

Interaction Layer Characteristics in U-xMo Dispersion/Monolithic Fuels

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November 2010

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This report is a result of a multinational/multi-laboratory
strategy to produce a data bank for understanding fuel/matrix interactions in
Reduced Enrichment for Research and Test Reactors fuel



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SUMMARY

Published data concerning the interaction layer (IL) formed between U-xMo fuel alloy and aluminum (Al)-based matrix or cladding materials was reviewed, including the effects of silicon (Si) content in the matrix/cladding, molybdenum (Mo) content in the fuel, pre-irradiation thermal treatments, irradiation, and test temperature. The review revealed that tests conducted in the laboratory produce results different from those conducted in an irradiation environment. However, the laboratory testing relates well to thermal treatments performed prior to irradiation and helps in understanding the effects that these pre-irradiation treatments have on in-reactor performance. A small, Si-enriched IL, formed during a step in the fabrication process, seems to be stable during irradiation, helping to prevent the rapid growth of an irradiation-induced IL. Moreover, the Si-enriched IL seems to be important in delaying the onset of rapid growth of fission gas bubbles.

Conclusions from irradiated fuels data have been repeated many times in the literature review. However, as related to the “Desired Characteristics of the IL” mentioned near the beginning of this report, several more conclusions can be drawn:

1. An IL with phases akin to UAl_3 is desired for optimum fuel performance, but at low temperatures, and especially in an irradiation atmosphere, the desired (Al+Si)/(U+Mo) ratio of three is difficult to produce. When the fuel operating temperature is low, it is important to create a pre-irradiation IL enriched in Si. This pre-formed IL is relatively stable, performs well in terms of swelling resistance, and prevents rapid IL growth during irradiation. Fabrication-related heat treatments should be limited in order to maintain a thin, Si-enriched layer containing potentially beneficial phases.
2. At higher operating temperatures ($>150\text{--}170^\circ\text{C}$), IL formation in reactor may not be so dependent on pre-irradiation IL formation, especially at high burnup; a pre-fabricated IL seems to be less stable at high burnup and high operating temperature. Moreover, the (Al+Si)/(U+Mo) ratio of three occurs more often at higher temperature. For these two reasons, it is important at high operating temperature to also have a matrix with significant Si content to create an IL in-reactor with the right characteristics.
3. Out-of-reactor testing seems to indicate that Si in the matrix material is required in some concentration (2%, 5%, ?) to provide for a thin, Si-enriched IL formed before irradiation of a fuel plate. It ensures that the IL contains beneficial phases or prevents formation of some known to promote poor fuel performance. Significant progress has been made in determining the desired characteristics of the IL.
4. The use of a fuel with stable gamma phase appears to allow more predictable performance regarding both a beneficial pre-irradiation layer and the fuel performance (low swelling) to high burnup. Destabilization of the gamma phase may create problems with IL breakaway growth.
5. A theory whereby prevention of the $\text{U}_6\text{Mo}_4\text{Al}_{43}$ complex phase in interaction layers formed during fabrication may be a key to good irradiation performance. Si additions to the matrix allow for solubility of Mo in the desirable

(U,Mo)(Al,Si)₃ or perhaps (U,Mo)(Al,Si)₄ phase, helping to prevent formation of the complex phase. Keeping alloy Mo content as low as possible may also help so long as decomposition does not occur in fabrication, forcing Mo into the interaction layer. This theory may explain a number of apparent anomalies observed in testing results.

6. More work is needed in order to prescribe the conditions to best produce a beneficial IL. Another necessity is a better understanding of any correlation between beneficial characteristics of the prefabricated IL and the irradiation conditions to which it will be subjected.

Two spreadsheets/databases were constructed to compile this data. One contains data obtained from irradiated samples, including those irradiated by neutrons, heavy ions, or protons. The other contains data obtained from samples subjected to thermal treatments in the laboratory. The data was used to test some of the theories proposed/re-stated as a result of the literature review. The results were less than satisfactory. Because a large number of experimental criteria were varied between tests, accurate comparisons could not be made. These results indicate that a more systematic, comprehensive set of experiments should be conducted to test these theories.

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ACRONYMS

| | |
|-------|---|
| EDS | energy dispersive spectroscopy |
| EPMA | electron probe micro-analysis |
| FSW | friction stir welding |
| HIP | hot isostatic press |
| IL | interaction layer |
| HEU | highly enriched uranium |
| LEU | low-enriched uranium |
| NDA | neutron diffraction analysis |
| OM | optical microscopy |
| PFZ | precipitate-free zone |
| RERTR | Reduced Enrichment for Research and Test Reactors (program) |
| RRFM | research reactor fuel management |
| SEM | scanning electron microscopy |
| TEM | transmission electron microscopy |
| XRD | x-ray diffraction |
| XRF | x-ray fluorescence |

Interaction Layer Characteristics in U-xMo Dispersion/Monolithic Fuels

1. INTRODUCTION

One goal of the Reduced Enrichment for Research and Test Reactors (RERTR) program is to replace research reactor fuels based on enriched uranium ($\geq 20\%$ $^{235}\text{U}/\text{U}$) with low-enriched uranium (LEU) fuels because these reactors are often remotely located and have little physical security. To date, the program successfully replaced many fuel types with a dispersion fuel using uranium silicide. Other reactors have fuel and reactor core designs that require fuels with a higher uranium density.

The use of U-xMo-based fuels is of great interest because uranium density can be enhanced compared to the uranium-silicide-based fuel. The development process associated with this fuel type requires acceptable (no cladding breach, low swelling, etc.) and predictable fuel performance. The fuel must also be easily fabricated, with fuel performance tolerant of minor variations in the fabrication process.

The performance of U-xMo dispersion fuels, the first type to be developed, is responsive to small variations in the alloying of the Al-based matrix material. The interaction layer (IL) that forms due to interdiffusion of fuel and matrix (dispersion fuel) or cladding (monolithic fuel) plays an important role in fuel performance. The layer can create dimensional changes in the fuel¹ due to the density changes when new phases form as fuel and matrix species mix. The layer may possess a thermal conductivity that affects fuel performance,² or it may be brittle such that, in the case of monolithic fuel, fuel/cladding separation could result.³

Of particular interest is fission-gas-induced swelling in the IL. The swelling of the U-xMo fuel is predictable,⁴ but the swelling of the fuel element as a whole (plate or rod) is not. This variation in swelling has been correlated with the IL in that gas bubbles form preferentially in the IL. While some of the variables in matrix alloy or fabrication parameters appear to correlate with the characteristics of IL formation and fuel performance, these correlations have not been explained.

This paper will review the characteristics of IL formation and analyze the body of available data to trend these characteristics against composition, temperature, and other variables thought to influence IL formation.

2. DESIRED CHARACTERISTICS OF THE INTERACTION LAYER

The manuscripts that discuss fuel performance and its relationship with the characteristics of the fuel/matrix interaction show diverse opinions on the details of this relationship, but common opinions on the desired characteristics of the IL based on experimental observation include:

1. The IL should be thin as produced by fabrication and grow slowly in-reactor
2. $(U,Mo)(Al,Si)_3$ should be the primary phase (seems to be particularly resistant to the nucleation and growth of fission gas bubbles)
3. The IL should be enriched in Si, especially near the fuel if stratified.

A review of the studies from which these ideas have come may help to understand the ideas and a collection of this data into one database may help to analyze the set.

Another goal is to understand what fabrication steps (particularly the time-at-temperature for elevated temperature processes), matrix alloy compositions, fuel compositions (Mo concentration), and irradiation conditions create these characteristics.

3. IL DATABASE – LITERATURE REVIEW

Many reports were gathered in an attempt to review all data as a single set. However, two data sets were created: one where radiation was used to create the interaction layers and another in which radiation was not used.

Three different methods were employed in studies not using radiation. One involved using diffusion couples created by mating fuel and matrix/cladding materials and heating them to stimulate interdiffusion, creating the interaction layer(s). These were assembled using traditional diffusion-couple techniques. Similar tests were also done where the “couples” were first bonded together using friction stir welding (FSW) and then subjected to high temperatures. The third type of test involved the heat treatment of dispersion fuels, in which the IL develops where the Al alloy matrix surrounds the particle of fuel.

Testing that involved irradiation usually consisted of post-irradiation examinations (PIEs) of as-irradiated dispersion fuels. In some cases the fuel was not fissioned, but the dispersion fuel was irradiated using heavy ions. Tests are also reviewed here where compositions observed in interaction layers were re-created by casting an alloy with similar composition to an IL or to phases within an IL. These materials were then heated and irradiated using energetic ions or protons.

To assemble the data sets, two spreadsheets were created: one populated with data created in the absence of irradiation, and the other generated from characterization of irradiated materials. The following sections describe the source references for the data and discuss this literature and others where data was not extracted.

3.1 Testing not Involving Irradiation

Most of the studies that have been done have been out-of-reactor due to several advantages over performing experiments in-reactor. These experiments typically are less expensive and much less time consuming because complex tests need not be designed, fabricated, or qualified, and no time in-reactor or accelerator is required. Two main types of ex-reactor study are conducted: diffusion couple studies and dispersion fuel studies. In both cases, the U- and Al-based alloys are bonded and annealed at relatively high temperatures and short times compared to reactor operating conditions.

3.1.1 Data Categories and Reference Listing

The categories of interest used to populate the spreadsheet for testing performed in the laboratory were:

Testing without Irradiation

- Method of IL creation
- Temperature
- Time at temperature
- Mo concentration in the fuel
- Si concentration in the matrix/cladding
- Characterization techniques
- IL composition: U, Mo, Al, Si
- Ratios: (Al+Si)/(U+Mo), Al/Si, (Al+Si+Mo)/U
- IL thickness
- IL phases identified.

