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PROGRESS IN QUALIFYING LOW-ENRICHED U-MO DISPERSION FUELS

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ABSTRACT

The U.S. Reduced Enrichment for Research and Test Reactors program is working to qualify dispersions of U-Mo alloys in aluminum with fuel-meat densities of 8 to 9 gU cm^{-3}. Postirradiation examinations of the small fuel plates irradiated in the Advanced Test Reactor during the high-temperature RERTR-3 tests are virtually complete, and analysis of the large quantity of data obtained is underway. We have observed that the swelling of the fuel plates is stable and modest and that the swelling is dominated by the temperature-dependent interaction of the U-Mo fuel and the aluminum matrix. In order to extract detailed information about the behavior of these fuels from the data, a complex fuel-plate thermal model is being developed to account for the effects of the changing fission rate and thermal conductivity of the fuel meat during irradiation. This paper summarizes the empirical results of the postirradiation examinations and the preliminary results of the model development. In addition, the schedule for irradiation of full-sized elements in the HFR-Petten is briefly discussed.

1. Introduction

For the past several years the focus of the fuels area of the U.S. Reduced Enrichment for Research and Test Reactors (RERTR) program has been the development of aluminum-based dispersion fuels that will accommodate uranium densities in the fuel meat of 8 to 9 gU cm^{-3} [1]. Our primary focus has been on determining the irradiation behavior of candidate fuels. Thus far, data are available from three irradiation tests of very small fuel plates--RERTR-1, -2, and -3. The first two tests resulted in the identification of U-Mo alloys with Mo contents of at least 6 wt.% as very promising candidates [2]. The third test, which is the principal subject of this paper, was focused on the behavior of the U-Mo fuels under high-temperature (up to ~250°C) irradiation conditions.

The small test plates irradiated in RERTR-3 contained either atomized or machined fuel particles ranging in composition from, nominally, 6 wt.% Mo (U-6Mo) to 10 wt.% Mo (U-10Mo); actual compositions ranged from 6.7 to 10.6 wt.% Mo. Of the plates discussed in this paper, one set was fabricated using either atomized or machined U-10Mo powder that had been given a solution heat treatment in the gamma phase; all other plates discussed in this paper were fabricated using powder in the as-fabricated state. The fuel plates in the RERTR-3 test measured 10.0 mm x 41.1 mm x 1.52 mm; the meat was in an elliptical zone nominally 0.76 mm thick and contained, nominally, 8 gU cm^{-3} of fuel meat. The actual average uranium densities in the plates were probably somewhat higher, and uranium densities in some areas of the plates were significantly higher.

2. Postirradiation Data

Because of the small size of the test plates, volumetric measurement with the customary immersion method would not yield swelling data with sufficient accuracy; therefore swelling was determined from plate thickness measurements. The measured plate-thickness increases are shown in Fig. 1, normalized to a meat fission density of 10^{21} cm^{-3} (~30% burnup), as a function of beginning of life (BOL) fuel centerline temperature. These BOL temperatures were calculated using a one-dimensional (1-D) heat transfer model. It is clear from Fig. 1 that the plate swelling is a strong function of temperature and that there appears to be no clear difference, within the measurement uncertainty, among the various compositions. It is important to note at this point that these thickness increases are quite small, and since thickness measurements always overestimate the swelling, the maximum fuel-meat swelling at this burnup is no more than 10% at the highest temperatures tested.

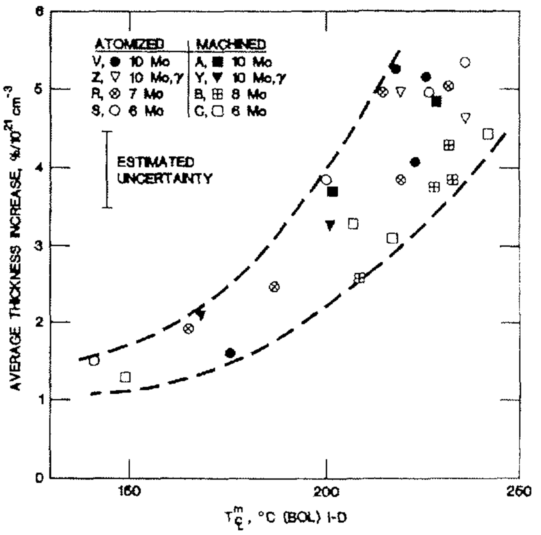


Fig. 1. Thickness increase of RERTR-3 test fuel plates normalized to a fuel-meat fission density of 10^{21} cm^{-3} (30% burnup) vs. calculated BOL peak fuel meat temperature.

