INTERACTION LAYER GROWTH CORRELATIONS FOR
(U-Mo)/Al AND Si-ADDED (U-Mo)/Al DISPERSION FUELS

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ABSTRACT

The interaction layer (IL) growth correlation of (U-Mo)/Al dispersion fuel for in-reactor tests is considerably different from that for out-of-pile tests because it contains factors that enhance diffusion kinetics by fission damage in the IL. This enhancement during irradiation was formulated by a combination of the fission rate and fission-fragment damage factor in the IL. Using a computer code (TRIM), fission damage factors were obtained as a function of the thickness and Al-composition of the IL. As a result of data fitting, the model correlation was set as a function of four major factors: fission rate, fission-damage factors, temperature and time. One important change from the old model used in the PLATE code is that the new one has the square-root dependence on the fission rate, whereas the old one has a linear dependence. The preliminary PIE results from RERTR-6 have shown a significant reduction in the IL thickness for the Si-added plates as well as prevention of pore formation. The reduction in IL growth followed an exponential decay function of the Si-content.

1. Introduction

The interaction layer (IL) thickness of U-Mo/Al dispersion fuel is usually described by an Arrhenius-type equation for out-of-reactor data. This equation is typically obtained at high temperatures, around 550°C. A correlation was developed for IL thickness from annealing tests of dispersion fuel.

The correlation for the out-of-pile data, when extrapolated to the reactor operation temperatures ~150°C, predicts IL thicknesses that are many orders of magnitude smaller than those measured in the irradiation tests. This is due to fission-fragment-enhanced diffusion (FED) caused by fission-fragment damage in the IL. Therefore, a new correlation applicable for the irradiation
tests was needed. We modeled the FED in the new correlation as a combination of the fission rate in the fuel and the fission-fragment damage factor in the IL. Specifically, the FED was implemented into the correlation as the square-root term of the fission rate multiplied by the fission-fragment damage factor. This is a change from the old model used in the PLATE code which has a linear dependence on the fission rate [1].

An iteration method was used to fit the model to the measured data from various in-reactor tests, primarily from RERTR tests and KOMO tests performed at KAERI.

A prominent observation from the recently available PIE data from the RERTR-6 test is that the IL thicknesses of the Si-modified plates are significantly smaller than the pure Al-matrix plates. In this paper, we present evaluation results for the effect of the Si addition in terms of the IL thickness and include some results of correlation work.

2. Post irradiation observations

The cross section of U-Mo/Al dispersion fuels from high-temperature annealing tests showed that three distinct layers with different compositions formed between U-Mo and Al [2]. Some layers appeared to be two-phase mixtures.

Irradiation test results, however, showed only a single layer with apparently a uniform composition throughout the IL, although the composition is different for each test [3], ranging from (U₀.₈Mo₀.₂)Al₃ to (U₀.₈Mo₀.₂)Al₇. The Al-content of an IL depends on the irradiation temperature, fuel loading, and fission density. At low temperatures the Al/(U+Mo) mole ratio tends to be higher. The difference in morphology of the diffusion zone in irradiated fuel compared to that of out-of-pile annealed fuel has been attributed to the phenomenon of fission-fragment induced amorphization [4]. Although amorphization of crystalline material during irradiation has been studied extensively in the literature [5], there has been no direct evidence on the amorphization of the IL of U-Mo/Al dispersion fuel until recently [6].

3. Interaction layer growth correlation for out-of-pile tests

The IL growth correlation, for out-of-reactor data obtained at 500 - 600°C, can be described by the parabolic law as follows:

\[ Y^2 = kt \]  \hspace{1cm} (1)

where \( Y \) is IL thickness, \( k \) is the interaction rate constant given by the Arrhenius equation

\[ k = k_0 \exp \left(-\frac{Q}{RT}\right) \]  \hspace{1cm} (2)

where \( k_0 \) is a constant, \( Q \) is the effective activation energy for IL growth, \( R \) is the gas constant, and \( T \) is the temperature.

For spherical coordinates Eq.(1) transforms to:

\[ \frac{(\beta-1)(1-\alpha)^{2/3} + [1+(\beta-1)\alpha]^{2/3} - \beta}{2(1-\beta)} \cdot r_0^2 = kt \]  \hspace{1cm} (3)
where \( r_0 \) is the initial fuel radius, \( \beta \) is the ratio of the volume of product formed to the volume of fuel consumed, and \( \alpha \) is the fraction of the original fuel volume consumed. The constant \( \alpha \) varies in the range, \( 0 \leq \alpha \leq 1 \). The constant \( \beta \) varies depending on the product density, which is closely related to the Al/(U+Mo) ratio of the reaction product. The method to estimate \( \beta \) is available elsewhere [7].

