MODELLING OF U-MO/AL INTERACTION LAYER GROWTH IN FULL-SIZE DISPERSION FUEL PLATES

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ABSTRACT

In order to simulate the U-Mo/Al interaction layer (IL) growth behavior in full-size dispersion fuel plates, the existing IL growth correlation was modified with a temperature-dependent multiplication factor which transforms around a threshold fission rate. Test data from FUTURE, E-FUTURE, SELENIUM, and SELENIUM-1a were utilized to determine and validate the updated IL growth correlation. Irradiation behavior of the plates was simulated with the DART-2D computational code. The general agreement between calculated and measured fuel meat swelling and constituent volume fractions as a function of fission density demonstrates the plausibility of the updated IL growth correlation. The simulation results also confirm that IL growth rate differs in different fission rate/temperature regimes.

1. Introduction

As one of the intrinsic characteristics impacting U-Mo/Al dispersion fuel performance, the fission-enhanced interdiffusion between U-Mo fuel particles and the Al matrix has been investigated previously, and several empirical correlations were developed based on either out-of-pile or in-pile test data [1]-[4]. However, the existing correlations of interaction layer (IL) growth during irradiation were fitted for the U-Mo/Al dispersion fuels in geometry of either a miniature plate or a fuel rod [1], using life- and space-averaged power and fuel meat temperature. Its applicability to the full-size fuel plates when using time-varied power history was not confirmed. Moreover, irradiation data of fuel plates that have an improved design, such as Si addition in the Al matrix or coated fuel particles, was either scarce or not available when the correlations were developed. Therefore, the existing correlations are not sufficient to capture the IL growth behavior in current fuel design. This study used DART-2D, a 2-dimensional dispersion fuel performance code, to develop a proper description of the IL growth behavior based on the most recent irradiation data of full-size dispersion fuels. The formulation of the updated IL correlation and its validation results are presented in this paper.

2. The existing IL growth correlation and its limitations

This section describes the existing empirical correlation of IL growth during irradiation and its limitations.

2.1 Average power and temperature vs. time-varied power and temperature

Eq. (1) is the latest IL growth correlation during irradiation published [1]:

$$Y_0^2 = 2.6 \times 10^{-16} (\dot{f})^{0.5} \cdot t \cdot \exp(-\frac{7700}{R \cdot T}) \quad (1)$$

where Y_0 is IL thickness in cm, $\dot{f}$ is fission rate in f/cm^3/s, t is irradiation time in s, R = 1.987 cal/K/mol, and T is the temperature in K. The constants in Eq. (1) were fitted for the U-Mo/Al dispersion fuels with a pure Al matrix based on RERTR (-4, -5, -6, -7, and -9) and KOMO-4 test data. The fuel temperatures used to represent the test samples are arithmetic averages of the BOL (beginning-of-life) and EOL (end-of-life) fuel temperatures for the RERTR plates and life-average values for the KOMO-4 rods [1]. The power/fission rate used was averaged over the fuel life and dimensions as well.

