OBSERVATIONS OF THE NUCLEATION AND EVOLUTION OF POROSITY IN U-Mo FUELS

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Argonne National Laboratory
Nuclear Engineering Division
Argonne, Illinois 60439

*On assignment from the
Australian Nuclear Science & Technology Organisation (ANSTO)
PMB 1 Menai NSW 2234 – Australia

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ABSTRACT

The irradiation of UMo fuel dispersed in a matrix of aluminum results in the formation of a reaction product that coats the fuel particle surface. In some instances pores form at the interface between the reaction product and the aluminum matrix. Depending on the irradiation conditions the pores may grow, link-up to form larger pores and in severe cases form a continuous network that results in unacceptable pillowing of the fuel plate. Such observations have been made in US and other international irradiation experiments. Evaluation of the irradiation conditions and the nature of porosity formation in these experiments permit us to draw some conclusions on the influence of various parameters such as fission density, fission rate, temperature and composition. In particular, the irradiation experiments RERTR-4 and RERTR-5, conducted in the Advanced Test Reactor (ATR) are reviewed.

I. Introduction

Post irradiation examination of mini-plates irradiated in the ATR in experiments RERTR-4 and 5 revealed the formation of porosity in the fuel meat. Porosity was found to occur in two discrete locations (Figure 1). This pattern was repeated in every mini-plate observed to contain porosity. The irradiation conditions and power histories from the RERTR-4 and 5 experiments have recently been finalized and now permit a more quantitative assessment of the influence of various irradiation parameters such as fission density, fission rate and temperature on porosity formation and growth.

Figure 1. Transverse cross section of mini-plate R6003F-7Mo from RERTR-4. The two zones of porosity are readily apparent.

II. ATR and Experiment Details

The ATR in Idaho, USA has a complicated core as shown in Figure 2. The fuel assemblies follow a serpentine pattern that flow around the irradiation positions. The three B positions that were used for RERTR-4 and 5 are shown in Figure 2 and listed in Table 1. The experiment basket was positioned in the irradiation position so that the edges of the fuel plates always face the core. At the end of each cycle the basket was rotated through 180° so that the opposite edge faces the core. A strong neutron flux gradient exists across the width of the mini-plates and by rotating the fuel plates at the end of each cycle the burn-up becomes more uniform. The power profile is tabulated in Table 2.

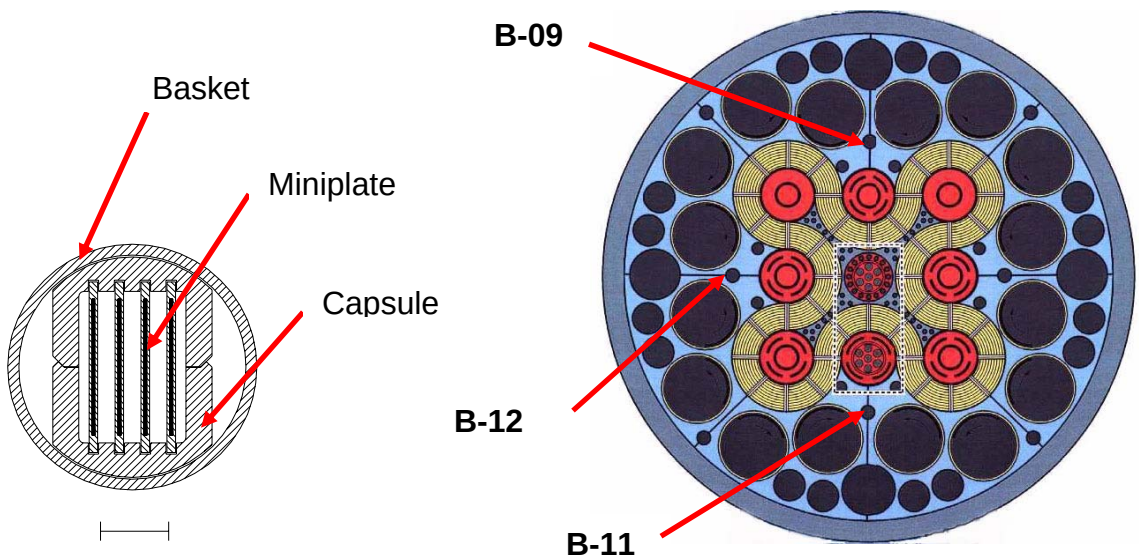

Figure 2. The ATR core and the design of the RERTR irradiation basket. The three B-holes used for RERTR-4 and 5 are B-09, B-11 and B-12.