The 1-D BOL temperatures plotted in Fig. 1 only serve to illustrate the trend in swelling with increasing temperature. The actual fuel temperature is a complex function of irradiation time and position in the fuel meat. Not only are there substantial temperature gradients in the fuel meat, but the temperature changes during the irradiation as a result of the competing effects of decreasing thermal conductivity and U-235 burnup. This issue is treated in detail later in this paper.

Two irradiation effects singly, or in combination, may be responsible for the observed temperature dependence of the swelling. One is the behavior of fission gas in the U-Mo alloy (the swelling owing to solid fission products can be considered to be athermal); the other is irradiation-enhanced interdiffusion of the fuel and the matrix aluminum. Inspection of the optical micrographs shown in Fig. 2 suggests that the latter is the major contributor to the temperature dependence of the swelling. Note

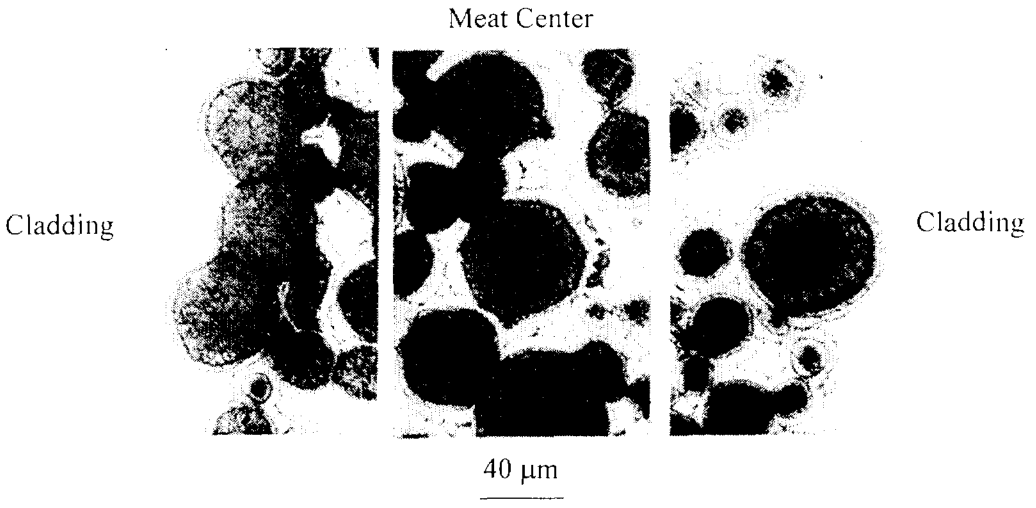
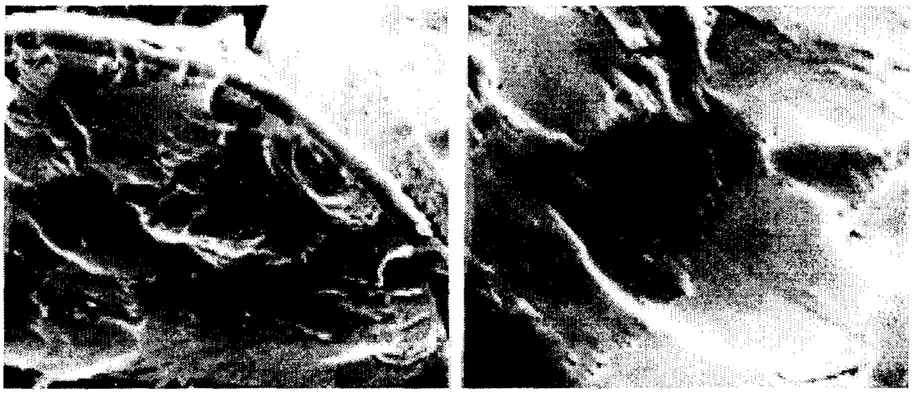