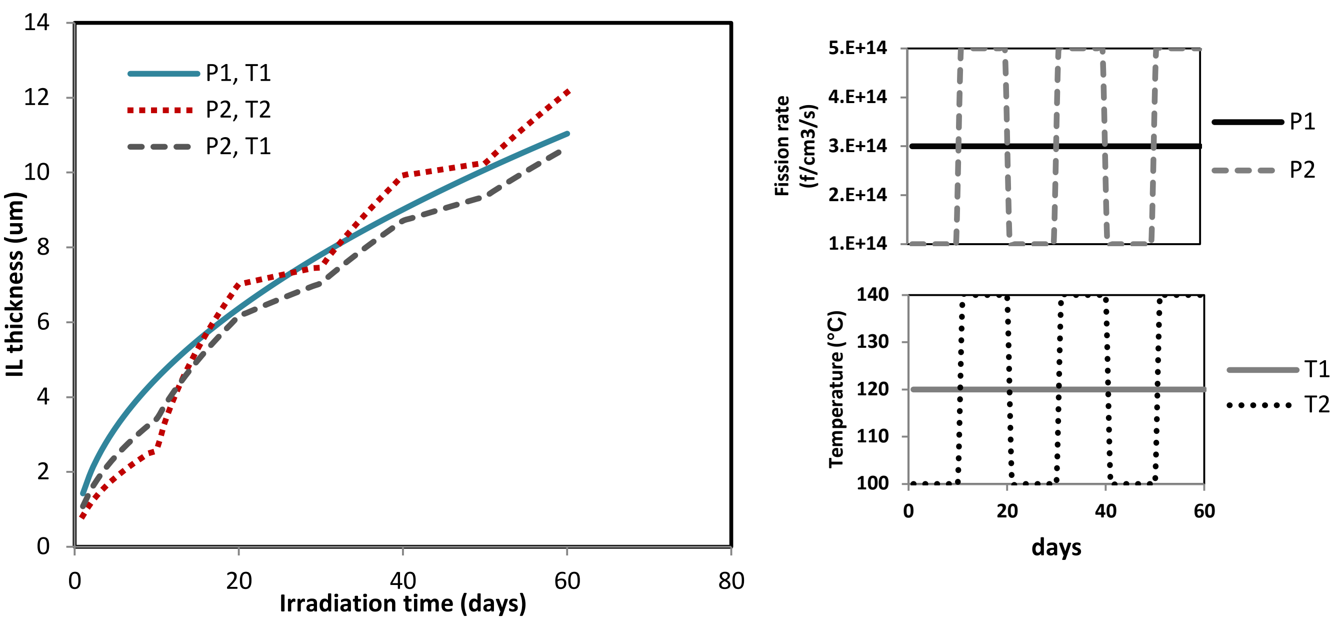

Fig. 1 Comparison of IL growth behavior through different operating histories.

IL thickness is not a linear function of life-averaged fission rate as expressed in Eq. (1). In other words, the same average power and temperature may not generate an identical IL thickness, if the operating history varies. An example of IL growth through different operating histories is shown in Fig. (1). All three cases in Fig. (1) have the same average power and temperature, and they achieved the same burnup at the end of irradiation. However, the final IL thicknesses are all different (the largest difference is ~ 13%). When the thickness is converted to IL volume, an even larger discrepancy is expected. To minimize this kind of uncertainty, it is necessary to calculate through realistic operating history instead of using averaged power and temperature.

2.2 The IL reduction effect of Si addition

As a possible way to reduce IL formation, Si was added into the Al matrix [5][6]. To describe the IL reduction effect of Si addition, Kim et al. [1] proposed to multiply an additional factor f_{Si} to Eq. (1). f_{Si} was fitted based on the best available data at that time, RERTR-6 and KOMO-4 test data, and expressed as a function of temperature and Si concentration:

$$f_{Si} = (1.201 - 0.00062T)\exp[-(10.333 - 0.021T)W_{si}] + (0.00062T - 0.201)\exp[-(0.00081T - 0.302)W_{Si}] \quad (2)$$

There was an effort in the past to simulate the E-FUTURE test [7], which is an irradiation test of Si-added full-size dispersion plates at the BR2 reactor in Belgium. Some limitations of the IL growth correlation expressed in Eqs. (1) and (2) were pointed out, and the accuracy of Eq. (2) was one of them. Since the method of introducing Si into the Al matrix is different in the E-FUTURE plates and in the RERTR and KOMO fuels, Eq. (2) may not be applicable and needs further investigation.

2.3 The IL growth behavior for coated particles

A design of coating fuel particles with a thin layer of diffusion barrier was put forward to further reduced IL formation and to stabilize the dispersion fuel behavior at high burnup [8]. Two full-size plates fabricated with fuel particles coated with a layer of Si or ZrN were irradiated in the SELENIUM test at the same operating conditions as the E-FUTURE test for direct comparison. Fig. 2 displays the irradiated microstructures of SELENIUM U7MD1221 (Si coating), SELENIUM U7MD1231 (ZrN coating), and E-FUTURE U7MC6111 (6% Si in matrix) plates taken at similar fission densities, demonstrating that the SELENIUM fuels have improved microstructural behavior over the E-FUTURE plates [9][10]. Nevertheless, IL formation still occurred at fission densities above 5.1×10^{21} f/cm^3 (Figs. 2 (a) and (b)) in the SELENIUM plates, and the IL volume fraction as a function of fission density is plotted in Fig. 3.

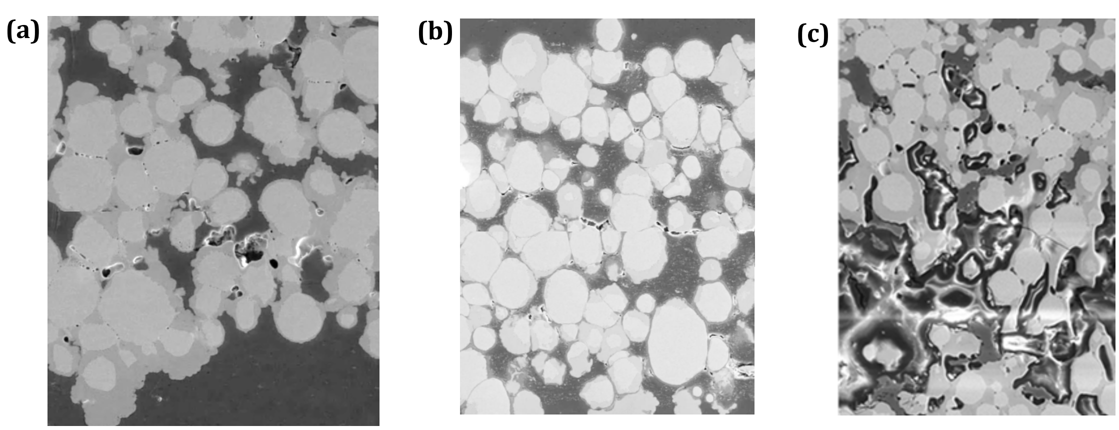

Fig. 2 Scanning electron microscopy (SEM) images of (a) SELENIUM-U7MD1221 (Si coating) plate taken at 5.1×10^{21} f/cm^3, (b) SELENIUM- U7MD1231 (ZrN coating) plate taken at 5.2×10^{21} f/cm^3, and (c) E-FUTURE-U7MC6111 (6% Si in matrix) plate taken at 5.2×10^{21} f/cm^3 [11].