Information to populate this spreadsheet was obtained from the following publications:

- Allenou, J., et al., RRFM, Vienna 2009⁵
- Allenou, J., et al., J.Nucl. Mater. , v.399 (2010)⁶
- Cornen, M., et al., RRFM, Lyon, 2007⁷
- Cornen, M., et al., RRFM, Hamburg, 2008⁸
- Dubois, et al., RRFM, Budapest, 2005⁹
- Keiser, D. D., et al., RERTR 2008 – 30th IM, Washington DC¹⁰
- Keiser, D. D., et al., Defects and Diffusion, 2007¹¹
- Kim, Y. S., et al., RERTR 2005 – 27th IM, Boston¹²
- Lee, J. S., et al., J Nucl. Mater., 2002¹³
- Mazaudier, F., et al., RRFM, Sofia, 2006¹⁴
- Mazaudier, F., J Nucl. Mater., 2008¹⁵
- Mirandou, M. I., et al., J Nucl. Mater., 2003¹⁶
- Mirandou, M. I., et al., RERTR 2002 – 24th IM, San Carlos¹⁷
- Mirandou, M. I., et al., RERTR 2005 – 27th IM, Boston¹⁸
- Mirandou, M. I., et al., RERTR 2004 – 26th IM, Vienna¹⁹
- Mirandou, M. I., et al., J Nucl. Mater., 2009²⁰
- Mirandou, M. I., et al., RERTR 2007 – 29th IM, Prague²¹
- Mirandou, M. I., et al., RRFM, Sofia, 2006²²
- DeLuca, L. S., Knolls Atomic Power Laboratory Report, 1957²³
- Palancher, H., et al., RERTR 2008 – 30th IM, Washington DC²⁴
- Palancher, H., et al., “ ,” J. Appl. Crystallogr., 2007²⁵
- Park, J. M., et al., J Nucl. Mater., 2008²⁶
- Park, J. M., et al., RERTR 2006 – 28th IM, Cape Town²⁷
- Park, J. M., et al., RERTR 2005 - 27th IM, Boston²⁸
- Perez, E., et al., Defect and Diffusion Forum, 2007²⁹
- Perez, E., et al., RERTR 2009 - 31st IM, Beijing³⁰
- Ryu, H. J., et al., JPED 2006³¹
- Ryu, H. J., et al., J Nucl. Mater., 2003³²
- Ryu, H. J., TMS Letters, volume 3, 2006³³
- Ryu, H. J., et al., RERTR 2006 – 28th IM, Cape Town³⁴
- Mirandou M., et al., Mater. Charact. 2009³⁵
- Yang, J. H., et al., RERTR 2009 - 31st IM, Beijing³⁶

3.1.2 Comments

Each study produced observations related to several variables, such as Si or Mo content, temperature, the mixture of phases that form in the layer, the relationship to precipitate-free zones (PFZs) in the matrix material, and stratifications of the IL. In addition to the data collected, it is also important to review these observations to find commonalities between bodies of research.

3.1.2.1 *Mo and Si Content*

Various studies have been performed in order to determine IL thickness as a function of Mo and Si content in the U and Al alloys, respectively. Based on the literature, Mo does not have as strong of an influence as Si does. Park et al.²⁶ conducted a study in diffusion couples using binary Al-Si alloys with 2 and 5 wt% Si to compare results with those using pure Al. These results reveal a clear decrease in IL thickness with the addition of Si; however, they also indicate that a Si threshold exists, after which it is no longer beneficial to add more Si to the system. Both Cornen et al.⁸ and Perez et al.²⁹ confirm this result with several diffusion couples containing various amounts of Si. Cornen et al. present thickness versus Si content data that shows a downward trend with a Si threshold value of about 5 wt%, with U-10Mo at 550°C, while the data Perez presents shows a very similar trend at 550°C, but with a threshold value of about 2 wt%. Note, however, that the layer thickness after one hour at 550°C that Perez showed was much less than that of Cornen for all Si concentrations. The rate of growth may be very sensitive to material and experimental conditions. Both agree, however, that Si content less than 2 wt% has very little influence on IL thickness.

Allenou et al.⁶ has recently reviewed data from 450°C diffusion couple experiments and showed evidence for microstructure and phase composition differences that perhaps helps explain the differences exhibited by fuels with high and low Si matrix compositions. With higher Si, the layers are thinner, with a highly Si-enriched sub-layer near the fuel and no Si-free complex phases. With lower Si contents, the sub-layer near the fuel is depleted in Si. Of more importance, perhaps, is that the phases present in the layered ILs change with Si content of the matrix. Allenou et al.⁶ noted this, showing there were no Si-bearing phases next to the fuel if the Si content was less than 5 wt% (see more detail below when phase content is discussed).

Others also studied the effect of Si content in the matrix on the composition of the IL. In annealing dispersion fuels, and based on energy dispersive spectroscopy (EDS) and electron probe micro-analysis (EPMA) evidence, Ryu et al.³¹ observed that a fuel plate annealed at 580°C for 10 hours did not have any evidence of Si accumulation in the IL; however, when annealed at 550°C for 25 hours, a 20 at.% Si was present in the IL. In diffusion couples, Mirandou et al.,¹⁹ Cornen et al.,⁷ and Park et al.²⁶ report the accumulation of Si in the IL. In FSW diffusion couples, Keiser et al.¹¹ mentions that the Al and Si content vary significantly within the IL, but the sum of the two remains constant around 70 at.% based on EDS/transmission electron microscopy (TEM) results. Very similar results were found by Mirandou et al.^{20,35} These results are consistent with most of the diffusion couple studies.

It has long been shown that Si additions to the U-Al mixture suppress the formation of the UAl₄-type phase, promoting and stabilizing the UAl₃-type phase. Thurber and Beaver³⁷ showed that Si additions to 48U-52Al (wt%) showed an increase in the UAl₃/(UAl₃+UAl₄) ratio from 0 to 100% as Si is added, up to 3 wt% Si. Chakraborty et al.³⁸ showed that when starting with a UAl₄-type phase and then annealing to transform to UAl₃, the transition could be suppressed by adding Si. Mirandou et al.,¹⁹ using x-ray diffraction (XRD), reported this fact in a U(Mo)/Al (Si) diffusion couple.

A soon-to-be-published article by Perez et al.³⁹ discusses the results of diffusion couples of U-7, 10, and 12Mo with only pure Al at 600°C. They find the increasing Mo content may increase the tendency to form U₆Mo₄Al₄₃, a phase, later in this review, shown to be unstable under irradiation. The implication is that if the U-xMo alloy can be kept stable during IL formation, lower Mo concentrations may be better. Carrying this theory forward and applying the idea that Si increases the solubility for Mo in more

favorable phases may help explain many anomalies in the experimental data (see irradiation section later in this report).

3.1.2.2 The Precipitate-Free Zone (PFZ)

Because Si is thought to be the most influential factor in controlling IL thickness, a lot of work has been done to understand the role it plays in this interaction. Kim et al.,¹² through thermodynamic considerations, estimate the stability of the IL when Si is added to Al. The authors mention that experimental evidence supporting the predictions is given in the works of Mirandou et al.¹⁹ and Park et al.²⁸ Mirandou et al.^{19, 22} mention a diffusion couple in which a 50- μ m-thick Si-depleted zone was observed; consequently, a high-Si content IL with composition approximately $(U, Mo)(Al_{0.67}, Si_{0.33})_3$ was observed. Mirandou has referred to this Si-depleted region as a PFZ because it is thought that the Si precipitates originally present in the Al alloy serve as a Si reservoir and diffuse into the IL region upon annealing to form the high-Si content ILs.²² Mirandou²⁰ presents data suggesting that the PFZ depends on the Al (Si) alloy and the temperature. For example, at 550°C, the PFZ is typically 4 to 6 times larger than the IL (which contains 25% Si) when using Al A 356 (7% Si) matrix and is not observed in case of an IL containing 12 % Si and a matrix of Al 6061 (0.6% Si).. At 340°C for Al 6061, the PFZ is observed, and the IL contains 47% Si. Cornen et al.⁷ has also reported a Si-depleted region in the Al alloy near the IL and a Si-rich region near the fuel side or near the Al side of the IL, depending on the Si content in the Al matrix. This differs slightly with observations made by Park et al.²⁶ where he mentions that the IL Si content varies significantly but has found high-Si content regions near the Al side of the IL. Also, he comments that in some couples, the Si content drops near both the U and Al interface with the IL, but the Al + Si content remains constant at approximately 80% as determined by EPMA.

Diffusion couples produced by FSW and using Al alloys with Si also show regions of high Si in the IL. This has been observed in U(Mo)/Al 6061 by Keiser¹¹ and Mirandou et al.³⁵ at 500°C and 550°C. Both works agree in that no PFZ is observed but Si is accumulated in the IL. At 340°C Mirandou et al.³⁵ reported Si content as high as 58 at.% within the IL and the presence of a PFZ. Similar behavior was observed in U(Mo)/Al A356 (7 wt% Si) by Mirandou et al.²⁰ at 550°C and 340°C. These results in FSW couples are again consistent with the ones obtained in chemical diffusion couples.

3.1.2.3 Multi-Layered IL

The observation of different layers or stratifications within the IL has also become a point of interest to scientists studying this system. Mazaudier, Mirandou, and Ryu have all observed three layers within the IL in the UMo/Al system.^{14, 17, 19, 32} The composition of these three layers is not yet agreed upon based on this literature, but each of these articles suggests the presence of three layers. While Mirandou observed all three layers, she asserts that there is no clear boundary between them, while the others suggest the layers are distinct. Ryu's diffusion couple work produced three layers, but out of the aforementioned works, this is the only one that does not identify a ternary phase present within the IL. However, the third layer is listed as an unidentified phase and, upon further research, could be determined as a ternary compound. This may be important when considering the addition of Si, because Si enrichment could be created by enrichment of the binary-based phases, UAl_3 or UAl_4 with Si substitution for Al, or by formation of a new phase that would depend on the presence of all three elements. The properties of the IL may vary depending on how the IL was enriched.

The formation and characteristics of multiple layers may be related to the addition of Si to the Al. Mirandou showed that two-layer structures were formed when Si was alloyed in the matrix material.¹⁹ Cornen originally suggested that the IL is comprised of two layers when Si is included.⁷ However, as co-author with Allenou in a paper that further examines the earlier results, Cornen mentions the observation of three layers.⁵ Allenou determined that three layers exist based on a study conducted at 450°C, and the composition and thickness of those layers are influenced by the amount of Si present in the system. This study presented results for two types of couples: those with Si content between 2 and 5 wt% and those with greater than 5 wt% Si. The most recent reference from Allenou et al.⁶ states that

450°C treatments produce bi-layers regardless of Si content, but the phases present are different if the matrix Si content is above or below 5 wt%. Mazaudier speculated on the phase make-up of the three layers seen in that study,¹⁴ but they are different from those presented originally by Allenou. Park et al. showed stratified and layered structures when Si was present in the matrix material, especially when the U-Mo contained 2% Zr.²⁶

3.1.2.4 Phase Identification

Due to the complex microstructure of the IL, it has been difficult to firmly agree upon the identification of the phase constituents. According to the most recent work, the IL may contain the ternary compounds $\text{UMo}_2\text{Al}_{20}$ and $\text{U}_6\text{Mo}_4\text{Al}_{43}$. This assertion is based on work done using XRD, micro-x-ray diffraction (μXRD), and TEM, which determine crystal structure and can accurately identify the compounds of interest in the UMo versus the AlSi system. Since the compositions of all of these compounds are so similar, these diffraction techniques must be used. With XRD, Mirandou identified the four compounds: (1) $(\text{U},\text{Mo})\text{Al}_3$, (2) $(\text{U},\text{Mo})\text{Al}_4$, (3) $\text{UMo}_2\text{Al}_{20}$, and (4) $\text{U}_6\text{Mo}_4\text{Al}_{43}$.¹⁶ Following this work, both Allenou⁵ and Palancher²⁴ used μXRD for verification. The results of both studies establish the presence of these four phases, except Allenou does not observe UAl_4 and instead mentions the presence of $\text{U}_3(\text{Al},\text{Si})_5$ because the study involved the addition of Si to the system.