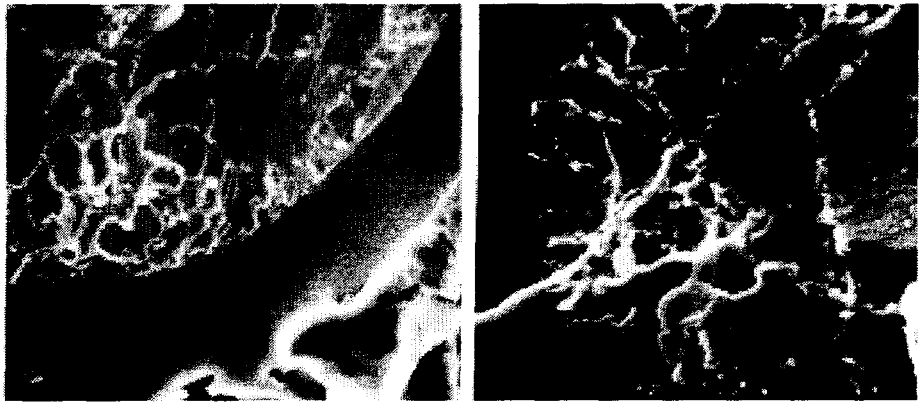
Fig. 2. Optical micrographs of sections of plate V03 taken near the center of the plate showing the change in interaction-layer thickness from the cladding/fuel-meat interface to the center of the fuel meat.

the large increase in interaction thickness when going from a fuel temperature of 175°C (1-D BOL) at the cladding interface to 217°C at the center of the meat in U-10 Mo. It should also be noted that although virtually all of the matrix aluminum near the center of the plate has been consumed, resulting in a mass of reacted and unreacted fuel, there is no evidence of instability of this fuel mass.

There are indications of the presence of small gas bubbles at the grain boundaries in the unreacted fuel. This is better illustrated in the SEM fractographs shown in Fig. 3. Small gas bubbles have begun to form in the 175°C sample and are more numerous and larger at 217°C. However, the 175°C sample reached only 30% burnup compared to 40% for the 217°C sample. Comparison of the bubble morphology in this latter sample with that of the same fuel irradiated previously in RERTR-1 to the



30% Burnup, 175°C



40% Burnup, 217°C

40% Burnup, 65°C

Fig. 3. Fuel microstructure of U-10Mo at low and high temperature, showing apparent athermal fission gas behavior.

same burnup, but at a much lower temperature of ~65°C, leads us to conclude that there is no effect of temperature on the fission gas behavior over the temperature and burnup range tested thus far, and that the temperature dependence of the measured plate swelling is the result of fuel-aluminum interdiffusion. Fission-gas bubbles begin to form on grain boundaries somewhere below 30% burnup, and between 30 and 40% burnup fission-induced grain refinement starts, providing additional grain boundaries for fission-gas precipitation. This process, which has been described in detail previously [3], will progress with further burnup, eventually covering the entire fuel particle.

The evolution of the unreacted fuel microstructure does appear to be a function of composition, as shown by a comparison of U-10Mo and U-6Mo in Fig. 4. Grain refinement has evidently started at a lower burnup in the U-6Mo sample and has already covered significant fractions of the grains. All fuel compositions are expected to develop microstructures like those shown in Fig. 4 for the