\( k_0 \) and \( Q \) were obtained by using data from the dispersion fuel annealing tests [2,8], and the results are \( k_0 = 3.94 \times 10^{16} \, \mu m^2/s \) and \( Q = 300 \, kJ/mol \).

### 4. Interaction layer growth correlation for irradiation tests

The analytical method described in the previous section is used to fit IL thickness data measured from irradiation tests. The interaction rate constant given in Eq. (2) was modified to consider FED by adding a square-root dependence of fission-fragment damage rate to the interaction constant as follows:

\[
k^i = A(F_r F_d)^{0.5} \exp\left(-\frac{Q'}{RT}\right)
\]  

(4)

where \( A \) is a constant, \( F_r \) is the fission rate in the U-Mo, \( F_d \) is the fission damage factor, \( Q' \) is the average effective activation energy during irradiation.

The fuel fission rate is typically given by reactor physics calculations. However, the damage distribution in the diffusion zone must be evaluated. Using the TRIM code [9], fission damage factors as a function of IL thickness were calculated for U-Mo/Al dispersion fuels. The modeling processes are discussed below.

#### 4.1 Fission rate dependence

A square-root dependence on the fission rate is similar to that obtained in ion-beam mixing experiments reported in Ref. 10. For the current case, fission damage affects FED in two ways. It amorphizes the newly grown IL and generates defects in the amorphous IL, from which the square-root dependence on the combination of \( F_r \) and \( F_d \) and also FED originate.

For IL thickness data from RERTR-1 and -2 tests, \( Y^2/t \) showed a square-root dependence on \( F_r \) at 343 K.

#### 4.2 Fission-fragment damage rate dependence

Since the distribution of fission-fragment population in the IL changes as the IL grows, the actual strength of the fission rate on IL growth also changes. We employed \( F_d \) “fission damage factor” to consider this as given in Eq. (4). We utilized TRIM results to obtain the damage distribution in the IL as it grows.

Figure 1 shows an example of TRIM results for the average distribution of instantaneous vacancy production in \((U_{0.8}Mo_{0.2})Al_3\) along the direction perpendicular to the impinging surface by a fission-fragment. Considering most of the vacancies annihilate by spontaneous recombination with interstitials and the TRIM estimate is a factor of 3 higher [10], only a small
portion of Frenkel pairs actually escape. The magnitude is not of interest. Instead, the distribution with respect to IL thickness is important because it is applied to obtain damage factors. As will be discussed later in this section, the damage factors are normalized values. A xenon fission-fragment with the initial energy of 90 MeV was chosen to represent the average fission-fragment. The range for this fission-fragment increases from 13 μm in the (U0.8Mo0.2)Al₃ to 17 μm in the (U0.8Mo0.2)Al₆.

Fission in the fuel is uniform so that the fission-fragment generation can be modeled as point sources distributed in a square array of 0.5-μm. Thus, the total vacancy production in the IL by the ions originating from the fuel at a spatial point can be obtained by summing the vacancy produced by ions starting from different sources 0.5-μm apart from each other. By using this scheme, the total damage rate expressed in the number of vacancies produced per angstrom IL length by fission-fragments originated from the fuel is obtained as shown in Fig. 2.

Because fissile uranium is also contained in the IL, fission-fragments are generated there as well. Damage created by these fission-fragments is modeled similarly to the case for fission-fragments from the fuel. The fission-fragment sources are distributed as point sources 0.5-μm apart on the line. Because fission is isotropic, half of the ions from a source go to the right and the other half go to the left. The total damage by the ions generated in the IL at a spatial point is the sum of the damages by all ions exerted at the point.

Since U density in the IL is lower than that of the fuel, for example, the ratio of U atom density of the (U₀.₈Mo₀.₂)Al₃ to that of U-Mo fuel is 0.25, the actual fission-fragment source strength in the IL is 25% of the values obtained here. By reducing the source strength this way, the total damage rate in the IL from fission-fragments from the fuel and IL can be obtained. An example for an IL thickness of 13 μm is shown in Fig. 3.