Some of the earliest results with Al(Si) by XRD were given by Mirandou et al.¹⁸ The researchers indexed the phase of the IL formed at low temperature (340°C) as $\text{U}(\text{Al},\text{Si})_3$ and suggested that the phase was $(\text{U},\text{Mo})(\text{Si},\text{Al})_3$. The formation of an IL at low temperatures (<500°C) leads to a layer concentrated in Si.

In another investigation of the IL phase constituents, Perez performed TEM and electron diffraction analysis and identified the $(\text{U},\text{Mo})\text{Al}_3$, $\text{UMo}_2\text{Al}_{20}$, and $\text{U}_6\text{Mo}_4\text{Al}_{43}$ phases again, excluding UAl_4 .²⁹ While this work confirms the presence of these phases, there is still debate on the development of the correlated microstructure.

Using couples formed by FSW and after a thermal treatment at 550°C, Mirandou et al.^{20,35} identified UAl_3 , $\text{U}(\text{Al},\text{Si})_3$, and $\text{U}_6\text{Mo}_4\text{Al}_{43}$ phases in the IL using XRD. They conducted an investigation using high-intensity XRD with synchrotron radiation.²¹ At 550°C, Mirandou reported the presence of the $\text{U}(\text{Al},\text{Si})_3$, $\text{UMo}_2\text{Al}_{20}$, and U_3Si_5 phases, and at 340°C the same phases were identified excluding the ternary compound. These results agree somewhat with studies performed via mechanically clamped couples, but some discrepancies remain.

Annealed dispersion fuel tests compare favorably with diffusion couple experiments. Using EDS and EPMA, Kim and Ryu determined the phases present in the IL.^{12,32} Kim identified only $(\text{U},\text{Mo})(\text{Al},\text{Si})_3$ with the addition of Si in the matrix while Ryu identified $(\text{U},\text{Mo})\text{Al}_3$ and $(\text{U},\text{Mo})\text{Al}_4$ when pure Al was used in samples annealed at 550°C. However, Lee did neutron diffraction analysis (NDA) to determine phases and observed UAl_2 and UAl_3 in the IL of fuels annealed at 400 and 500°C. When the dispersion fuel system was analyzed using μXRD , different results were obtained. Palancher identified UAl_2 , UAl_3 , and $\text{U}_6\text{Mo}_4\text{Al}_{43}$ in ILs formed during anneal at 600°C.²⁴ In a separate publication, Palancher²⁵ identified the presence of $(\text{U},\text{Mo})\text{Al}_3$, $(\text{U},\text{Mo})\text{Al}_4$, $\text{UMo}_2\text{Al}_{20}$, and $\text{U}_6\text{Mo}_4\text{Al}_{43}$ in a diffusion couple and a dispersion fuel annealed at 600 and 500°C, respectively. These results are somewhat contradictory, although the results based on the dispersion fuels are based on tests run at different temperatures.

In a very recent study, Keiser et al.⁴⁰ attempted to identify the phases formed in ILs created during fabrication steps of dispersion fuels, specifically U-7Mo in a matrix of 4043 Al (4.8 wt% Si) or an Al-2 wt% Si alloy. The layers were created by the initial roll bonding (500°C for one hour) and a blister anneal treatment (485°C for thirty minutes). The plates studied then received an additional hot isostatic pressing (HIP) process (seventy-five minutes heat-up to 500°C and hold for 30 minutes) to ensure a good bond. Basically, a series of heat treatments resulted in exposure to ~500°C for a little more than 2 hours. The TEM and scanning electron microscopy (SEM) characterization was particularly detailed. The gamma

phase in the U-7Mo particles partially decomposed, and an IL formed in all cases. A thicker IL formed adjacent to decomposed areas in the fuel particle. The thin regions contained $U(Al,Si)_3$, $U_3Si_3Al_2$, $USi_{1.88}$ and U_3Si_5 . The thicker interaction layers were Si deficient compared to the typical thin IL and contained $U(Si,Al)_3$ and $U_6Mo_4Al_{43}$.

A previous study by Keiser et al.¹⁰ characterized a similar fuel plate with Al-2Si matrix material but did not identify the $U_6Mo_4Al_{43}$ phase. The authors noted in the current article⁴⁰ that others had also observed the U-Si binary phases in diffusion couples where the “matrix” alloy contained a high concentration of Si (>5 %). The fact that very different layers can be formed with fairly minor changes in heat treating conditions implies that selection of fuel and matrix materials may be very important if a particular phase mixture and composition of the IL is desired.

In addition, that study is supported by a contemporaneous characterization of diffusion couples where U-7, 10 and 12 wt% Mo were heated at 600°C for 24 hours in contact with pure Al, and in contact with pure Al, Al-2 wt% Si and Al 5% wt% Si at 550°C for up to twenty hours.³⁰ The same authors participated in this study. UAl_3 , UAl_4 , $U_6Mo_4Al_{43}$, and UMo_2Al_{20} were all observed in the couples without Si, and the Al concentration remained constant at 80% across the IL. In the other set of tests, with Si included, $(U, Mo)(Al,Si)_3$ was found with considerable concentrations of Mo and Si, and UMo_2Al_{20} . The UAl_4 -type phase and the $U_6Mo_4Al_{43}$ were not found. These results are consistent with the results from many of the other studies, confirming the effect of Si. This specific variation in the phases present in the IL as a function of Si additions to the Al will be addressed again later in this report when the results of irradiation testing are discussed. They become part of an interesting theory as to the influence of the Si on irradiation performance.

The earlier work on this subject relied heavily upon EDS and EPMA to perform phase identification, which are composition-based techniques that cannot distinguish different phases with similar compositions. For this reason, most of the literature first identified the IL as either UAl_3 or UAl_4 because the average compositions matched these compounds.

Perez, Kim, and Ryu used a combination of EDS and EPMA to determine the phases present.^{29,12,32} Based on the average composition determined by EPMA, Perez established that the IL was the $(U,Mo)Al_4$ intermetallic compound with Mo substituting for U. Kim suggests that the IL is the $U(Al,Si)_3$ with a high concentration of Si substituting for Al. However, Ryu observed the presence of both UAl_3 and $UAl_{4.4}$ since UAl_4 has a solubility range. While most account Mo as a substitution for U, Cornen suggests that it could substitute for Al or Si.⁸ Based on this and EDS data, Cornen proposes that the phase constituents of the IL are UAl_2 and UAl_3 .

3.1.2.5 Effect of Temperature

Temperature effects have been alluded to in the previous discussions. Both Ryu and Mazaudier assert that the formation of layers within the IL is temperature dependent.^{32,15} Based on these results, it appears that 500°C is the threshold temperature below which the IL remains one layer and above which two or even three layers form. Ryu mentions that when the dispersion fuels were annealed at 500°C, only one layer formed, but at temperatures of 525 or 550°C, two or more layers formed.³² Note that these experiments did not include Si. In another paper in 2006, Ryu observes three layers in dispersion fuels annealed at 550, 580, and 600°C, but no phase identification was given for the third layer.³² In this same study, recall that Ryu et al.³² noted that Si concentrated in the IL only at temperatures less than 580°C.

Mazaudier also presents similar data showing that at 440 and 500°C, the IL consists of only one layer while at 550°C, the IL is in general divided into two and sometimes three zones.¹⁵ This temperature-dependent stratification seems to apply to both diffusion couple and dispersion fuel anneal experiments.

Mirandou et al.'s investigations often made comparisons between results obtained from testing at 340°C to those created at 500 or 580°C.^{20,35} While the same phases were often found regardless of

temperature, Si was more easily concentrated in the IL at 340°C, especially if the matrix had a lesser amount of Si.

3.2 Testing Involving Irradiation

Irradiation testing was examined separately from the ex-reactor studies because there are several factors that could make the process of IL formation very different. One is that under irradiation, especially at low temperature, some of the IL phase can exist with an amorphous structure. Another is the diffusion processes occur at high rates at much lower temperatures (higher temperatures are used in the laboratory studies to enhance diffusion without irradiation). In addition to these effects, if the fuel is fissioning, fission fragments are thought to destabilize precipitates in the Al alloys and facilitate PFZ formation.

3.2.1 Data Categories and Reference Listing

The categories of interest used to populate the spreadsheet reflecting the ‘Irradiation Testing’ data were:

Testing with Irradiation

- Mo concentration of fuel
- Si concentration in the matrix/cladding
- Peak temperature
- Enrichment
- Uranium density
- Ion fluence
- Average fission density
- Burnup, at.% ²³⁵U
- Characterization techniques
- IL composition; U, Mo, Al, Si
- Ratios: (Al+Si)/(U+Mo), Al/Si, (Al+Si+Mo)/U
- IL phases identified.

Note that “phases identified” is recorded without a footnote only if a technique was utilized where crystal structure could be identified. Often a phase is presumed to be present because microchemical analysis indicates the proper ratios of elements present. These too are often listed in the database but with a footnote or only in a “comment” section.