Table 1. Experiment position and irradiation history for RERTR-4 and 5. S=South, N=North, W=West.

Cycle Number	RERTR-4	RERTR-5	EFPD
123B	S (B-11)	W (B-12)	42.2
123C	N (B-09)	W (B-12)	13.4
124A	W (B-12)	S (B-11)	57.5
124C	S (B-11)	Out of pile	40.1
125A-1/2	S (B-11)	Out of pile	54
125B	S (B-11)	Out of pile	49.9

Table 2. Local to average ratio of fission power profile from the inner edge to the outer edge of the mini-plate. X1 and X10 are the edges of the fuel meat.

Position	X1	X2	X3	X4	X5	X6	X7	X8	X9	X10
Dist from Edge (mm)	0	2.2	4.4	6.7	8.9	11.1	13.3	15.6	17.8	20
Fission Power Profile	1.46	1.27	1.15	1.06	0.97	0.92	0.85	0.81	0.78	0.73

III. Irradiation History

A number of mini-plates from RERTR-4 and 5 have been selected to illustrate the effect of irradiation parameters on the evolution of porosity in UMo fuel. One fuel plate from RERTR-3 has also been included as it adds further data of value in this study. They are listed in Table 3.

Table 3. Mini-plates selected for study.

Plate Number	Capsule Position	Composition. (a- atomized)	RERTR Test No.	Fission Density (10^{21} f/cm^3)	Burn-up (%U235)	Reaction Thickness (μm)
V6019G	B-4	U-10Mo[a]	5	3.2	49.0	15
V6001M	A-1	U-10Mo[a]	4	4.4	66.2	12
S6006C	C-6	U-6Mo[a]	4	6.1	82.5	22
Z03		U-10Mo[a]	3	1.8	29.8	5

The goal of this study is to identify differences in irradiation parameters where porosity is observed to have formed relative to areas where it has not. Therefore three positions across the fuel width have been selected for study. They are the center and two positions equidistant from the centerline. The exact position of the porosity varies from mini-plate to mini-plate based on plate specific variables such as meat width, thickness and local loading. On average however, the center of the porous region is ~3.5 mm from the meat edge. Therefore the peaking factors at those two locations are 1.2 or 0.8 (see Table 2) depending on which edge is closer to the core.

IV. Fission Density

The data shows that fission density is a prerequisite but not a key determinant in the formation of porosity. There is a minimum quantity of fission gas necessary to drive the formation and swelling of porosity, but there are examples where the presence of porosity is not linked to fission density. V6001M was irradiated in RERTR-4 and reached a fission density of 4.4×10^{21} f/cm^3, considerably higher than 3.2×10^{21} f/cm^3 of V6019G in RERTR-5. Yet despite such a substantial difference, porosity had evolved on both sides of mini-plate in V6019G while in V6001M it was not observed. Furthermore, the fission density achieved in Z03 (RERTR-3) was 1.8×10^{21} f/cm^3 and yet porosity has formed as shown in Figure 3.

It should be noted that each mini-plate was sectioned only once, in the transverse direction at the mid-plane. To be conclusive it would be necessary to make numerous sections or at least repeatedly grind and polish the metallographic samples to ensure that porosity had not been missed.

IV. Temperature

Temperature is an important consideration in the formation of porosity because it exerts a strong influence over the rate of reaction between fuel and matrix. It is also intimately

related to the fission rate. There are examples however that indicate that temperature is not the primary driving force for porosity formation. In Fig. 4 the fuel meat centerline temperature has been calculated for V6019G in the two regions of porosity, and also in the pore free region in the center. The center of the plate maintains a temperature greater than the right side of the plate for the entire irradiation, excluding the 14 days of the second cycle. Despite maintaining an elevated temperature the center region of the fuel plate failed to develop any porosity. The temperature differential between the location of porosity and the center remains less than 15°C for the period of the irradiation.