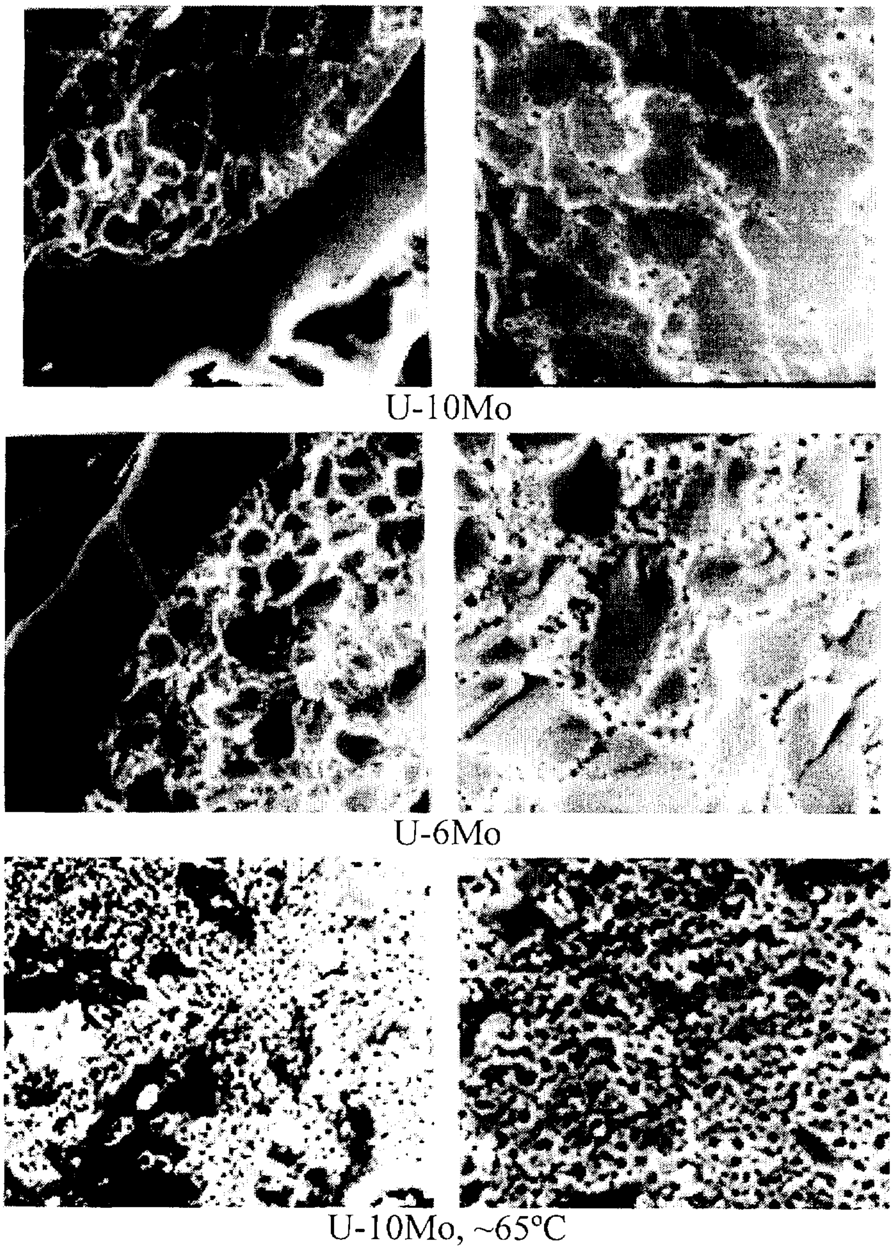


Fig. 4. Comparison of fuel microstructures of U-10Mo and U-6Mo at ~40% burnup irradiated at T>200°C, and at 70% burnup irradiated at ~65°C.

low-temperature, 70%-burnup sample from RERTR-2, but the lower-Mo compositions will complete their restructuring at lower burnup and should therefore have a somewhat higher fuel-swelling rate.

In summary, we have observed through metallographic examinations that (1) fuel plate swelling is stable and modest, (2) overall swelling owes predominantly to temperature-dependent U-Mo/Al interdiffusion up to the burnup where the matrix aluminum is fully consumed by this interdiffusion process, (3) the aluminide interaction product appears stable and contains no fission gas bubbles, (4) the swelling behavior of the unreacted fuel appears to be athermal in the range of burnup and temperature tested, and (5) reducing the molybdenum content of the alloy results in somewhat higher rates of interdiffusion and fission gas swelling. Although not discussed in this paper, we have also shown that small ternary additions of other elements to the alloy, shown to reduce the rate of thermally driven interdiffusion, do not reduce the rate of irradiation-induced interdiffusion.

3. Fuel Plate Thermal Analysis

3.1 Motivation

As described above, results from postirradiation examinations of U-Mo alloy dispersion fuels indicate that the interaction between the fuel and matrix phases is apparently a sensitive function of temperature. As the interaction proceeds, a low-conductivity reaction-product phase builds up, while at the same time the high-conductivity aluminum-matrix phase is depleted. Thus, fuel temperatures are likely to increase with burnup even if plate powers decrease. This interplay 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. For this reason, a complex thermal model was developed to calculate fuel temperatures, taking into account the changing volume fractions of fuel, matrix, and reaction-product phases within the fuel meat, as well as gas generation/swelling in the fuel phase. Then, within the context of this best-estimate temperature calculation, a fuel-matrix reaction-rate equation can be developed in an integral manner.