Information to populate the “Irradiated” database was obtained from the following publications:

- Conlon, K. T. and Sears, D. F., RRFM, Sofia, 2006⁴¹
- Conlon, K. T. and Sears, D. F., RRFM, Lyon, 2007⁴²
- Gan, J., et al., RRFM, Vienna, 2009⁴³
- Gan, J., et al., RRFM, Hamburg, 2008⁴⁴
- Golosov, O. A., et al., RRFM, Lyon, 2007⁴⁵
- Golosov, O. A., et al., RERTR 2007 – 29th IM, Prague⁴⁶
- Golosov, O. A., et al., RRFM, Hamburg, 2008⁴⁷

- Hofman, G. L., et al., RERTR 2006 – 28th IM, Cape Town⁴⁸
- Hofman, G. L., et al., RERTR 2003 – 25th IM, Chicago⁴⁹
- Keiser, D. D., et al., RRFM, Hamburg, 2008⁵⁰
- Keiser, D. D., et al., RRFM, Vienna, 2009⁵¹
- Keiser, D. D., et al., Global 2009, Paris, 2009⁵²
- Kim, K. H., et al., Nucl. Eng. & Des., 2002⁵³
- Kim, Y. S., et al., RERTR 2006 – 28th IM, Cape Town⁵⁴
- Leenaers, A., et al., J Nucl. Mater., 2004⁵⁵
- Leenaers, A., et al., RRFM, Hamburg, 2008⁵⁶
- Leenaers, A., et al., J Nucl. Mater., 2008⁵⁷
- Meyer, M. K., RERTR 1999 – 22nd IM, Budapest⁵⁸
- Meyer, M. K., et al., J Nucl. Mater., 2002⁵⁹
- Miller, B. D., et al., RERTR 2008 – 30th IM, Washington DC⁶⁰
- Palancher, H., et al., J Nucl. Mater., 2009⁶¹
- Palancher, H., et al., RRFM, Sofia, 2006⁶²
- Park, J. M., et al., RRFM, Hamburg, 2008⁶³
- Richt, A. E., et al., Research Reactor Fuel Element Conference, Gatlinburg, 1962⁶⁴
- Ryu, H. J., et al., RERTR 2006 – 28th IM, Cape Town⁶⁵
- Ryu, H. J., et al., J Nucl. Mater., 2009⁶⁶
- Ryu, H. J., et al., Nucl. Eng. And Tech., 2008⁶⁷
- Van den Berghe, et al., J Nucl. Mater., 2008⁶⁸
- Welcomme, E., et al., RRFM, Vienna, 2009⁶⁹
- Wieschalla, N., et al., J Nucl. Mater., 2006⁷⁰
- Wieschalla, N., et al., RERTR 2005 - 27th IM, Boston.⁷¹

3.2.2 Comments

3.2.2.1 *Mo and Si Content*

The Si concentration in the interaction layer is important both in minimizing its growth and also in enhancing characteristics/ properties (phases, amorphous character during irradiation, ability to inhibit rapid swelling within the IL, etc.) that are important to facilitate the fuel to perform to a high burnup.

While the Mo content (within a range of 5–12 wt%) in the fuel is important to maintaining stability of the gamma phase, therefore preventing rapid swelling within the fuel and perhaps secondary effects of IL growth, it may also affect the types of phases that form in the IL, as some are stabilized by the presence of Mo. One indication of the phase mixture being affected was in an ¹²⁷I bombardment study in which UAl₂ was identified in a U-6Mo/Al fuel but not in a U-10Mo/Al fuel.^{70,71} However, in a similar study, U-7Mo/Al fuel was irradiated in a similar fashion, and no UAl₂ was detected.⁶⁹

Keiser et al.⁷² examined U-10Mo dispersion fuel as-fabricated and also after irradiation, comparing it with the examination of U-7Mo,⁷³ both from the RERTR-6 experiment. Fabricated under similar

conditions, the U-10Mo formed very little IL, showing only a few tenths of a micron layer in some areas, while the U-7Mo formed a ~ 2 μm layer. Both fuels were in a 6061 Al matrix. After irradiation the IL on the U-7Mo particles remained relatively stable, growing only slightly. There was significant IL growth on the U-10Mo particles, especially on the high fission-rate (2.9×10^{14} f/cm³s) and high fission density (3.4×10^{21} f/cm³) side of the fuel plate. The other edge of the plate, with approximately two-thirds of the fission rate and density side (and therefore heat flux), had grown to less than half the 6.8- μm thickness, on average.

The discussion focused on the effects of having a pre-formed layer rather than the effects of Mo. However, a contemporary study of pre-formed, fabrication-driven interaction layers did note that there were effects of the Mo content of the fuel.⁴⁰ The study indicated that availability of Mo to participate in the IL formation can allow some of the complex phases ($\text{UMo}_2\text{Al}_{20}$ and $\text{U}_6\text{Mo}_4\text{Al}_{43}$) to form. It would seem that fuel alloy with a higher Mo content might allow the phases with a high Mo concentration to form more easily, if the Mo concentration is high enough to ensure the gamma phase is stable. If the Mo concentration is low enough to allow the gamma U-Mo phase to transform to alpha uranium, then Mo is rejected to form U_2Mo and perhaps provide a source of Mo to the IL.

As will be discussed later in this report, the formation of these Mo-rich higher phases may play an important role in the stability, or instability, of the IL during irradiation. The effect of Si may then be synergistic with the Mo effects as Si additions to the $(\text{U},\text{Mo})(\text{Al},\text{Si})_3$ phase increase the solubility for Mo in this phase, perhaps helping to prevent the formation of the complex, undesirable phases. This Keiser et al.⁴⁰ work may help explain the effects of pre-formed layers. Previous results of the RERTR-6 experiment showed that the IL thickness was related to the Si content in the matrix alloy. Kim et al. compared the thickness of IL layers as a function of Si in the matrix Al alloy (0.2, 0.9, 2, 4.8% Si) and concluded that with at least 2% Si, there was an effect on thickness, or growth rate.⁵⁴ Keiser et al. recently made other observations concerning this experiment and noted that when a significant amount of Si was available, the IL was enriched in Si.^{50,51,52} While Si enrichment had been observed routinely in out-of-reactor experiments, it is not always seen in in-reactor experiments.

Similar observations were also made in examination of the French IRIS experiments⁵⁶ as well as in the KOMO experiments, noting that Si in the matrix did suppress IL growth. However, the researchers did observe that only when there were Si-rich precipitates near the fuel particle was the IL enriched in Si and growth was significantly reduced. The RERTR-6 pre- and post-irradiation examinations implied that perhaps the high-temperature fabrication methods used to produce RERTR-6 plates had produced a pre-irradiation IL, enriched in Si, and the Si enrichment persisted during irradiation. Recently, an addendum to this report (Appendix A) prepared by French researchers illustrates that fabrication techniques, even one hour at $\sim 450^\circ\text{C}$, form a small, irregular IL comprised of Si-rich IL phases ($\text{U}(\text{Al},\text{Si})_3$ and U_3Si_5) when a 2.1% Si matrix material is utilized. No layer is found when the matrix contains only 0.3% Si. These observations were made on plate materials as used in the IRIS 3 experiment.

These two effects are also shown in one of the Keiser studies⁵⁰. However, in this same study, Keiser et al. note that in a more aggressive test (higher fission rate), RERTR-7, the IL has very little observable Si enrichment in the areas with the highest burnup (BU) and therefore the highest fission rate. Areas with less aggressive operating conditions showed a thin IL with high Si concentration produced during fabrication.

Kim et al.⁷⁴ reflected on the same RERTR-6 results, and those of RERTR-7, and compared them to a more aggressive test, RERTR-9, where an enhanced uranium density is thought to have overwhelmed the 2% Si in the matrix, especially at high burnup. The growth of fission gas bubbles accelerates as the Si effect appears to diminish as the burnup increases. They recommended a Si concentration of $\sim 5\%$. Park et al.⁶³ also showed that even a 2% Si addition to the Al matrix in KOMO-3 resulted in a reduction in the IL thickness. However, 2% Si seemed to be insufficient to promote a $\text{U}(\text{Al},\text{Si})_3$ -type composition ratio throughout the IL.

The results of the IRIS4 experiment were recently compared to results from the IRIS3 and IRIS2 experiments to investigate the effects of matrix Si and an oxide coating on the fuel particles on fuel/matrix interaction in-reactor.⁷⁵ IRIS2 contained U-7.6 Mo fuel in a pure Al matrix, IRIS3 tested fuel (U-7.3Mo) with 2.1 wt% Si added to the matrix Al, and IRIS4 used oxidized (oxide coated) fuel (U-7.3Mo) particles in matrix material of both pure Al and Al-2.1%Si. A comparison of the swelling characteristics of the plates is shown, demonstrating that the oxide coating prevents rapid swelling (pillowing) to higher fission densities (comparing IRIS4 in pure Al to IRIS2). Comparing IRIS4 results with and without matrix Si may show a slight added benefit of the Si.

However, the authors indicate that it is not clear that these benefits are demonstrated as being additive, as IRIS3 (with Si but no oxidation) shows the greatest delay of rapid swelling. Unfortunately, the IRIS3 plate operated at a beginning heat flux less than those of IRIS2 or IRIS4, likely adding extra resistance to swelling.

Likewise, Izhutov et al.⁷⁶ recently presented results of testing in the MIR reactor (IRT-M and IRT-U FA) to investigate the effects of Si content in the matrix or coatings (oxide or ZrN) on the fuel particles. The fuel was 9.4%Mo, or U-8.8%Mo for the oxidized particles, and there was <0.3% Si in the basic matrix Al, but additions of 2, 5, and 15% Si were made to Al as variables for the matrix material. The “measure of effectiveness” for the variables was in reducing the amount of IL volume formed as compared to the base fuel design. They saw little effect of 2% Si addition to the matrix, but 5 and 15% Si both produced a 50% reduction in the volume of IL formed in their test.

This study also compared the base fuel with and without oxidized particles and showed that the oxide coating had no effect. Remember that the fuel swelling with and without oxidized fuel (for IRIS2 versus IRIS4) showed a delay in the onset of rapid swelling for oxidized fuel particles in the IRIS experiments.⁷⁵ Microstructural examinations may show the reasoning for the variation in the results of these two studies.

Recent SEM examinations have been made of the AFIP-1 experiment,⁷⁷ a test containing large dispersion U-7Mo fuel plates, one with a matrix of Al-2Si and the other with a 4043 Al (4.8% Si) matrix. The study was designed to compare interaction layers before and after irradiation. The test operated at a maximum fuel centerline temperature of ~150°C, a peak heat flux of 325 W/cm², and to an average fission density of 4×10^{21} f/cm³. The pre-formed interaction layers were thin and did not encompass the entirety of the surface of the fuel particles, especially the fuel with a Al-2%Si matrix.

After operation in the reactor, both fuels showed considerable IL formation, to the point of nearly consuming the fuel matrix, especially in the high-power regions of the U-7Mo/Al-2%Si fuel. The implication is that the Si in each was depleted eventually and the IL grew rapidly at that point. This seems to be unusual for a matrix with 4.8% Si. No chemical analysis of the irradiation-stimulated IL material was reported to confirm this.

The results of all of these tests show that Si additions to the matrix Al alloy can enhance an IL enriched in Si. The Si-enriched layer reduces the IL growth in-reactor, and this growth rate reduction reduces the probability that fission gas bubbles will grow rapidly in the IL, improving the resistance to fuel swelling. An IL pre-formed during fabrication, enriched in Si, also seems to enhance the effect of matrix Si. All of these observations seem to be consistently true if the fuel is operating at conservative operating conditions. However, at very aggressive conditions (high temperature, high fission rate, high burnup, etc.) the matrix Si may be overwhelmed, with the IL losing Si concentration and growing rapidly and the fuel swelling quickly (pillowing). The implication is that in these cases, a higher concentration of Si should be added to the matrix, and there are several observations where additions in excess of 2% Si were needed to be effective.