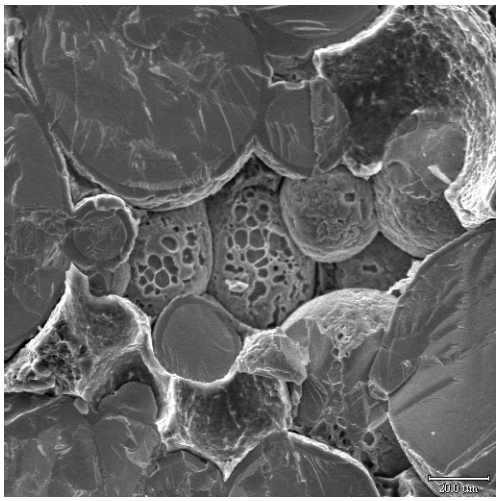

Figure 3. SEM image of Plate Z03 from RERTR-3. Porosity evolution is evident on the outer surface of the spherical U-Mo particles.

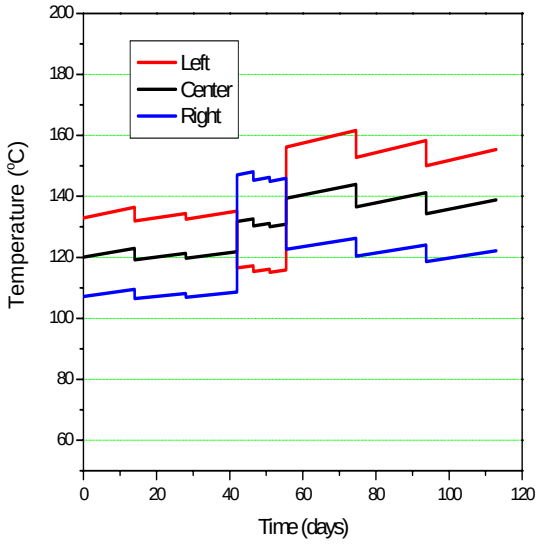

Figure 4. Graph of mini-plate V6019G centerline temperature calculated at the left, center and right positions across the width of the plate. .

At the interface between the fuel meat and the cladding the temperature drops due to the higher conductivity of the cladding and the lack of a heat source. Porosity is observed to form at this location in addition to the hotter centerline in some cases despite the temperature drop as shown by Lemoine et al. [1] in Figure 5.

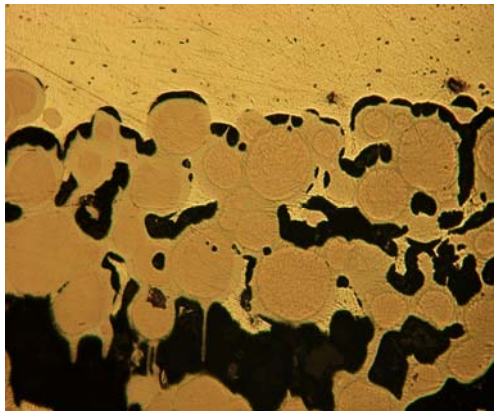

Figure 5. OM image of the interface between the fuel meat and cladding; FUTURE [1].

V. Fission Rate

Fission rate, like temperature is an important irradiation variable that is expected to exert an influence on the evolution of porosity as it is intimately linked to heat generation and consequently temperature. Like temperature however, it does not appear to be the sole reason why porosity forms in different locations. In Fig.6 it is clear that the fission rate in the fuel particles is largely constant throughout the irradiation and is greater in the center of the plate than the right side for almost the entire irradiation. Similarly, the fission rate of mini-plates in high flux positions is considerably higher across the entire plate than mini-plates of lower flux and still no porosity forms in the center of the plate. However, one should consider the fission rate (i.e., damage rate) in the interaction product itself rather than in the U-Mo fuel, because this is where the porosity forms.

Figure 7 shows the damage rate history for mini plate V6019G as calculated with the TRIM code [2].

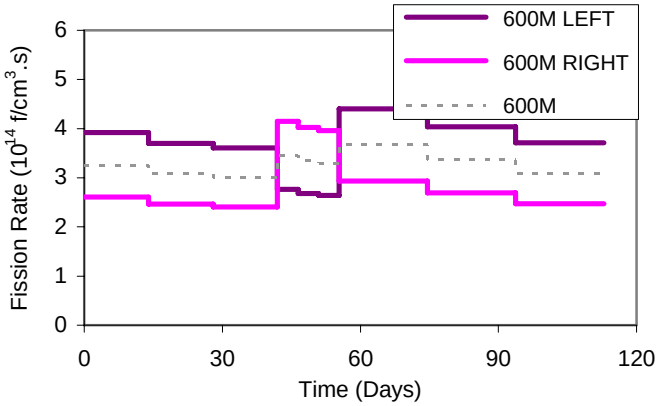

Figure 6. V6019G fission.