3.2 Model Description

The thermal model developed is based upon 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 (while under irradiation), allowing fuel-matrix interaction to proceed based upon a physics-based reaction-rate equation; a correlation developed for U_3Si_2/Al interaction was taken as an initial guess [4]. As the reaction-product phase increases and the matrix phase depletes, the effective fuel-meat thermal conductivity is changed through the use of an analytical multiphase conductivity model [5]. The multiphase conductivity model employed was derived for a two-phase material from purely theoretical considerations. The matrix aluminum constitutes one phase and the fuel, consisting of the spherical fuel particles surrounded by a uniform spherical shell of reaction product, constitutes the other. The reaction product produces a thermal resistance to radial heat flow out of the spheres that increases with time as the reaction-product thickness increases. This resistance is calculated directly and used to decrease the fuel alloy thermal conductivity accordingly. The revised value for the fuel thermal conductivity, representing both the fuel and interaction-product phases, is then used in the multiphase conductivity model to reevaluate the effective fuel-meat thermal conductivity.

As fuel-matrix interaction proceeds, volume fractions of the meat constituents change. The volume fraction of the reaction product is initially zero, but increases with irradiation time/burnup. The matrix phase decreases from its as-fabricated value as it is consumed both by the fuel-matrix reaction and by incorporation into solution with the 'unreacted' fuel alloy. The incorporation of matrix aluminum into solution with the 'unreacted' fuel alloy kernels has been observed during the postirradiation examinations [6], and the effect this has on decreasing the fuel-phase density (and increasing its volume) is an important phenomenon because it affects the calculation of the meat effective thermal conductivity. The fuel-phase mass is consumed by the fuel-matrix reaction; however, the volume of 'unreacted' fuel actually increases owing to swelling and to the incorporation of aluminum into solid solution. Keeping track of constituent masses and densities for each meat control volume allows the change in volume fractions with time to be calculated. This in turn leads to a degradation of the effective meat thermal conductivity with time/burnup. 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 at various locations in the fuel meat.

3.3 Model Results

The fuel-plate thermal model was employed to perform preliminary calculations for two U-10Mo/Al fuel plates irradiated in the Advanced Test Reactor (ATR) as part of the RERTR-3 irradiation experiment [7] and subsequently examined destructively to quantify the extent of fuel-matrix interaction [6]. The two fuel plates, V03 and V07, were subjected to significantly different plate powers, and the extent of fuel-matrix interaction observed during postirradiation examination differed significantly between the two plates. Both of these fuel plates were fabricated using as-fabricated spherical U-10Mo fuel powder produced by the Korea Atomic Energy Research Institute (KAERI) by centrifugal atomization.

Figure 5 shows the detailed power histories for the two fuel plates as calculated using the ATR whole-core MCNP model. Also shown are the peak fuel temperatures calculated using the thermal model. It is apparent from this figure why simply using a calculated beginning-of-life fuel temperature would be inadequate for the purposes of correlating fuel-matrix interaction. It is interesting to note that the calculated fuel-meat thermal conductivity of plate V03 decreased during irradiation from ~40 W m^{-1} K^{-1} at BOL to ~7.5 W m^{-1} K^{-1} at the end of the irradiation.

Figure 6 shows the fuel-matrix interaction thicknesses calculated by the thermal model, along with the measured data. The silicide-based interaction rate correlation has an Arrhenius dependence upon temperature with an activation energy of 8,520 cal/mol (35,670 J/mol). This activation energy was not changed, but the pre-exponential term was reduced by approximately 25% to obtain the agreement shown with the thickness data for V03. The calculation results shown for V07 were

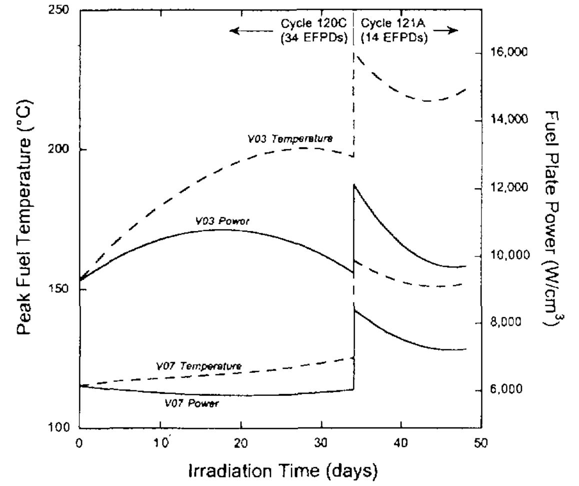


Fig. 5. Input power histories and calculated temperature histories of plates V03 and V07.