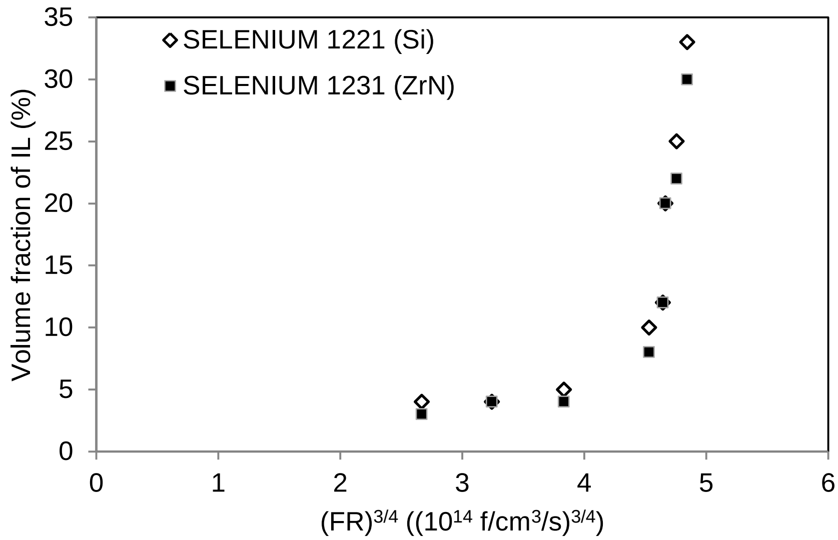

Fig. 3 Measured IL volume fractions in the SELENIUM plates [12].

The interaction of Al with U-Mo in the coated particles occurs through damaged areas in the coating [12]. A hypothesis was proposed based on the data in Fig. 3 and its similarity to ion-mixing results in Nb-Si system [13][14] – part of the fuel plate has a temperature and fission rate independent intermixing behavior changing at a threshold fission rate $\dot{f}_{threshold}$ to a thermally activated behavior [12]. This temperature dependency is typically referred to as the Q-curve behavior in ion mixing studies. This transition behavior was not described in the existing IL growth correlations. However, it was attempted to use Eq. (1) to fit the IL volume fraction in the thermally activated regions where $\dot{f} > \dot{f}_{threshold}$. By reducing the rate coefficient from 2.6×10^{-16} to 0.1×10^{-16}, Eq. (1) was able to describe the IL growth behavior in the regions where $\dot{f} > \dot{f}_{threshold}$. The reduction of the rate coefficient was explained with the reduced U-Mo/Al contact area at coated particle surfaces, i.e. limited to breaches in the coating.

3. Modifications of the IL growth correlation

Irradiation data from 6 full-size U-7Mo/Al dispersion plates irradiated at the BR2 reactor were used for updating the IL growth correlation. These 6 plates were selected, because they represent all 3 types of dispersion fuel design available: pure Al matrix, Si addition in the Al matrix, and coated particle. The fabrication characteristics and irradiation conditions of these 6 plates are listed in Table 1.

Table 1. Fabrication and irradiation characteristics of the 6 selected plates irradiated at BR2.

Test	FUTURE [9]	E-FUTURE [9]		SELENIUM [9]		SELENIUM - 1a [16]
Irradiation period	2002-2003	2010-2011		2012		2015-2016
Plate ID	U7MTBR07	U7MC4202	U7MC6301	U7MD1221	U7MD1231	U7MD1222
U-Mo powder type	Atomized	Atomized	Atomized	Atomized +	Atomized +	Atomized +

				600nm Si	1μm ZrN	600nm Si
Mo content (wt%)	7.3	7.5	7.3	7.2	7.2	7.2
Enrichment (%^{235}U)	19.8	19.8	19.8	19.8	19.8	19.8
Fuel loading (gU/cm^3)	8.5	8.1	8.0	8.0	8.0	8.0
Matrix type	Al	Al+4.1wt%Si	Al+6.0wt% Si	Al	Al	Al
Cladding material	AG3NE	AG3NE	AlFeNi	AG3NE	AG3NE	AG3NE
Max. heat flux BOL (W/cm^2)	351	453	472	421	466	278
Total EFPD (days)	40	77	77	70	70	98
Plate average burnup (%^{235}U)	25	48.1	47.5	47.9	47.5	53.1
Plate average FD (f/cm^3 UMo)	1.8×10^{21}	3.6×10^{21}	3.6×10^{21}	3.5×10^{21}	3.5×10^{21}	4.0×10^{21}
Plate max burnup (%^{235}U)	31.9	71.3	71.4	69.2	69.6	73.5
Plate max FD (f/cm^3 UMo)	2.4×10^{21}	5.5×10^{21}	5.5×10^{21}	5.3×10^{21}	5.3×10^{21}	5.5×10^{21}
Life average fission rate (f/cm^3 UMo/s)	5.2×10^{14}	5.4×10^{14}	5.4×10^{14}	5.8×10^{14}	5.4×10^{14}	4.7×10^{14}