3.2.2.2 The Precipitate Free Zone (PFZ)

The PFZ seemed to be of more consequence in irradiation tests, especially those where the samples are active fuel. The reason was speculated that the PFZs are created more efficiently in-reactor due to the

fuel causing recoil damage in the surrounding matrix. The damage apparently allows the precipitate to dissolve more readily. Indeed, there seems to be a PFZ around the fuel in many studies (e.g., Keiser et al.⁵⁰), at least within the resolution of the characterization techniques used.

3.2.2.3 Multi-Layered IL

There are few, if any, reports of multi-layered IL formation in irradiated samples, even in ion-irradiated samples, which are more easily examined.

Phase Identification: While many phases have often been “identified” by composition data obtained using x-ray fluorescence (XRF) EDS analysis in the SEM or EPMA, the most convincing data were obtained using XRD, μ XRD or neutron diffraction analysis (NDA) because actual crystal structure information is obtained using the latter techniques. Analyses involving only chemical composition are unreliable because a number of phases could be analyzed simultaneously using any of the techniques, and only the diffraction techniques may identify each of them. Only TEM uses diffraction (electron) and fluorescence (XRF) to examine individual phases unambiguously. The following discussion, and the spreadsheet/database acknowledge positive identification only through TEM with electron diffraction, XRD, μ XRD, or NDA. The database notes composition-based identification separately within “note” or “comment” fields.

Richt et al.⁶⁴ had identified very early that Si additions encouraged the formation of UAl_3 phase as opposed to UAl_4 , and noted that the UAl_3 became amorphous during irradiation. Richt was working with U-Al-Si alloys to be used as fuel. The compositions used were such that excess Al would be expected if the dominant phase were UAl_3 , but the UAl_3 remained stable as it was the only crystalline phase found after irradiation; UAl_4 was not found using XRD, but an etching technique indicated that perhaps the composition was present but the phase was amorphous. Interpreting these observations, the authors were apparently implying that perhaps the UAl_3 composition did drift towards UAl_4 , as excess Al was absorbed, and the UAl_4 formed was always amorphous.

As new fuel studies began using U-Mo alloys, there was an early identification of IL phase as (U, Mo) Al_7 based on some other early diffusion couple testing.⁷⁸ Another early paper describing characteristics of IL layers produced in-reactor found several layers, each with a different composition: one composition near (U,Mo) Al_3 and the other near (U,Mo) $Al_{4.4}$.⁵⁵ Note that these early studies contained no real phase identification.

Conlon and Sears⁴¹ showed that U-10Mo dispersed in Al formed an IL when irradiated and, using NDA, identified the major phase in the IL as (U,Mo) Al_3 , and essentially no “higher” aluminides, like UMo_2Al_{20} or $U_6Mo_4Al_{43}$, often observed in diffusion couple tests. The fuel burnup was 20 at.% ^{235}U . In a later study, Conlon and Sears⁴² showed that the results remained the same to higher burnup (~60 at.% ^{235}U) because the major phase was (U,Mo) Al_3 . However, this time much more UAl_2 and a small amount of UMo_2Al_{20} were also found, but not enough to account for the excess Al in the IL if the UAl_2 that formed at high burnup was a decomposition of UAl_3 . Only 4% of UAl_4 was detected, but of course if there was an amorphous phase of that composition it would be difficult to detect.

Golosov et al.⁴⁶ used NDA to examine U-9 wt%Mo/Al fuel following “equivalent” burnups of 33 to 97%. After irradiation, the IL was likely (U,Mo) Al_x and was amorphous (only 0.5% crystalline UAl_3 at all burnup levels). Upon annealing after irradiation at 50°C to 550°C (1 hr at each 50-°C step), it was found that at temperatures of 350°C and above, the fraction of UAl_3 increased to about one-third of the IL phases at 550°C, but at 550°C the IL was also growing in size from the thermal treatment. In a subsequent analysis,⁴⁷ small-angle neutron scattering showed Guinier-Preston zones and small particles of UAl_3 , indicating that the “amorphous” label in many other studies may be related to the characterization technique, where the amorphous character is related to size and orientation specificity of the particles being examined.

Park et al.^{63,79} did not make complete phase identifications, but did chart where and under what conditions the (Al+Si)/(U+Mo) was ~3, 4, or higher. They concluded that the ratio was low during high-temperature irradiation, low if the IL was formed prior to irradiation, and higher if the irradiation temperature was low and there was no pre-formed IL.

TEM can provide both chemical and structural information from very small and specific areas. The difficulty is preparation of a TEM sample from the very reactive and radioactive irradiated metallic fuel. Gan⁴³ and Van den Berghe⁶⁸ both were able to produce such specimens and provide some very important results. The work of Gan et al. showed that the IL in a U-7 wt%Mo/Al-2%Si dispersion fuel was apparently amorphous with composition data showing the “Al+Si” content was very high (80–90 at.%).

The Gan et al. study used samples from the fuel experiment RERTR-6. It is known that an IL formed during fabrication of the fuel used in this experiment, and it is suspected that at least under some irradiation conditions, the pre-formed IL can affect the nature of the phases and composition of the IL subsequently in-reactor. It would be instructive to add the fabrication process steps to the spreadsheet representing irradiated fuels. However, this level of detail is not often revealed in the published work.

The issue of the effects of a pre-formed IL will be revisited in a subsequent section of this article (see “Effect of Temperature”), as others have noticed this effect as well.

Ion Irradiation – IL Phases Produced

The use of particle irradiation to simulate the combined effects of fissioning and a neutron flux has been used to study the formation of an IL in an irradiation environment. Birtcher and Baldo⁸⁰ irradiated U-6Mo (coated with Al) at 150°C using 3-MeV Kr ions. Based upon the elementary composition of the IL that formed, UAl₃ was the suspected phase. Despite the fact that there was no Si, there was no mention of a suspected presence of UAl₄.

Palancher et al. also irradiated fuel in the laboratory, using 80-MeV ¹²⁷I ions.⁶² They used U-7Mo and U-10Mo dispersion fuels in pure Al and in Al alloyed with Si. The irradiation was performed at 170°C or less. The environment allowed an interaction layer to grow on the fuel particles, more slowly in the samples which had Si in the matrix. Micro-XRD revealed only the UAl₃-type phase. Of course one could say that if an amorphous phase, like UAl₄, was also present, the μ XRD may not have revealed its presence. A more recent study by most of these same experimenters⁶¹ used the U-7Mo and U-10Mo in pure Al, irradiated with 80-MeV ¹²⁷I ions at less than 170°C, and performed a much more detailed XRD and μ XRD analysis and obtained the same results. The UAl₃ phase was the only IL phase identified.

A previous study by Wieschalla et al., involving some of the same researchers,⁷⁰ was performed using U-6Mo and U-10Mo in Al, irradiating with 120-MeV ¹²⁷I ions at a maximum temperature of 200°C but to about half of the dose of the latter study, yielded different results. Almost equal quantities of UAl₂, UAl₃ and UAl₄ were found in the U-10Mo sample, and nearly equal quantities of UAl₃ and UAl₄ were found in the U-6Mo sample. Another test was run at 80 MeV with ¹²⁷I ions and at a maximum temperature of 100°C. The sample broke, limiting the characterization that could be done; there was evidence of an amorphous phase, but its existence was not proven.

These ion irradiations may provide information about the conditions that create amorphous IL phases and their compositions. Taking these results together, one might surmise that the lower temperature test produced an amorphous-phase IL, the higher temperature test (<170°C) produced UAl₃ in crystalline form, but perhaps other phases also in amorphous form. The highest temperature test (<200°C) produced all three crystalline phases, as an amorphous phase could not be supported at that temperature. The study by Miller et al.⁶⁰ irradiating with energetic ions and protons and discussed in more detail in the next section of this article, would support the last observation because crystalline phases were not made amorphous by 200°C proton irradiation. A follow-up study by Gan et al.,⁸¹ discussed in more detail in the next section, also produced consistent results.

Just as in-reactor fuel experiments have been used to investigate the effects of matrix silicon on IL formation, Jungwirth et al.⁸² looked at the potential for optimizing the Si content in the matrix material. They formed fabrication-related IL in dispersion fuels with U-7Mo fuel in Al matrix material containing 2, 5 and 7 wt% Si. Although the times at temperature for the fabrication procedures were not given, it was observed that the fuels where the matrix contained 5 and 7 wt% Si had formed a Si-rich IL. The fuel was then ion irradiated (80-MeV ¹²⁷I) at 150–300°C to ~1000 dpa to observe the effect of irradiation on the IL in each sample. It was observed that the thin Si-rich IL had either been completely replaced by an IL depleted in Si, or partially replaced, on the Al matrix side of the IL. This result is consistent with the fuel plate examinations where operating conditions were aggressive or the burnup had exceeded a point where the Si in the matrix proved insufficient to maintain a Si-rich IL.^{74,76,77}

3.2.2.4 Effect of Irradiation on IL

Irradiation created from neutrons and fission recoil, knock-on damage in particular, creates effects that can be important to interaction layer formation and properties. For example, damage-induced point defects can accelerate diffusion, producing conditions similar to high temperature where vacancy concentrations are large. In addition, the damage can produce instability in some phases, helping to dissolve precipitates or decrease order in crystalline phases, particularly intermetallic phases. The latter effect can cause a crystalline phase to become completely amorphous.

Ryu et al.⁶⁶ chose to examine the differences between observations made of testing that did not involve irradiation and those that did. Rather than concluding that different phases are present because irradiation can shift equilibrium temperatures lower, much of the difference is explained by allowing the irradiation to create amorphous phases, undetectable by standard diffraction techniques.

Heavy particle irradiation (neutron, ion, etc.) is known to also enhance diffusion, the process by which the interaction layer is created. An analysis of sample cross-sections from ¹²⁷I ion-irradiated U-7Mo in Al, performed in a manner nearly identical to the studies mentioned previously, showed clearly the effects of irradiation in stimulating the growth of the IL.⁶⁹ In this study, ion fluence, flux, and incidence angle were varied to observe the influence of these variables. The increase in IL thickness with fluence was clearly demonstrated.

Kim et al.⁸³ produced a model based upon thermal diffusion and enhanced by irradiation, using fission rate to correlate with the experiment or to provide an estimate of future fuel performance.

Some of the more important observations concerning the difference between thermally activated IL formation and that formed in-reactor were by Park et al.,^{63,79} who recently described the results of the KOMO-3 irradiation test using dispersion and monolithic rod-type fuels. The dispersion fuels were constructed of U-7Mo or U-7Mo-1Zr in Al or U-7Mo in Al-2%Si. Characterization of the fuel with the Si-bearing matrix showed a difference between the irradiation test and previous ex-reactor testing in that the Si-rich region was near the matrix instead of the fuel particle. Also, Si accumulation in the IL was not as prominent in the irradiation test. As part of the presentation of this work, the authors showed that the low-temperature irradiations were sensitive to whether an IL had been initiated during the fabrication process.⁷⁹ While the causes of these observed effects cannot be completely explained, they are likely to be important in developing such an understanding.