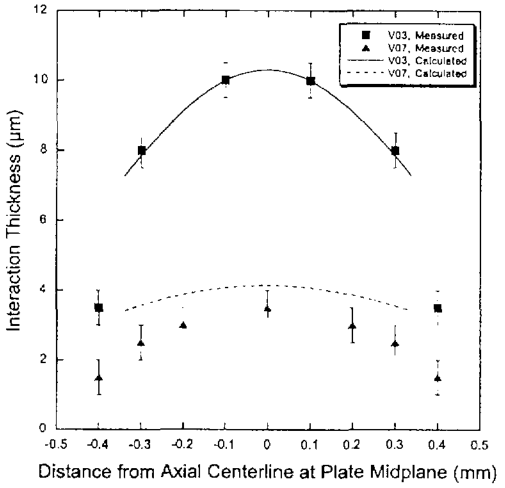


Fig. 6. Comparison of measured and calculated interaction-layer thickness profiles of plates V03 and V07.

obtained using the same parameter set as for V03, resulting in a slight overprediction of reaction thicknesses.

It must be emphasized that the calculations shown are preliminary in nature. Several aspects of the model are still under development and validation against a much larger set of data must be undertaken. Nevertheless, the qualitative agreement of these preliminary results with observed irradiation performance is encouraging.

4. Qualification Irradiations

The analysis of the RERTR-3 test discussed above is an important step in the qualification of the U-Mo fuels. However, since the RERTR-3 test was limited to burnups below 40% and because fuel-meat swelling determined from plate thickness measurements or by quantitative metallography has a large uncertainty, two additional irradiation tests are underway in the ATR–RERTR-4 and RERTR-5. These tests utilize larger, though still miniature, fuel plates so that the fuel-meat volume increase can be determined by the immersion method. The temperatures will be somewhat lower than those of RERTR-3, but the burnups achieved will be considerably higher--up to ~50% for RERTR-4 and up to ~80% for RERTR-5. Irradiation of RERTR-4 was completed in early January, and it will be shipped to ANL-East (Illinois) during the first half of March for postirradiation examination. Irradiation of RERTR-5 will be completed in the July-August 2001 time period, and its examination is expected to begin during November. A good capability to predict irradiation behavior of U-Mo fuels will result from the analyses of the RERTR-X tests, and the data obtained will be the basis for qualification.

As discussed at RRFM 2000, the RERTR program is also planning to irradiate several full-sized fuel elements as part of the qualification of both fuel and fabricator [8]. Unfortunately the schedule for the start of the first irradiations in the HFR-Petten, of 6-gU cm^{-3} U-7Mo fuel fabricated by BWX Technologies (BWXT) using atomized powder produced by KAERI, has been delayed by at least six months from that shown previously owing to commercial uncertainties resulting from a KAERI patent claiming protection of all dispersions using spherical U-Mo fuel particles. We hope finally to resolve this problem in early March, and a progress report and an updated schedule will be presented during the oral presentation of this paper on April 2.

In the meantime, the Argentine Comisión Nacional de Energía Atómica (CNEA) has requested to join the RERTR program's U-Mo fuel element qualification program. Discussions are currently underway regarding fabrication by the CNEA and irradiation in the HFR of one fuel element with a fuel-meat density of 7 gU cm^{-3}.

5. Conclusions

The postirradiation examinations of the RERTR-3 fuel plates have provided a wealth of information about the swelling of U-Mo fuel meat. The swelling is dominated by the fuel-aluminum interaction, and there is no indication that even complete depletion of the aluminum matrix will result in unstable swelling behavior. A complex fuel-plate thermal model is under development that should allow us to derive quantitative information on the interaction correlation and on the thermal conductivity as a function of the plate's thermal and burnup history. This model, when completed, can be used with externally supplied fission-rate histories and fission-product fuel-swelling correlations to predict the irradiation behavior of U-Mo fuel plates under actual irradiation conditions.

All of the information obtained so far from the RERTR program's irradiations indicate that the U-Mo fuel will perform well in research reactors. Additional irradiations are underway, and a series of full-sized fuel element irradiations is planned. Although the start of the irradiation of 6-gU cm^{-3} U-7Mo fuel elements in the HFR has been delayed, we still hope to be able to issue a qualification report by the end of CY 2003.

6. References

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