The transition of IL growth rate at a threshold fission rate was first noticed in the coated particle system as shown in Fig. 3. However, according to the IL formation analyses in Ref. [12], the behavior of the rapid growth of IL within a small range of fission rate should also occur in plates fabricated with uncoated particles. A multiplication factor $F_{reduction}$ with a threshold fission rate was therefore applied to all types of fuel plates and not limited to the coated particle systems.

$F_{reduction}$ was assumed to be a function of fuel meat temperature, similar to the f_{Si} in Eq. (2). A sigmoid function was applied to achieve a smooth transition between the two fission rate regimes where IL growth rate is largely different. The resultant function is

$$F_{reduction} = C_1 \cdot T + \frac{(C_2 - C_1) \cdot T}{1 + \exp(2 \times 10^{-14} \cdot (\dot{f}_{threshold} - \dot{f}))} \quad (3)$$

where C_1 and C_2 are constant coefficients, and they may vary with the change of fuel design. T is fuel meat temperature in K; $\dot{f}_{threshold}$ and $\dot{f}$ are threshold fission rate and fission rate respectively in f/cm^3/s.

The threshold fission rate $\dot{f}_{threshold}$ was determined based on the measured data: 6×10^{14} f/cm^3/s for the FUTURE plates and 8×10^{14} f/cm^3/s for the E-FUTURE and SELENIUM plates. C_1 and C_2 were fitted to measured fuel meat swelling and constituent volume fractions using DART-2D. $\dot{f}_{threshold}$, C_1, and C_2 for each type of fuel design are listed in Table 2. The IL growth correlation implemented in DART-2D is the product of Eqs. (1) and (3), which is:

$$Y^2 = 2.6\times10^{-16}(\dot{f})^{0.5}\cdot t\cdot\exp(-\frac{7700}{R\cdot T})\cdot(C_1\cdot T + \frac{(C_2 - C_1)\cdot T}{1+\exp(2\times10^{-14}\cdot(\dot{f}_{threshold} - \dot{f}))}) \qquad (4)$$

Table 2. Parameters in $F_{reduction}$ fitted for different fuel design.

Fuel design	Test data	$\dot{f}_{threshold}$ (f/cm^3/s)	C_1	C_2
Pure Al matrix	FUTURE	6×10^{14}	1×10^{-4}	7×10^{-4}
Al matrix with Si addition	E-FUTURE	8×10^{14}	1×10^{-4}	7×10^{-4}
Coated particles	SELENIUM, SELENIUM-1a	8×10^{14}	1.5×10^{-5}	1.05×10^{-4}

The fitting results show that, compared to the improved fuel design (Si addition in the Al matrix or Si-coated particles), the FUTURE plate has a lower $\dot{f}_{threshold}$, which can be explained with lower mixing heat. The reason for the higher $\dot{f}_{threshold}$ for ZrN-coated particles is however different. It cannot be directly explained with a different mixing heat, because there is no Si in the system to change the U-Mo-Al reaction rate. Instead, it could be related to a wetting phenomenon in which no wetting occurs even in the case of a breached coating as long as the diffusion rate is insufficiently high. C_1 and C_2 fitted for the plates fabricated with uncoated particles and the Al matrix with Si addition are ~ 7 times larger than those fitted for the plates with coated particles and an Al matrix. The difference is due to the significant reduction of diffusion paths between U-Mo and Al in the latter. For this reason, it is speculated that for a fuel plate composed with coated particles and an Al-Si matrix, very possibly, $\dot{f}_{threshold}$ will remain to be 8×10^{14} f/cm^3/s, but C_1 and C_2 may decrease further from those for the coated particle system with an Al matrix.

It is worth to mention that temperature is an important parameter in irradiation-induced intermixing as shown in the chemical ion-mixing experiments [13]. The fuel meat temperature is also related to thermal conductivity evolution, which creates a non-linearity in the dependence of temperature and fission rate and introduces a feedback dependency on the IL since it is also expected to influence thermal conductivity. It also creates a reactor dependency, since the cooling will affect the oxide layer growth, which in turn also affects the thermal conductivity. Therefore, it is more physical to use temperature threshold for IL growth behavior modelling. However, fission rate is employed to describe the transition behavior of IL growth rate in this study, because of multiple reasons: (1) fuel meat temperature is currently unmeasurable; (2) fuel meat temperature and fission rate are highly correlated; and (3) all plates investigated in this study were irradiated in the same reactor and no reactor dependency was involved. The temperature threshold will be investigated in the future using irradiation data from other reactors (different thermal conditions) and possible out-of-pile irradiation data.