Miller et al. studied U, Mo, Si, and Al alloys that were cast to produce (U,Mo)(Al,Si)₃, UAl₄, UMo₂Al₂₀, and U₆Mo₂₀Al₄₃ compounds.⁶⁰ They were then irradiated with protons at 200°C to 3 dpa. The irradiation was thought to perhaps induce some of the phases to become amorphous. However, with the exception of some precipitate coarsening in the UAl₄, the alloys were all very stable. As previously mentioned, the temperature may have been too high for the irradiation to cause the alloy phases to become amorphous.

In a follow-up study, Gan et al. looked at the alloys using TEM after bombarding the samples with 500-keV Kr ions to 10 and 100 dpa at 200°C.⁸¹ They had identified the phases UAl₄, U(Si,Al)₃,

(U,Mo)(Si,Al)₃, UMo₂Al₂₀, and U₆Mo₄Al₄₃ in the as-cast alloys. This study provided a more detailed evaluation of the conditions under which these phases became amorphous, although only one temperature was used. They examined the materials at low doses to 10 dpa and then 100 dpa and noted whether the crystalline phases had become amorphous and which had swollen (formed voids). A connection between void and gas bubble formation is not necessarily implied.

U(Si,Al)₃ never became amorphous (at 200°C) while the similar phase in the quaternary system, (U,Mo)(Si,Al)₃, was amorphous at ~2 dpa. Neither phase had formed voids by 100 dpa. The UAl₄ also never became amorphous at 200°C and formed no voids to 100 dpa. The more complex phases, UMo₂Al₂₀ and U₆Mo₄Al₄₃, became amorphous at less than 10 dpa and less than 1 dpa, respectively. The UMo₂Al₂₀ formed no voids to 100 dpa while the U₆Mo₄Al₄₃ showed no voids at 10 dpa but had formed voids and/or large gas bubbles by 100 dpa. Again, 200°C is perhaps too hot to sustain amorphous phase in some of the other intermetallics. The low stability and void-forming characteristics of the U₆Mo₄Al₄₃ make it suspect for being related to gas bubble formation at low fission density in operating fuel.

The combined results of these ion irradiation studies and the observations give rise to a set of observations that provide a potential theory for the combined effects of Mo and Si on fuel stability. Much of this is outlined in a recent publication by Perez et al.³⁹ The suggestion that the presence of the U₆Mo₄Al₄₃ phase is necessary to prevent early breakaway swelling of the fuel requires an understanding of what conditions encourage its formation; they can then be avoided. The high Mo content would suggest that minimizing the local concentrations of Mo would be important. So, use as little Mo in the fuel as possible without allowing U-Mo instability (transformation to alpha phase); the transformation would force rejection of Mo to the IL. Si enhances the solubility of Mo in the (U,Mo)(Si,Al)₃ phase, allowing for the presence of more Mo without deleterious effects.

This theory/model ties together many observations. These include:

1. Si in the Al matrix improves fuel performance
2. Formation of a Si-rich IL prior to irradiation enhances performance (inhibits rapid fuel swelling)
3. An IL where (Si+Al)/(U+Mo) is ~3 is most stable
4. High-temperature irradiations are less affected by Si additions.

A synergism such as this between Mo and Si may explain many of the differences in the results of experiments where Mo and Si are allowed to vary independently. It also may explain the reason large amounts of IL are sometimes seen without gross fuel swelling as in the AFIP-1 test⁷⁷—large volumes of IL can form if they consist of stable phases. Note also that AFIP-1 fuel had thin as-fabricated IL very low in Mo and high in Si.

3.2.2.5 Effect of Temperature

Equally important observations have been made concerning some of the experiments led by a number of French researchers. There are many references for the detailed examinations of the three “IRIS” experiments and the “FUTURE” experiment, but Dubois et al. [84] conducted a thorough review of these experiments including observations made concerning the IRIS1 and IRIS2 experiments. The basic findings were:

- The IL was thicker in the IRIS2 experiment, at a lower accumulated fission density, but accumulated at a higher fission rate, heat flux, and operating temperature.
- The Al/(U+Mo) ratio was 6–8 for IRIS1, 4.6–5.8 for IRIS 2, and the authors compared these to the FUTURE experiment ratio of 3.3–4.7 and the UMUS experiment ratio of ~3. The latter experiments operated at higher temperatures, with UMUS fuel operating at ~225°C.
- The IRIS1 and IRIS2 ILs were amorphous, although neither operated hot enough to clearly have allowed crystalline phases, and none were found.

Ryu et al. recently extended these observations to include another experiment.⁶⁶ The authors examined the data for the IRIS-1, KM004, IRIS-2, and FUTURE experiments (in order of increasing heat flux and operating fuel temperature, 140 to 340 W/cm² and 75 to 130°C, respectively). Data on the IL composition from each experiment were gathered from Leenaers, et al., JNM, 2004; Golosov, et al, RRFM 2007; and Huet, et al., RRFM 2005. For those four experiments, a consistent trend exists in the Al/(U+Mo) ratio, decreasing from 6–7 to 3.3–4.7 as the operating temperature is increased. PIE results indicate that low temperatures and high fission rates produce conditions that are conducive to amorphization of the IL phases.

Likewise, Park et al.⁶³ showed that KOMO-3 IL compositions differed between those formed in cooler (120–150°C) regions and those in hotter (160–200°C) regions, the (Al+Si)/(U+Mo) being closer to 3 in the hotter region and 4 or higher in the cooler region. This was true with or without Si in the matrix. As stated above, comparing theirs with the work of others (Keiser, RERTR-6), Park et al. showed that low-temperature irradiations were sensitive to whether an IL had been introduced during fabrication, with the reduction of IL growth more prominent if a pre-formed IL is present prior to irradiation. Si enrichment in the IL, high in the pre-formed layer, reduces with irradiation and is inconsistent if there is no pre-formed IL, as in KOMO-3. At high temperatures, the Si enrichment was always present.

Keiser et al.⁵² recently showed even more convincing data concerning the importance of pre-irradiation IL layers, especially that those interaction layers enriched in Si are particularly stable. Subsequently,⁴⁰ characterization of U-7Mo/4043 Al or U-7Mo/Al-2Si plates showed that pre-treatment IL can produce thin layers rich in Si, or thicker Si-depleted layers. They suggest that the phases formed in the thin layers (binary U-Si phases and U(Al, Si)₃) may be eventually beneficial to irradiation performance.

Many recent publications address the correlation between the composition and thickness of ILs formed prior to irradiation and the IL growth rate and composition formed during subsequent irradiation. Also, many of Keiser et al. and Park et al.'s observations have been considered in more detail.

4. LITERATURE REVIEW – CONCLUDING REMARKS

The differences between studies where the IL is created in the absence of irradiation and those where irradiation was included are significant. They indicate that the results from one kind of study cannot be used specifically to explain the processes determining structure and composition of the IL produced in the other.

However, the importance of the testing results where no radiation was used may result in a prescription for optimization of the pre-treatment of fuels. A small, Si-enriched IL, formed during a step in the fabrication process, seems to be stable during irradiation, helping to prevent the rapid growth of an irradiation-induced IL. Moreover, the Si-enriched IL does seem to be important to delaying the onset of rapid growth of fission gas bubbles. Therefore, prior testing where dispersion fuels are heat-treated or diffusion couples are tested may be important in optimizing the pre-irradiation IL layer formation.

Conclusions from irradiated fuels data have been repeated often in the literature review. However, as related to the “Desired Characteristics of the IL” mentioned at the beginning of this article, several concluding remarks can be made:

1. At low temperatures, and especially in an irradiation atmosphere, the (Al+Si)/(U+Mo) ratio of three is difficult to produce, but phases related to UAl_3 are preferred because they seemed to be related to good fuel performance. In the cases where fuel operating temperatures are low, it is important to create a Si-enriched, pre-irradiation IL. The IL is relatively stable, performs well in terms of swelling resistance, and prevents rapid IL growth during irradiation. Fabrication-related heat treatments should not be excessive in order to maintain a thin, Si-enriched layer containing potentially beneficial phases.
2. At higher operating temperatures ($>150\text{--}170^\circ\text{C}$), the IL formation in-reactor may not be so dependent on pre-irradiation IL formation, especially at high burnup because the pre-fabricated IL seems to be less stable at high burnup and high operating temperature. Moreover, the (Al+Si)/(U+Mo) ratio of three occurs more often at higher temperature. For these two reasons, it is important at high operating temperature to also have a matrix with significant Si content, which is important for creating a thin IL in-reactor with the right characteristics.
3. Out-of-reactor testing seems to indicate that Si in the matrix material is required in some concentration (2%, 5%, ?) to provide for a thin, Si-enriched IL formed before irradiation of a fuel plate. It ensures the IL contains beneficial phases or prevents formation of some phases known to promote poor fuel performance. Significant progress has been made in understanding what characteristics this IL should have.
4. The use of a fuel with stable gamma phase appears to allow more predictable performance, regarding both the reliable forming of a beneficial pre-irradiation layer and the fuel performance (low swelling) to high burnup. Destabilization of the gamma phase may create problems with IL breakaway growth.
5. One theory does seemingly explain the roles of Mo and Si as a synergistic effect. It is based on the notion that certain phases in the IL, especially $\text{U}_6\text{Mo}_4\text{Al}_{43}$, create poor fuel performance and manifest as breakaway fuel swelling. One could then conclude that Mo in the IL creates the problem. The role of Si is to increase the solubility of Mo in the $(\text{U},\text{Mo})(\text{Al},\text{Si})_3$ phase, and an IL dominated by this phase is more stable. Simply, Mo creates the IL instability (with regard to swelling, not growth), and Si helps to mitigate its effect. Si does also seem to inhibit IL growth rates; it is not clear if these are related. It has been shown that thick layers can form during irradiation without attendant high swelling.
6. More work is needed in order to prescribe the conditions to best produce a beneficial IL. Another necessity is a better understanding of any correlation between beneficial characteristics of the pre-fabricated IL and the irradiation conditions to which it will be subjected. The theory expressed in (5) above may be prove to be such an understanding.

5. DATABASE/SPREADSHEETS

Table 1 shows several examples of entries into the “Irradiation” part of the database/spreadsheet.

Table 2 shows another two entries but from the spreadsheet containing information on ILs created without an irradiation atmosphere. These are just examples of the type of information stored from the references mentioned earlier.

Table 1 (a, b). Example(two entries) from Interaction Layer Database/Spreadsheet, Irradiated Fuel.