FED is the highest at the U-Mo fuel-IL interface and the lowest at the IL-Al interface as shown in Fig. 3 because of the damage contribution of the fission-fragments originating from the U-Mo particle. As the IL grows, this will cause the Al diffusion at the IL-Al interface to be less than that at the fuel-IL interface. This not only decreases the rate of IL growth, but also yields an IL...
with a lower $\text{Al/(U+Mo)}$ mole ratio at the fuel side of IL, which may, in part, explain the observation that the overall $\text{Al/(U+Mo)}$ ratio of an IL decreases as the IL grows. The lowest damage rates at the IL-Al interface controls more the IL growth than those at the fuel-IL interface. The damage rates for all IL thickness cases were collected. The procedure was followed for $\text{Al/(U+Mo)}=3, 4, 5$, and $6$. The results were normalized to the peak values of each case, and are designated as “damage factors” and plotted in Fig. 4. Because the density of the IL increases as the $\text{Al/(U+Mo)}$ ratio decreases, the damage factors for the case with $\text{Al/(U+Mo)}=3$ are the highest. The damage factors for each case decrease to the minimum for IL thicknesses greater than the fission-fragment range.

**Fig. 3** Overall damage rate distribution in the 13-μm thick $(\text{U}_{0.8}\text{Mo}_{0.2})\text{Al}_3$ by ions originated both from the fuel and the IL.

**Fig. 4** Comparison of damage factors with respect to IL thickness between IL cases with different $\text{Al/(U+Mo)}$ ratios.

### 4.3 Data fitting

As discussed in Section 2, the composition of the IL changes depending on the irradiation temperature, fuel loading, and burnup in the range of $\text{Al/(U+Mo)} = 3.3 \text{ - } 7$. The $\text{Al/(U+Mo)}$ ratio of the IL was assumed to change with the fuel temperature based on data from Refs. 3, 11-14. The $\text{Al/(U+Mo)}$ ratio was correlated with the irradiation temperature as follows:

For $T \leq 496 \text{ } K$, $x = 15.4 - 2.44 \times 10^{-2} \text{ } T,$ \hspace{1cm} (5a)

For $496 \text{ } K \leq T$, $x = 3.3.$ \hspace{1cm} (5b)

where $x$ is the $\text{Al/(U+Mo)}$ ratio and $T$ in K. From the irradiation tests, we found that $x$ always decreased irreversibly. Therefore, we used a scheme for data fitting by which $x$ remained unchanged from the previous time step for a temperature decrease and only decreased according to Eq. (5) when there was a temperature increase.

The volume fraction of each phase can be calculated from the IL thickness using the ratio of the volume of the diffusion reaction product to the fuel volume, $\beta$, given in Eq. (3) and the relation

$$r_2^3 = \beta r_0^3 + (1 - \beta) r_1^3$$ \hspace{1cm} (6)

where $r_1$ is the fuel radius at time $t$ and $r_2$ the radius of fuel plus the reaction product thickness at time $t$, i.e., $r_2 = r_1 - Y$.
As the interaction layer grows, fuel particles begin to contact one another. To account for this situation in the quantitative analysis, the contiguity of dispersed phase was considered. The correlation for the contiguity ratio was established using measured data from RERTR irradiation tests as follows:

\[ C_F = 4.005 \times 10^{-3} V_{disp} - 3.432 \times 10^{-5} V_{disp}^2 + 9.206 \times 10^{-7} V_{disp}^3 \]  

(7)

where \( C_F \) is the contiguity ratio and \( V_{disp} \) is the volume fraction in percent of the dispersed phase, i.e., the volume of the fuel and reaction product.

The calculation procedure to fit the correlation to the measured data is illustrated in Fig. 5. A simple time-marching to the end of fuel life was used by repeating the algorithm in Fig. 5. KOMO-1 and -2 irradiation test data were used for data fitting because they have wide ranges of temperatures and fission rates. For a fuel rod, the IL thickness results were compared with the known measured values. If the two were different, a new set of values for the parameters, i.e., the activation energy and pre-exponential constant, were assumed. Then, the whole process was repeated. This iteration was continued until the best-fit to the data was achieved. The same procedure was performed for other irradiated fuels. The best fitting was obtained when the average effective activation energy \( (Q_i) \) was in the range of 46 – 76 kJ/mol, and the corresponding pre-exponential factor \( (A) \) was in the range \( 1.46 \times 10^{-14} \) – \( 1.94 \times 10^{-10} \) cm\(^3\) s\(^{-0.5}\), respectively. The fitting curves are compared to the data in Fig. 6. The average effective activation energy was allowed to change with the fuel test condition, particularly with the temperature, to achieve the best fitting. Accordingly, the constant \( A \) was also changed.

5. Silicon effect on interaction layer growth

5.1 Out-of-pile data

There are two cases in the 1950s literature dealing with the addition of 3 – 12 wt\% silicon in Al to reduce interaction between U and Al. [15,16]. Figure 7 summarizes the findings from these references. They clearly show a reduction in IL growth rates with the addition of Si. The KAPL
data (Ref.15) and Hanford data (Ref.16) are consistent, generally. The temperature ranges are 250 - 600°C for the KAPL data and 200 - 300°C for the Hanford data.