4. Simulation results of full-size dispersion plates using the updated IL growth correlation

To assess the accuracy of an IL growth correlation, it is key to have a reliable simulation tool to simulate the in-pile irradiation behavior of dispersion fuel. The DART-2D computational code [15] was developed for dispersion fuel performance modelling. It uses both empirical and mechanistic physics models to generate results which can be directly compared with PIE data. The code takes the neutronics-analysis-generated power history as input and calculates fuel meat swelling, constituent volume fractions, temperature, and thermal conductivity at each time step. More detailed description on the capabilities of the code, physics models implemented, and calculation procedures can be found in Ref. [15].

This section presents the DART-2D simulation results of the plates listed in Table 1 using the IL growth correlation developed in Section 3. The calculations utilized input information provided by SCK•CEN, including fabrication characteristics, nominal plate dimensions, coolant conditions, and MCNP-calculated power profiles. Other computation settings are: 1) time step length is 1 day; 2) the number of grids in fuel meat domain is 12 (plate width direction) × 18 (plate length direction), the same set-up as what was defined in the neutronics calculations; 3) fuel particle size distribution is represented with six size groups. In this way, the fact that the particles in the SELENIUM/SELENIUM-1a plates have a larger average size than those in the E-FUTURE plates is taken into account.

4.1 Arrhenius growth of IL

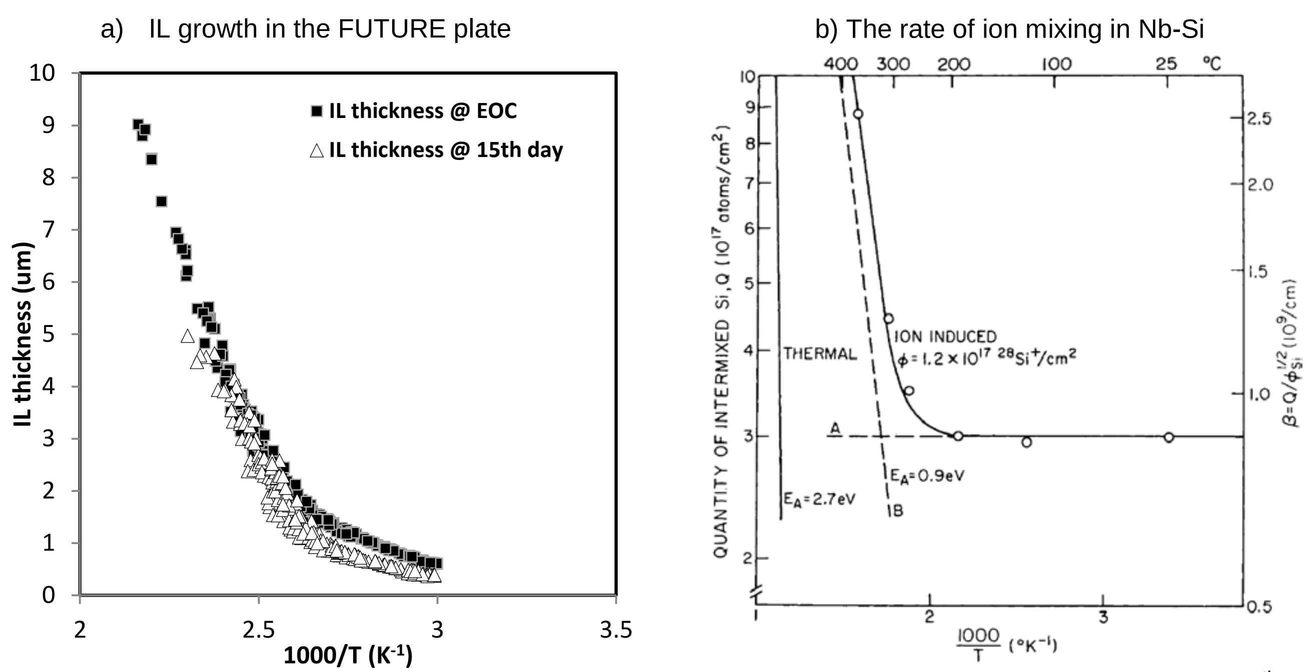

Fig. 4 Temperature dependences of (a) IL growth in the FUTURE plate at EOL and the 15th irradiation day and (b) the rate of ion mixing in Nb-Si system (adapted from Ref. [13]).

Fig. 4 depicts Arrhenius plots of typical IL growth in a full-size fuel plate (Fig. 4 (a)) and the rate of ion-beam mixing in Nb-Si system bombarded with Ar ions (Fig. 4 (b)). The resemblance between Figs. 4 (a) and (b) supports the hypothesis that a threshold fission rate/temperature for IL growth during irradiation does exist in full-size dispersion fuel plates proposed [12]. Note that, in Fig. 4 (a), the Arrhenius plot of IL growth evolves with burnup/irradiation time, due to the changes of system composition and configuration during irradiation.

