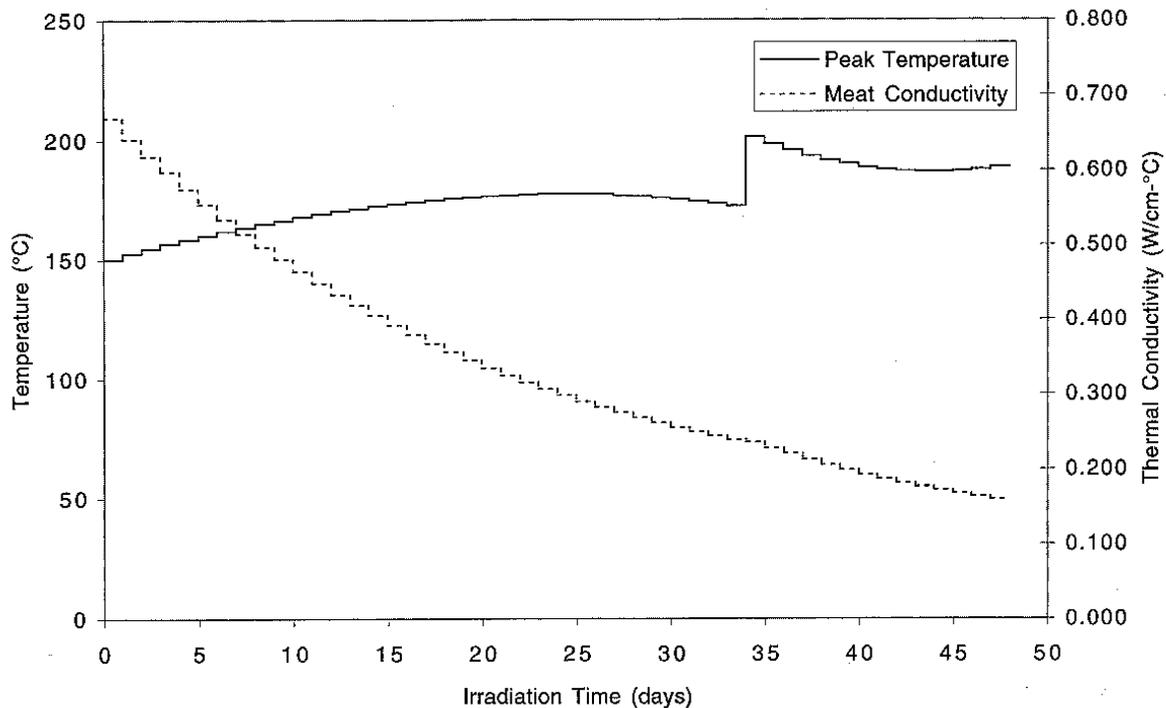


Figure 6. Thermal history of fuel plate V03 from RERTR-3 as calculated by PLATE.

CONCLUSION

The development of a plate-type dispersion fuel thermal analysis code was initiated in order to provide an integrated framework within which to evaluate many interdependent fuel behavior phenomena. Ultimately, the capability to calculate meaningful, best-estimate fuel temperatures during irradiation was the goal. The PLATE code that has resulted from this development effort has achieved this goal.

While many of the behavior phenomena are treated empirically in PLATE, the heart of the calculation is based on an evaluation of the fuel meat thermal conductivity that is obtained from a sound analytical model. The good agreement of code calculations with fundamental parameters measured during the postirradiation examination of U-Mo/Al fuel plates from the RERTR-3 irradiation test give confidence that the temperatures calculated by PLATE are sound.

REFERENCES

- [1] G. L. Hofman, J. L. Snelgrove, S. L. Hayes, and M. K. Meyer, "Progress in Development of Low-Enriched U-Mo Dispersion Fuels," Proceedings of the 2002 International Topical Meeting on Research Reactor Fuel Management, Ghent, Belgium, 17-20 March 2002.

- [2] Z. Hashin and S. Shtrikman, "A Variational Approach to the Theory of the Effective Magnetic Permeability of Multiphase Materials," Journal of Applied Physics, Vol. 33 (1962) pp. 3125-3131.
- [3] David Plancq, Private Communication, CEA-Cadarache, France (2002).
- [4] M. E. Cunningham and K. L. Peddicord, "Heat Conduction in Spheres Packed in an Infinite Regular Cubical Array," International Journal of Heat and Mass Transfer, Vol. 24 (1981) pp. 1081-1088.
- [5] J. Rest, "A Theoretical Model for the Irradiation-Enhanced Interdiffusion of Al and U Alloys," Proceedings of the 2000 International Meeting on Reduced Enrichment for Research and Test Reactors, Argonne National Laboratory Report, ANL/TD/TM01-12, July 2001, pp. 146-158.
- [6] K. H. Kim, H. J. Kwon, J. S. Lee, H. J. Ryu, J. M. Park, and C. K. Kim, "An Investigation on the Fuel/Matrix Reaction Behavior of U-Mo/Al Dispersion Fuels Prepared with Centrifugal Atomization," Proceedings of the 2000 International Meeting on Reduced Enrichment for Research and Test Reactors, Argonne National Laboratory Report, ANL/TD/TM01-12, July 2001, pp. 285-295.
- [7] M. K. Meyer, G. L. Hofman, R. V. Strain, C. R. Clark, and J. R. Stuart, "Metallographic Analysis of Irradiated RERTR-3 Fuel Test Specimens," Proceedings of the 2000 International Meeting on Reduced Enrichment for Research and Test Reactors, Argonne National Laboratory Report, ANL/TD/TM01-12, July 2001, pp. 201-213.
- [8] J. Gurland, "An Estimate of Contact and Continuity of Dispersions in Opaque Samples," Transactions of the Metallurgical Society of AIME, Vol. 236 (1966) pp. 642-646.
- [9] G. L. Hofman and M. K. Meyer, "Progress in Irradiation Performance of Experimental Uranium-Molybdenum Dispersion Fuel," Proceedings of the 2002 International Meeting on Reduced Enrichment for Research and Test Reactors, 3-8 November 2002, Bariloche, Argentina.