We anticipated that such alloy additions might have a similar positive effect on reducing the interaction rates in U-Mo/Al dispersion fuel. Out-of-pile tests at ~550°C were performed at CNEA and KAERI [17-19]. The results from these tests are also included in Fig. 7. For (U-Mo)-Al diffusion couples at high temperatures, the effect of the addition of Si in Al on the IL growth kinetics was also substantial. Mirandou reported a test result for Al-7Si showing that the Si-effect is also observed at relatively low temperature (at 340°C). The reason for the CNEA and KAERI data to have lower rates than those from Refs. 15 and 16 is attributed to the Mo effect in U.

![Graph showing IL growth rates in U vs. Al, U vs. (Al-12Si), (U-7Mo) vs. Al, and (U-7Mo) (Al-5Si) diffusion-couple tests [15-19].](image)

**5.2 Irradiation data**

There have been several studies that either intentionally explored or accidentally revealed the efficacy of Si addition during irradiation in reducing the IL growth rate between U (or U-Mo) and Al. Richt et al. [20] showed the IL growth rate in UA13/Al dispersion was lower with a Si addition than that without it. Hofman et al. [21] noticed the Si effect when they observed a systematic lower IL growth rate in U3Si/Al, U3Si2/Al and USi/Al dispersion fuels. Meyer et al. [14] found reduced IL thickness growth at U-Mo fuel contacting Al6061 cladding compared to the pure Al matrix.

<table>
<thead>
<tr>
<th>Plate ID</th>
<th>Plate location</th>
<th>Burnup (%U-235)</th>
<th>Nominal Si-content (wt%)</th>
<th>IL thickness (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R5R020</td>
<td>C5</td>
<td>49.60</td>
<td>0.2</td>
<td>11.3</td>
</tr>
<tr>
<td>R1R010</td>
<td>A2</td>
<td>39.14</td>
<td>0.9</td>
<td>1.9</td>
</tr>
<tr>
<td>R2R010</td>
<td>C3</td>
<td>47.64</td>
<td>2.0</td>
<td>1.7</td>
</tr>
<tr>
<td>R2R020</td>
<td>B2</td>
<td>44.92</td>
<td>2.0</td>
<td>1.4</td>
</tr>
<tr>
<td>R3R030</td>
<td>B5</td>
<td>49.30</td>
<td>4.8</td>
<td>1.5</td>
</tr>
</tbody>
</table>

The RERTR-6 test was the first irradiation test that included Si-added dispersion plates, i.e., ten U-7Mo/Al-xSi (x=0.2, 0.9, 2.0, 4.8 wt%) dispersion plates and six U-10Mo/Al-ySi (y=0.1, 0.2,
0.9 wt%) dispersion plates. The plate locations in the test capsule, nominal Si content in matrix Al, and measured IL thicknesses are given in Table 1. The fission rates and temperatures of these plates are shown in Fig. 8. The effect of Si addition in the Al matrix is prominent comparing the ILs between R5R020 and R3R030. The fission rates and temperatures of these plates are similar as shown in Fig. 8. These data were used to develop a correlation to calculate IL thicknesses for the Si-added plates. In order to match the predictions to the measurement, we reduced the pre-exponential constant (i.e., A in Eq. (4)). The ‘reduction factors’ defined as \( A(\text{Si-added Al})/A(\text{pure Al}) \) are plotted in Fig. 8(c).

![Fig. 8](image)

Fig. 8  (a) Average fission rates in fuel particles, (b) temperatures at plate-mid region, (c) reduction factors for the square of interaction layer thicknesses of Si-added RERTR-6 plates.

6. Discussion

6.1 Variable activation energy

In Fig. 9, for the out-of-pile annealing test data, a straight line is obtained whereas it is a curve for the irradiation data. This indicates that the average effective activation energy varied during irradiation. This apparent change in effective activation energy with temperature is attributed to a composition change of the IL, a change that is possible when the IL is amorphous.

The error bars on temperature indicate the temperature variances during the tests. As the error bars show, except for low temperature tests, the fuel temperature usually changes substantially during the test. It is for this reason that the growth of the IL needs to be modeled in an iterative way, and Fig. 9 should therefore be taken as an illustration of the fission enhanced interdiffusion presented here.