4.2 Comparison of calculation and measurement results of fuel meat swelling and IL volume fraction

DART-2D calculation results were compared with experimental data for fuel meat swelling and constituent volume fractions for each plate. All measurement data were obtained at SCK•CEN at the end of irradiations. For comparison, the calculation results at EOL were plotted against fission densities at the corresponding locations in the same way as the measurement data was plotted.

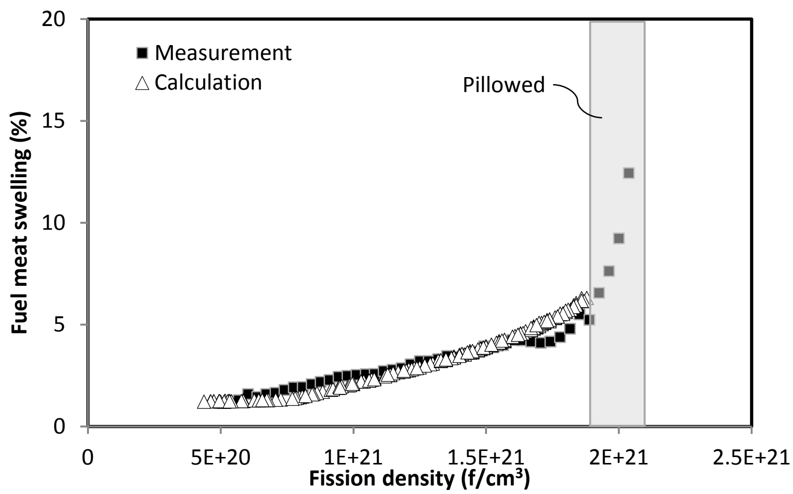

Fig. 5 A comparison of calculated and measured fuel meat swelling in the FUTURE plate.