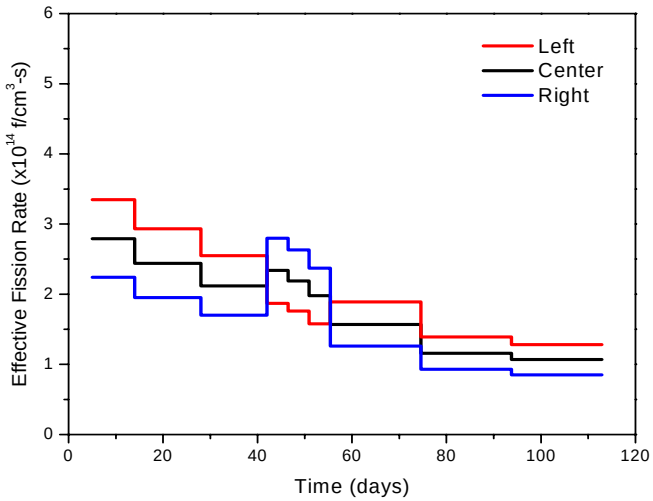

Figure 7. V6019G **effective** fission rate

VI. Interaction Layer Thickness

As mentioned previously there needs to be a minimum reaction thickness and quantity of fission gas present to nucleate and grow the pores. Based on Figure 3 it would appear that the threshold for this event is very low. The measured reaction thickness for mini-plate Z03 is ~5μm. However, thickness alone does not dictate whether porosity forms or not. For example, in V6019G, the reaction thickness is reasonably constant over the full width of the fuel meat. However, porosity was observed to form in two discrete locations approximately 3.5 mm from the edge of the meat but not in the center.

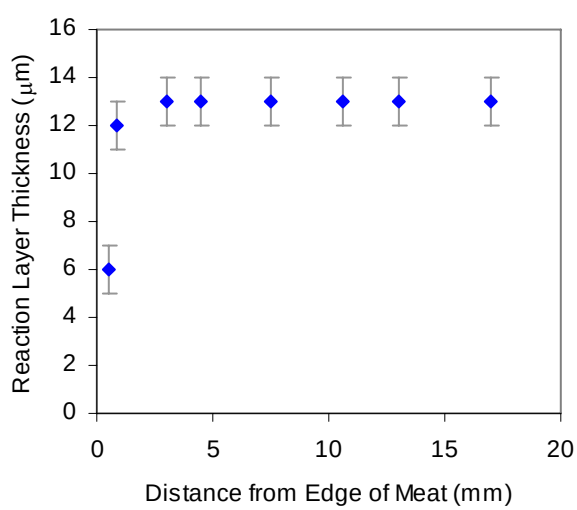

Figure 8. Interaction layer thickness in V6019G measured across the width of the mini-plate at the midplane.

VII. Composition

Given the quantitative information in the preceding sections it appears that neither the fission rate and fission density nor temperature can independently account for the location of porosity. Composition and its potential variation is the other readily identifiable variable that may be the key determinant. Fabrication variables such as uranium density have been considered as well but shown to be indeterminant.

Since the (U-Mo)Al_x interaction product is a fissionable material – a fuel – its composition, specifically its Al content expressed by 'x', could have a strong effect on its irradiation behavior; recall the vastly different fission gas bubble growth between U_3Si and U_3Si_2.

A measure of the composition of the interaction layer, i.e., its Al content is reflected in the ratio of the measured volume of interaction and the volume of matrix Al consumed in the formation of the former, V^I/V_c^{Al}.

This ratio is plotted in Figure 9 for several mini-plates of RERTR 4 and 5. The ratio, and therefore x in (U-Mo)Al_x, changes with temperature and loading presumably because when the available volume fraction of Al is diminished, more rapidly at higher temperature and/or higher fuel volume loading, U and Mo from the fuel particles continue to diffuse into the interaction layer, hence a decrease in x.

This is corroborated by EPMA analysis on irradiated fuels from the FUTURE [3] and IRIS [4] tests where, depending on the temperature and fission rates, x was found to decrease from ~6 to ~4.