(a)

| Author | Reference | Fuel | Al Alloy | Peak Temp. C | Enrichment % | U density g/cm ³ | Ion Fluence cm ⁻² | Avg. Fission Density f/cm ³ | BU at.% ²³⁵ U | Measurement Techniques |
|------------------|------------------|--------|----------|------------------|--------------|-----------------------------|------------------------------|--|--------------------------|------------------------------|
| | | Mo wt% | Si wt% | | | | | | | |
| J. Gan, et al. | RRFM 2009 Vienna | 7 | 2 | 109 | 19.4 | 6.2 | | 4.5×10^{21} | 45.8 | OM, SEM, TEM, EDS |
| Leenaers, et al. | JNM 335 p39 2004 | 7 | 0 | 130 (clad temp.) | | 8.47 | | 1.41×10^{21} | 32.8 | OM, SEM, EDS, WDS, EPMA, XRD |

(b)

| Author | Reference | Composition at % | | | | Composition Ratios | | | Phases | Comments |
|---------------------|------------------|------------------|-----|------|-----|--------------------|-------|----------------|-----------|--|
| | | U | Mo | Al | Si | Al+Si/ U+Mo | Al/Si | Al+Si+ Mo/U | | |
| J. Gan, et al. | RRFM 2009 Vienna | 10.3 | 4.3 | 76 | 9.3 | 5.8 | 8.2 | 8.7 | Amorphous | TEM; Spots A1,A2,A3 |
| | | 14.8 | 4.5 | 71.1 | 9.5 | 4.2 | 7.5 | 5.8 | Amorphous | TEM; Spots A,B,C |
| A. Leenaers, et al. | JNM 335 p39 2004 | 18.3 | 4.2 | 77.5 | | 3.4 | | 4.5 | | FUTURE, see Xe distribution; presumed IL to be UAl ₃ ; Ryu L1 |
| | | 15 | 3.2 | 81.9 | | 4.5 | | 5.7 | | FUTURE, see Xe distribution; presumed IL to be UAl ₄ ; Ryu L2 |

Table 2 (a,b). Example(two entries) from Interaction Layer Database/Spreadsheet, Unirradiated Samples.

(a)

| Author | Reference | Method of Producing Interaction | Anneal | | Mo in Fuel, wt% | Si in Matrix, wt% | Method of Analysis | Interaction Layer Composition, at. % | | | |
|---------------------|--|---------------------------------|--------|------------|-----------------|-------------------|-------------------------|--------------------------------------|----|--------|--------|
| | | | T, °C | Time, hrs. | | | | U | Mo | Al | Si |
| J. M. Park, et al. | JNM 374 p422 2008, see also Cape Town 2006 and Boston 2005 | Mechanically clamped | 580 | 5 | 7 | 0 | OM, SEM, EPMA, XRD | 17 | 2 | 81 | |
| | | | | | 7 | 2 | | 18 | 2 | 68 | 12 |
| | | | | | 7 | 5 | | 18 | 2 | 50 | 30 |
| | | | 600 | 3 | 7 | 0 | | 16 | 2 | 82 | |
| | | | | | 7 | 2 | | | | | |
| | | | | | 7 | 5 | | 18 | 2 | varies | varies |
| | | | | | | | | | | | |
| M. Mirandou, et al. | Boston 2005 | Friction stir welded | 340 | 1176 | 7 | 7.1 | OM, SEM, EDS, EPMA, XRD | 32 | 4 | 8 | 56 |
| | | | 340 | 552 | 7 | 7.1 | | 31 | 5 | 6 | 58 |
| | | | 340 | 1248 | 7 | 0.6 | | 36 | 5 | 9 | 50 |

(b)

| Author | Reference | Composition Ratios | | | Thickness, μm | Phases | Comments |
|---------------------|--|--------------------|--------|--------------|--------------------------|--------|--|
| | | (Al+Si)/(U+Mo) | Al/Si | (Al+Si+Mo)/U | | | |
| J. M. Park, et al. | JNM 374 p422 2008, see also Cape Town 2006 and Boston 2005 | 4.3 | | 4.9 | 135 | | App. avg. comps. EPMA |
| | | 4 | 5.7 | 4.6 | 25 | | App. avg. comps. EPMA, Si varies (high by Al; low by U) |
| | | 4 | 1.7 | 4.6 | 35 | | App. avg. comps. EPMA, Si drops near U and Al |
| | | 4.6 | | 5.3 | 240 | | App. avg. comps. EPMA |
| | | | | | 65 | | |
| | | 4 | varies | 4.6 | 63 | | App. avg. comps. EPMA, Al + Si remains constant at ~ 80% |
| M. Mirandou, et al. | Boston 2005 | 1.8 | 0.1 | 2.1 | 21 | | Al A356 |
| | | 1.8 | 0.1 | 2.2 | 5 | | Al A356 |
| | | 1.4 | 0.2 | 1.8 | 2 | | Al 6061 |

6. ANALYSIS OF IL DATA

The data set seems large enough to analyze trending with variables such as time, temperature, matrix alloy, etc. However, in practice, the abundance of variables makes it difficult to capture a group large enough to analyze without having to assume we can ignore some of the parameters. Despite this problem, one of the goals of this investigation was to analyze the data gathered in the spreadsheets to identify trends in the data that had not been observed previously. The following presents some of this analysis.

One conclusion reached by a number of the researchers whose work appears in the database is that the $(Al+Si)/(U+Mo)$ ratio for the IL formed in irradiated dispersion fuels is influenced by temperature and Si content of the matrix. The ratio is thought to be important in obtaining an IL with optimum properties, and the ratio is thought to be higher if the irradiation temperature is lower. The higher ratio is not desired.

Figure 1 shows the $(Al+Si)/(U+Mo)$ ratio plotted against the Si content in the matrix Al alloy. No trend is seen, although the number of data points does not allow a statistical review of the correlation. More importantly, the temperatures are also shown. Note that the data from the FUTURE experiment shows the “maximum cladding temperature” (fuel temperatures were not found in the available literature), indicating that the fuel temperatures were in the higher range in this data set. The first impression is that the Si content of the matrix does not influence the $(Al+Si)/(U+Mo)$ ratio, but note that the only data point with a Si concentration greater than 2 wt% is also the one at the lowest operating temperature. This is a good example of how the great number of variables impedes trending.

The data indicate some trend in the higher temperature experiments showing a lower ratio, but the lower temperature experiments demonstrate a fairly wide range. Perhaps the presence and nature of a pre-formed IL influences this data. Also, the way the temperatures were calculated, etc., is unknown, so these comparisons are difficult; conclusions should not be made without further review of analysis methods. These results do show that the several observations within a single experiment, with a single variable like KOMO-3, were analyzed, and the trend can be stated, but it is difficult to compare to other experiments because of the lack of detail shown in the literature.

Note that fuel burnup or fission density, fission rate, etc., are not used as variables here. Again, the number of variables makes the scope of the data set unusable to trend the data, unless some of the variables can be considered as constant (ineffective).

Another characteristic of the IL thought to be important to optimizing fuel performance is the enrichment of the IL with Si. Because it seems difficult to enrich the IL in Si consistently in-reactor, a pre-formed Si-enriched IL is thought to be beneficial. Therefore, the available data concerning out-of-reactor ILs produced by thermally activated diffusion may help reveal what conditions might produce the best pre-formed IL.

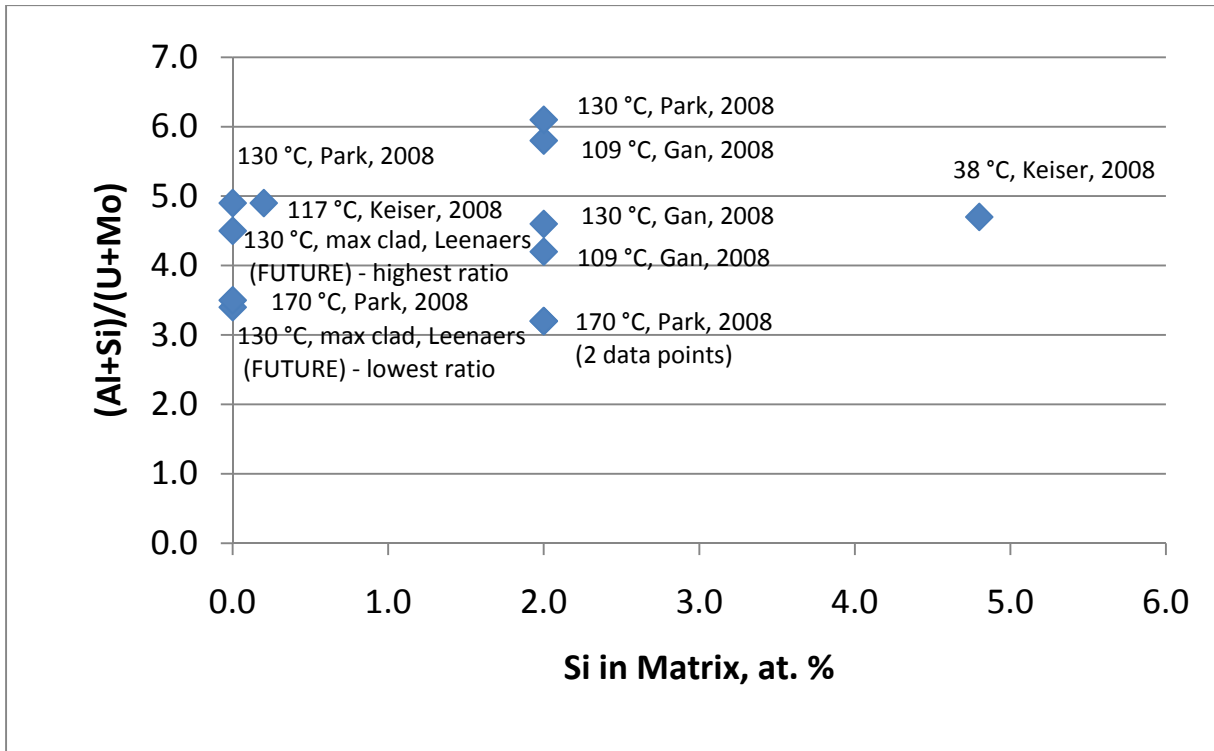


Figure 1. Data extracted from database showing (Al+Si)/(U+Mo) ratio as a function of Si concentration in matrix material. Reported temperatures are shown.

Figure 2 shows the Si/Al ratio of interaction layers formed without irradiation. These data all represent ILs that were noted to be layered, and the composition was measured both in the layer closest to the fuel and in that closest to the unaltered Al alloy (matrix). The only obvious correlation is that Si-rich layers can be formed near the fuel if the Al alloy contains sufficient Si (> 4 wt%). A single point at high temperature (600°C) does not fit the observation. Perhaps there is a temperature limitation. Again, the size of the database and data scatter make correlations difficult. Note that Mo content in the fuel is discounted in this analysis, as is time at temperature or IL thickness; too many variables, not enough data.