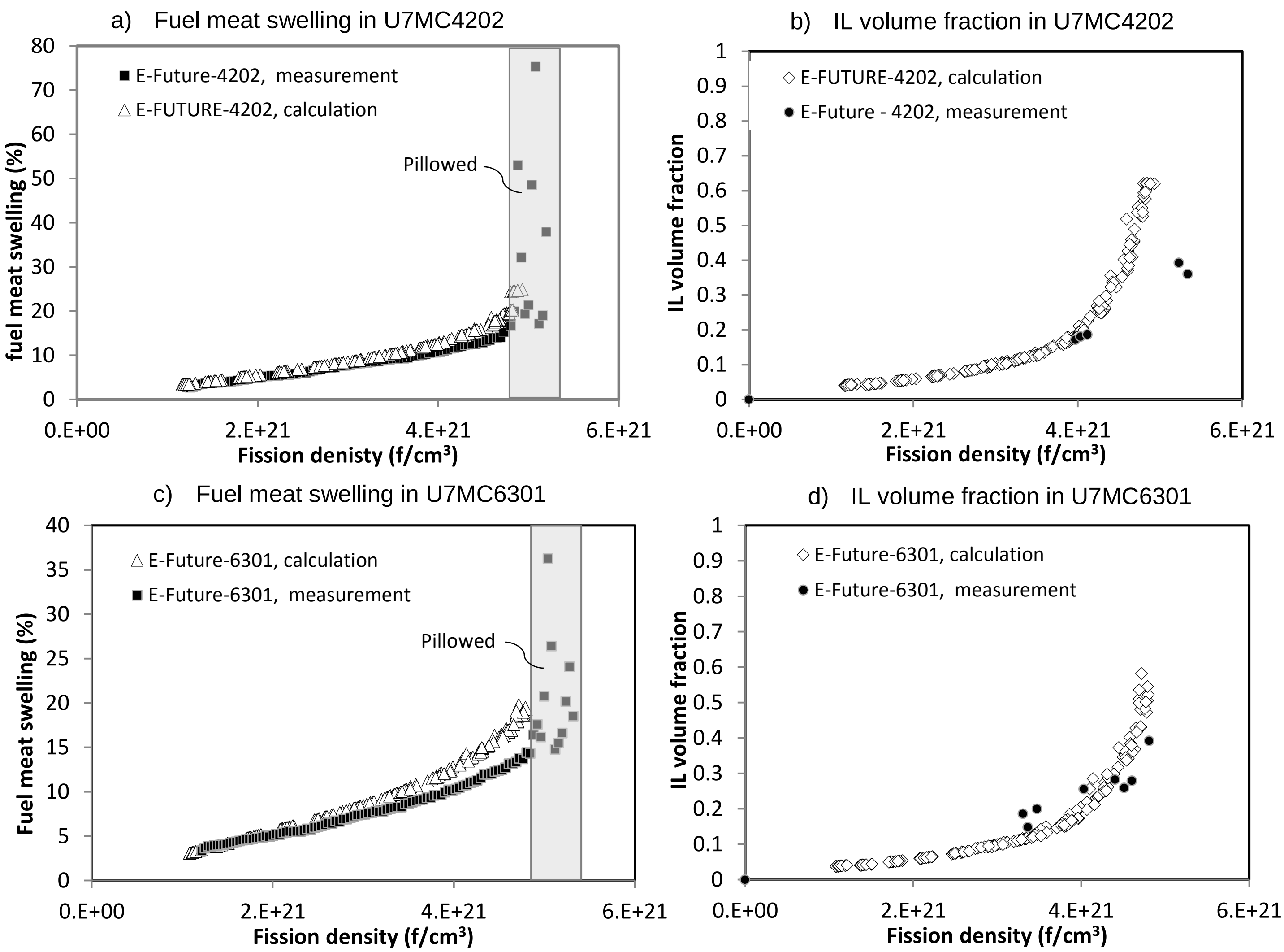

Fig. 6 Comparisons of calculation and measurement results of the E-FUTURE U7MC4202 ((a) and (b)) and U7MC6301 ((c) and (d)) plates respectively. Fuel meat swelling comparisons are in (a) and (c); IL volume fraction comparisons are in (b) and (d).

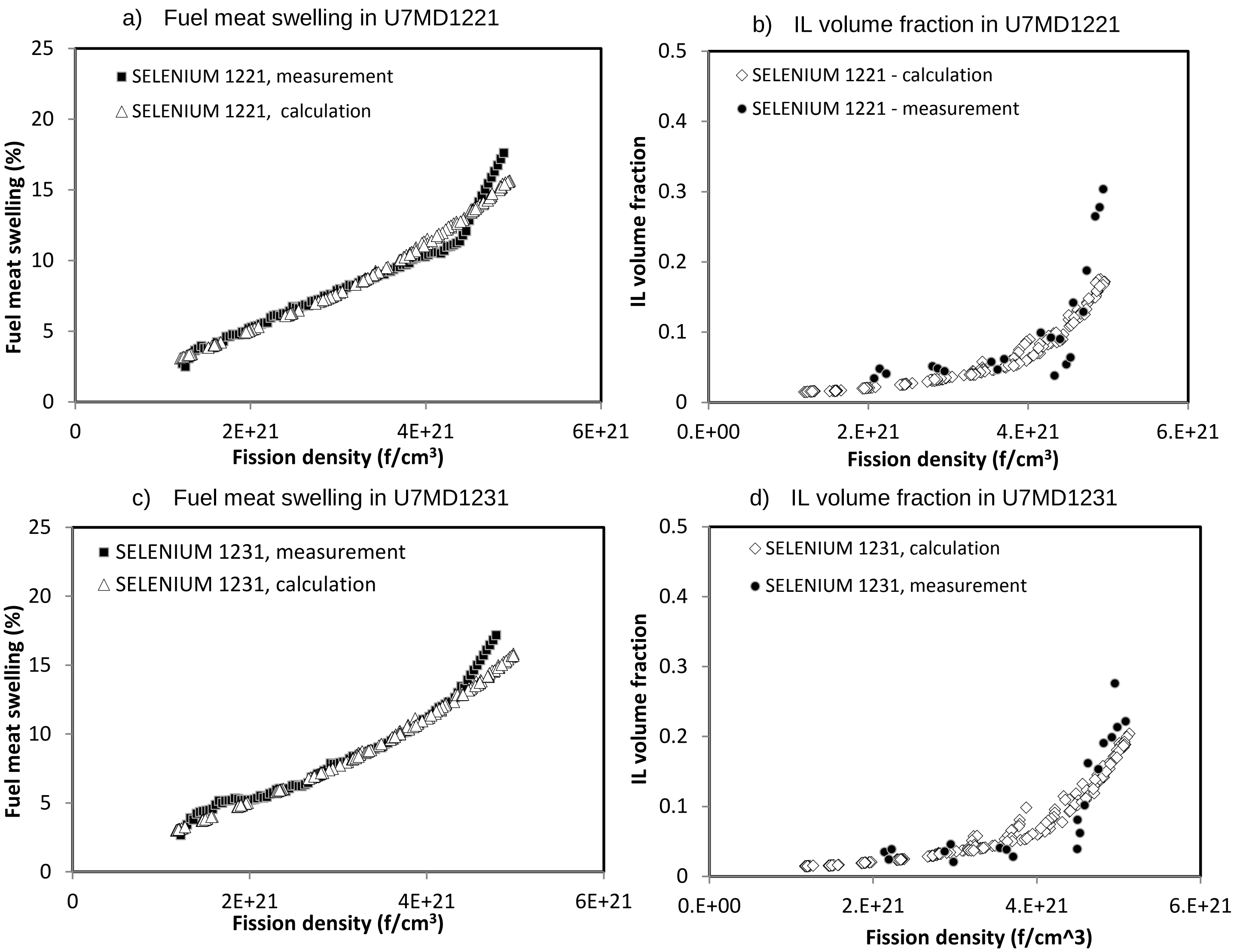

Fig. 7 Comparisons of calculation and measurement results of the SELENIUM U7MD1221 ((a) and (b)) and U7MD1231 ((c) and (d)) plates respectively. Fuel meat swelling comparisons are in (a) and (c); IL volume fraction comparisons are in (b) and (d).