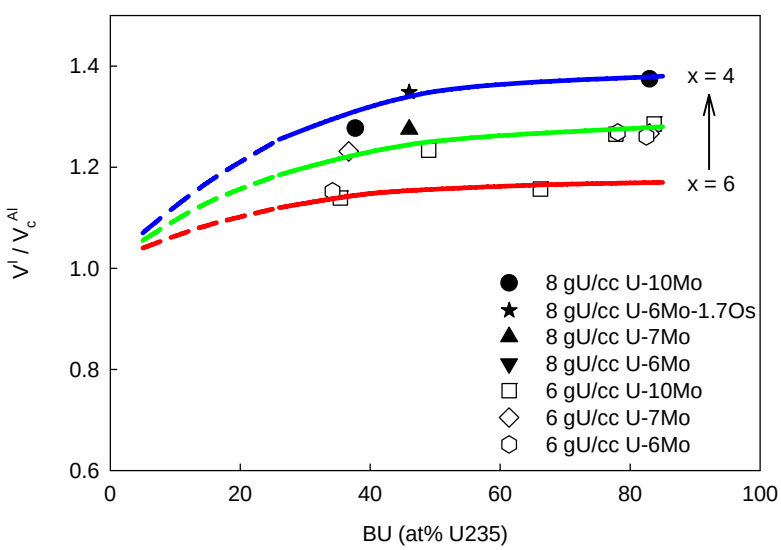

Figure 9. Volume changes RERTR-4 and 5 mini-plates during irradiation. V^I is the volume fraction of the interaction product and V_c^{Al} is the volume fraction of the consumed Al.

V Discussion

As the irradiation parameters and postirradiation measurement entities by themselves cannot explain the observed patterns in porosity, we have attempted to develop a simple multi-parameter function that captures the observations. We propose that there exists a critical damage rate, F_{cr}^{I}, above which value fission gas pores, that nucleate at the Al-interaction layer interface, can readily grow into the interaction product by rapid accumulation of fission gas from the surrounding interaction product implying a low viscosity – high fission gas mobility behavior.

The function consists of two parts; one part simply accounting for the dependence on the mobility and viscosity of the interaction product, the other part representing the accumulation of interaction product volume, the amount of fission gas, and the change in composition.

The function takes the following form:

$$F_{cr}^{I} = A^{l} \frac{1}{exp\left(-\frac{Q_1}{RT}\right)} \frac{1}{\sum \dot{F}^{I} \; exp\left(-\frac{Q_2}{RT}\right) \Delta t} \tag{1}$$

where A^l is a rate constant, that includes a factor accounting for different fuel loading,

$\dot{F}^{I}$ is the fission rate at the Al-interaction layer interface,

Q_1 is an activation energy characterizing the temperature dependence of the interaction product properties,

Q_2 is a compound "activation energy" characterizing the accumulation process,

T is the fuel temperature, and

Δt is the incremental increase in irradiation time.

Example calculations of F_{cr}^{I} with the best-fitted parameters (A^l = 0.052, Q_1 = 7500 cal, Q_2 = 100 cal, $\dot{F}^{I}$ is in 10^{14} f/cm^3/s, R = 1.987 cal/mol-K, T is in K, and Δt is in day) for several mini-plates from RERTR-4 and 5, which span the operating range of temperature and fission rate, are compared with the fission rate histories of these mini-plates in Figure 10.

The locations of observed porosity or absence there of coincide with whether the critical fission rate is reached.