![Fig. 9](image)

Fig. 9  Comparison of \( Y^2/t \) of annealing data and \( Y^2/(F_r/F_{r,avg})^{0.5} t \) of irradiation test data. For comparison, the irradiation test data were divided by their normalized fission rates with the average fission rate.
6.2 Square-root dependence on fission rate
We tried fitting with the power of the fission rate in the range of 0.5 – 1. The best fitting was achieved when we used 0.5. This is consistent with the crystalline radiation-enhanced diffusion (RED) theory [10], advocating the square-root dependence on the fission rate when defect annihilation is dominated by recombination, not by annihilation at sinks. Considering the availability of sinks such as excess free volume in amorphous materials, this appears contradiction. However, Averback and Hahn [10] showed that RED takes place in amorphous materials with kinetics similar to those in crystalline materials, i.e., with the square-root ion-flux dependence by “point-defectlike entities” that undergo recombination in the amorphous material. Since the purpose of the present work was to develop an interdiffusion correlation for use in fuel behavior modeling based on fitting a plausible rate equation to measured data and calculated test parameters, we considered a further pursuit for the physical meaning of the correlation belonged to the scope of an academic interest.

Fortuitously, however, some of the RERTR-6 test plates have the same numbers of $t F_r^{0.5}$ (i.e., time multiplied by the square-root of fission rate) and fuel temperatures as those of RERTR-7 although the individual $t$ and $F_r$ are very different. Thus, if the square-root dependence is valid in $Y^2 \propto t F_r^{0.5} \exp(-Q^f/RT)$, there must be some pairs between RERTR-6 and -7 that have the same measured IL thicknesses. To exclude the $Q^f$ dependence, of course, the plates with the same Si-content should be considered. When the RERTR-7 PIE is available and the results turn out this way, we will be able to confirm once again the validity of the current modeling.

6.3 Si-effect on IL thickness growth
In Fig. 7, the KAPL data show the effect of a Si addition in Al seems to increase with temperature. The Hanford data, however, obtained at lower temperatures than the KAPL data, show the opposite trend. Because the KAPL data were the only data obtained under the same condition for Si-added or no Si-added samples, we were inclined to rely on these data. Therefore, there still remain uncertainties regarding the right trend for the U-Al case.

The high temperature data from CNEA [17,18] and KAERI [19], with U-Mo instead of pure U, showed a different trend on the Si effect. The Si-effect was not discernable in some tests at high temperatures because the $\gamma$-phase decomposition effect was so large that the Si-effect was masked, whereas some lower temperature tests showed the effect. Both CNEA and KAERI data consistently show that the IL growth rate decreases as the Si-content increases. Comparing the data for (U-Mo) vs (Al-7Si) at 340 and 550°C reported by CNEA, the Si-effect seems to follow the similar temperature trend as the KAPL data. However, since the temperature trend of the Si-effect has never been systematically studied with the $\gamma$-phase U-Mo, judging the trend with only these data has not been successful.

Figure 8(c) shows that the reduction factor decreases exponentially as the Si-content in Al increases. Although this is an assessment of limited data, it clearly shows that the IL growth rate decreases as the Si-content increases. When more data are collected to elucidate the effect of Si-addition from RERTR-6 and -7, modifications for the activation energy for IL growth ($Q^f$) and the pre-exponential constant ($A$) may be obtained. Therefore, the reduction factors presented here should be considered as tentative results.
7. Conclusions

An IL growth correlation for spherical U-Mo particles dispersed in matrix Al was obtained based on data from high temperature annealing tests of U-Mo/Al dispersion fuel samples. The correlation has a classical Arrhenius form with a pre-exponential factor obtained from the best-fit as \( k_0 = 3.94 \times 10^{16} \text{ \mu m}^2/\text{s} \), and an effective activation energy as \( Q = 300 \text{ kJ/mol} \).

The reaction rate constant obtained from irradiation tests is: \( k_i = A(F_r {F_d}^{0.5}) e^{-Q_i / RT} \), where \( F_r \) is the fission rate in the fuel particles in \( \text{fiss/cm}^2\cdot\text{s} \), \( F_d \) is the unitless fission damage factor, \( Q_i \) is the average effective activation energy during irradiation. \( Q_i \) was best fitted within a range of 46 – 76 kJ/mol, and the corresponding \( A \) was in the range \( 1.46 \times 10^{-14} – 1.94 \times 10^{-10} \text{ cm}^{3.5} \text{s}^{-0.5} \), respectively.

The measured data from the RERTR-6 test showed that reduction factors for the IL thickness prediction decrease exponentially with the Si-content. When more data are collected from RERTR-6 and RERTR-7, the new correlation for the Si-added plates may be obtained.

Acknowledgments

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References

[6] Recent observations from IRM (Zarechny, Russia) and RIAR (Dimitrovgrad, Russia), Private communication, 2006.
[19] J.M. Park et al., This meeting.