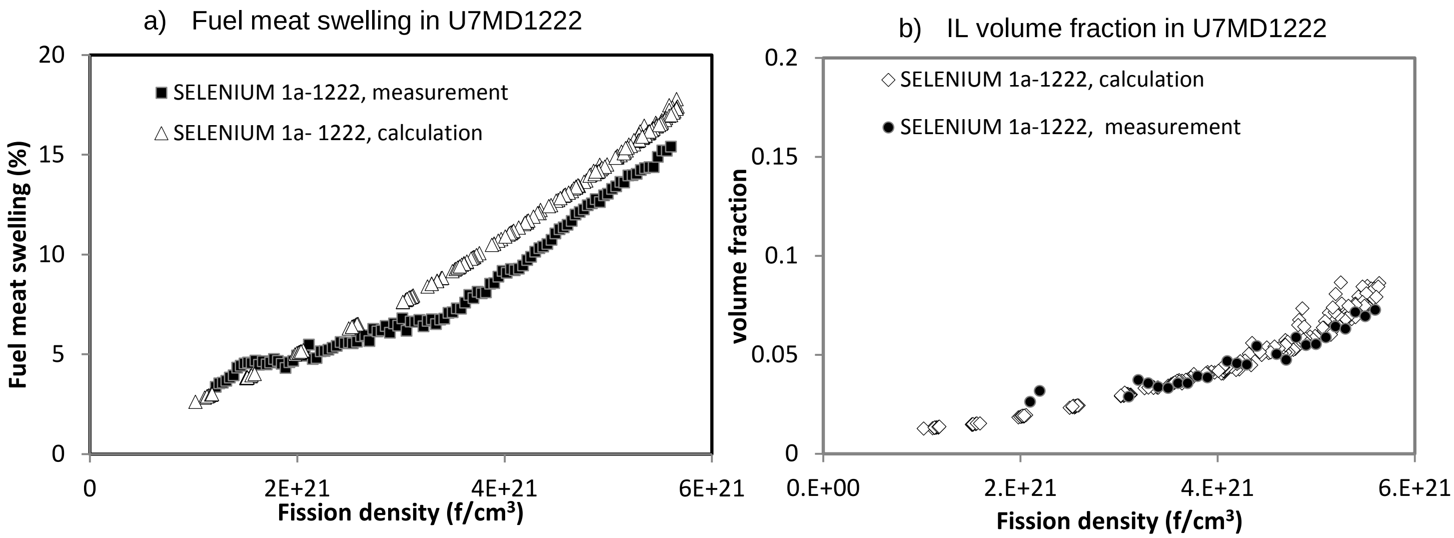

Fig. 8 Comparisons of calculated and measured (a) fuel meat swelling and (b) IL volume fraction in the SELENIUM U7MD1222 plate.

Figs. 5 – 8 display the calculated fuel meat swelling and IL volume fraction as a function of fission density in the FUTURE, E-FUTURE, SELENIUM, and SELENIUM-1a test plates respectively. The calculation results agree with measurement data generally. The pillowed regions in the FUTURE and E-FUTURE plates, indicated in Figs. 5 and 6, were not simulated because of the lack of reliable pillowing model and the large uncertainties in the measurements. For the SELENIUM plates (Fig. 7), the computational results generally agree with the measured fuel meat swelling over a fission density range up to ~ 4.5×10^{21} f/cm^3-UMo, then the two curves deviate at higher burnup in both plates. The discrepancy stems from the inaccurate U-Mo swelling correlation, which does not capture well the accelerated U-Mo swelling behavior after the fuel particles are fully recrystallized. With the implementation of a theory-based swelling model [15], the discrepancy will be improved. The comparison results in Figs. 5 - 8 demonstrate that the IL growth correlation developed in Section 3 reasonably represents the IL growth behavior in full-size dispersion plates.

5. Conclusions

An empirical IL growth correlation for U-7Mo/Al dispersion fuel has been proposed based on the PIE data of 6 full-size dispersion fuel plates irradiated at the BR2 reactor in 4 separate tests. The correlation is expressed as a modified Arrhenius equation multiplied with a temperature-dependent reduction factor. A Sigmoid function was employed to describe the transition of IL growth rate around a threshold fission rate. The IL growth correlation was applied to 3 different types of fuel design: pure Al matrix, Si addition in the Al matrix, and coated particle. Fuel meat swelling and IL volume fractions were calculated with the DART-2D dispersion fuel performance code using realistic power history and operating conditions. The calculated results are in reasonably good accordance with the measurement data for all 6 fuel plates. Therefore, it is reasonable to use the updated IL growth correlation for U-7Mo/Al dispersion fuel behavior simulation. Moreover, its plausibility was confirmed with the close similarity between the Arrhenius plot of IL thickness in a fuel plate and the temperature dependence of ion-mixing rate in Nb-Si system. For future work, the IL growth correlation will be tested with the irradiation data from different reactors. In addition, neither Si concentration nor coating material is treated as a parameter in the current IL correlation. Their effects on IL growth will be explored when more experimental data become available.

Acknowledgements

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