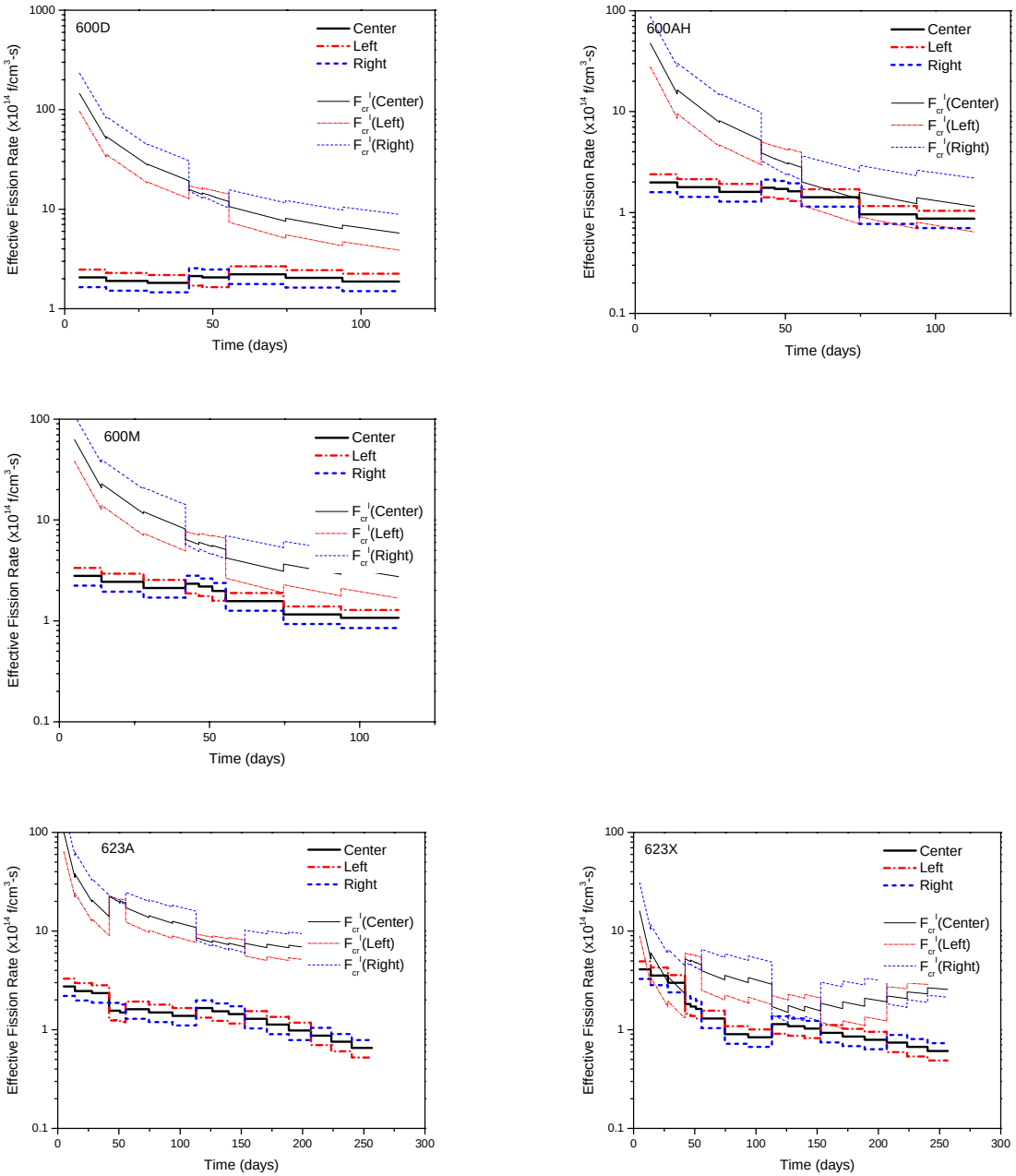

Figure 10. Comparison of calculated fission rate histories and critical fission rate function

VI. Conclusions

Given the quantitative information in the preceding sections it appears that neither the fission rate and fission density nor temperature can independently account for the location of porosity. However, the porosity patterns suggest that some critical combination of operating parameters is a prerequisite for the pores to nucleate and grow. This critical combination can be expressed by a rather simplified function of time,

temperature and fission rate as shown in Eq. (1). The parameters in Eq. (1) are fitting constants and are, as well as the functional form of Eq. (1), likely to change when more details of the porosity phenomenon become available. Two particular issues are: the composition and properties of the interaction product, and the fuel temperature during the course of the irradiation. Fuel temperature calculations rely on thermal conductivity data from ex-reactor measurement and may be subject to substantial uncertainty. The evaluation presented here may, with appropriate caveats, delineate an operational range for acceptable behavior. It is clear that for high temperature – high fission rate fuel operational conditions, a solution to the porosity problem can only be obtained by metallurgical changes to the fuel design.

As fission rate and temperature are given parameters for a particular reactor application the only possible independent change lies in eliminating fuel-Al interaction altogether through the use of a diffusion barrier between matrix and fuel particles, or by stabilizing the (U-Mo)/Al interaction product through alloy additions to either or both fuel and matrix. These subjects are dealt with in detail in several papers at this conference.

References

[1] P. Lemoine et al., RRFM 2004.
[2] J. F. Ziegler et al., TRIM, www.SRIM.org, 2005.
[3] A. Leenaers et al., J. Nucl. Mater., 335 (2004) 39.
[4] F. Huet et al., RRFM 2005.