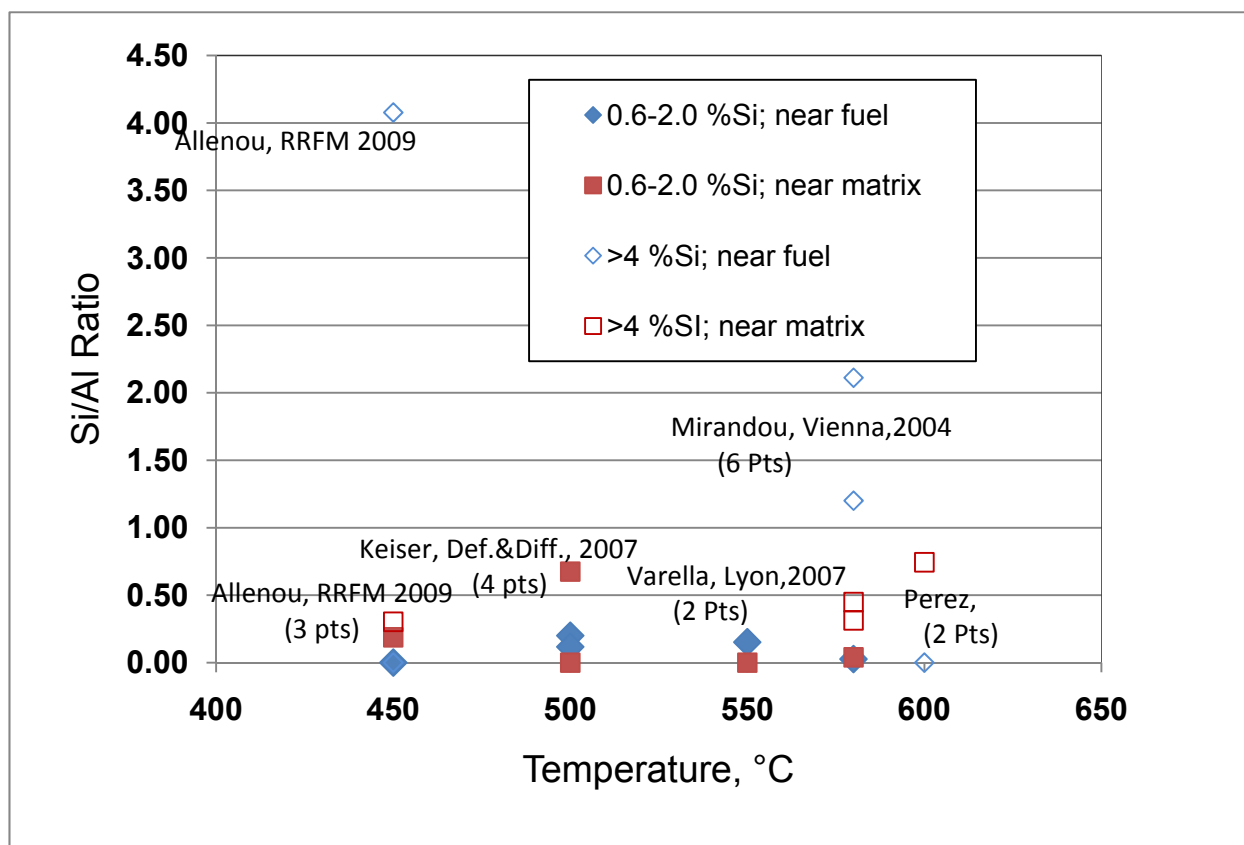


Figure 2. Interaction layer Si/Al ratio as a function of formation temperature. Shown are data where the IL was layered, so “near fuel” and “near matrix” data are both shown. Data is segregated by Si composition in the Al alloy “matrix” material.

Figure 3 was generated to add to the size of the data set. In this case all of the data shown in Figure 2 are plotted again, but additional data was added—data where only an averaged IL composition was measured or where there was no obvious layering within the IL (8 points total); all Si concentrations in the Al alloy are shown using the same marker. However, all Al alloys contained some Si. The lack of strong correlation is not surprising considering the large differences observed after minor changes in pre-irradiation heat treatment.⁴⁰ The relative instability of U-7Mo has not been sorted out of this data, nor has time-at-temperature been included as might have been suggested by the results of that study, although the database would then be too small to make a reliable conclusion.

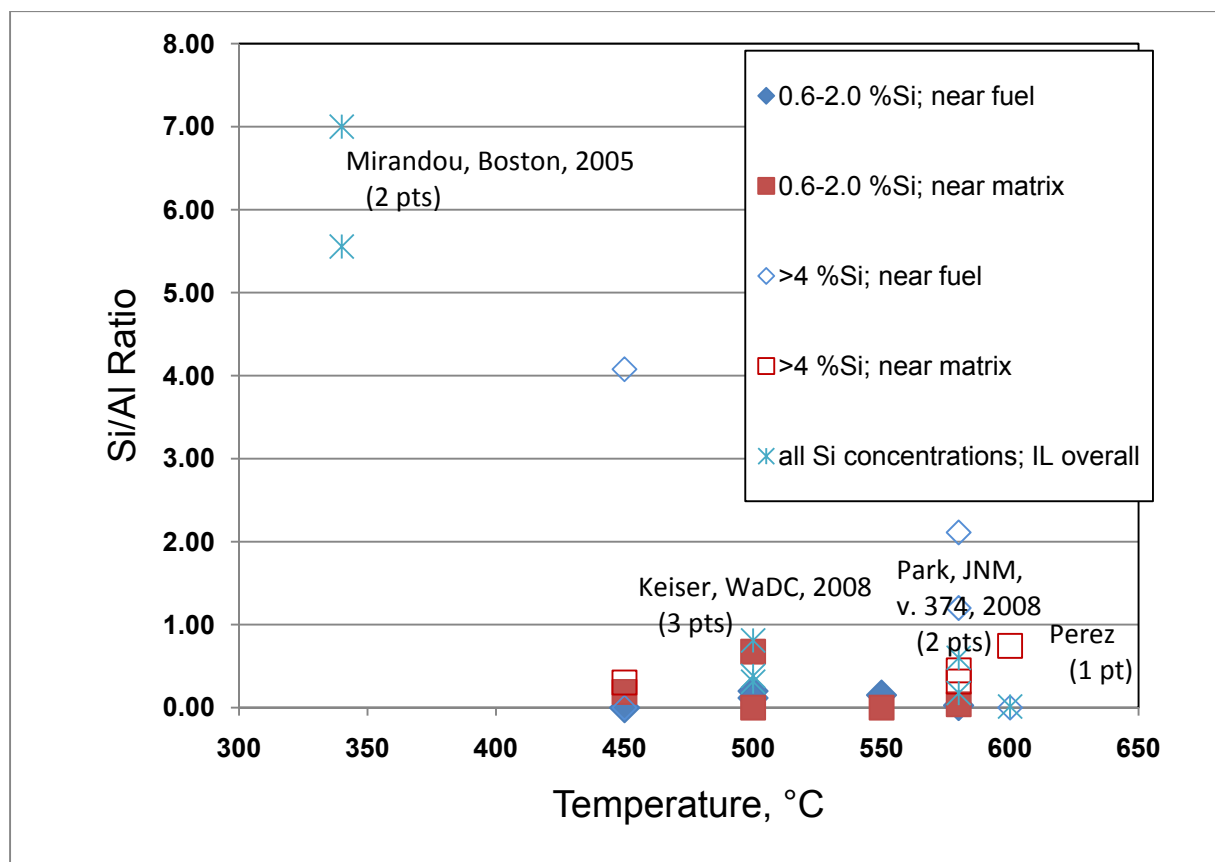


Figure 3. Interaction layer Si/Al data, along with “average” or non-layered IL data (“overall”).

There was some work, such as Mazaudier et al.¹⁵ using Al alloys with very low Si (0.25 and 0.3 %Si)– this data is not shown here.

Most of the earlier discussions of individual experiments noted some trends in the data associated with the specific experiment(s), despite the fact that the data sets were very small. These were small data sets where the variable parameters were relatively limited. We attempted to expand the data set to see if the trends remained. However, there were more variables associated with this data set making this difficult.

Trends discussed earlier have not been discounted, nor significantly supported. The larger data set can however, be used to define further experiments that could: (1) Assist in optimization of pre-irradiation heat treatments, (2) provide a side-by-side comparison of optimized U-7to8Mo and U-10to12Mo dispersion fuels to discover under what conditions a more stable fuel particle may be required, and (3) optimize the Si content of matrix material.

7. CONCLUSIONS

This work summarizes previously reported information on the IL that forms by interdiffusion between U-Mo alloy fuel particles and the Al-based matrix materials in dispersion fuels, or between U-Mo alloy foils and Al-based cladding/liner materials in monolithic fuel designs. The goal was to gather data concerning the composition of the IL (compositions and phases) as they formed in-reactor in RERTR test fuels, in simulated irradiation environments (heavy ion, etc.), and in the laboratory (time and temperature only). A database was to be generated in an effort to better understand the factors that caused the different characteristics of the IL that formed and how those characteristics affected irradiation performance.

In doing so, the discussions by the various researchers were also reviewed and similarities were noted. Fuel test data indicate that for optimum fuel performance the IL that forms should be thin and enriched in Si, especially near the fuel surface. It should also be composed of a (U,Mo)(Al,Si)₃ phase and should not contain secondary complex phases. Where the fuel operating temperature is low, it is important to create a pre-irradiation IL, enriched in Si. A matrix/cladding Al alloy containing at least 5% Si is advised to ensure an enriched IL. The pre-formed IL is relatively stable, performs well in terms of swelling resistance, and prevents rapid IL growth during irradiation. Fabrication-related heat treatments should not be excessive in order to maintain a thin layer, enriched in Si, and containing potentially beneficial phases.

A pre-formed IL is not as important if the fuel operates at higher temperatures (>150–170°C), especially at high burnup; a pre-fabricated IL seems to be less stable at high burnup and high operating temperature. Moreover, the (Al+Si)/(U+Mo) ratio of three occurs more often at higher temperature. For these two reasons, it is important at high operating temperature to also have a matrix with significant Si content to create an IL with the right characteristics in-reactor.

U-(10-12)Mo is more stable than alloys of lesser Mo concentration, and destabilization of the gamma phase has been observed to apparently lead to break-away IL growth. However, more Mo may promote unwanted, unstable phases in the IL, such as U₆Mo₄Al₄₃. If, however, the reduced Mo results in a destabilized fuel and it transforms to alpha phase, Mo will be rejected, promoting the unwanted phases in the IL. Si increases the solubility for Mo in the (U,Mo)(Al,Si)₃ phase, discouraging the formation of U₆Mo₄Al₄₃.

The database/spreadsheet was used to analyze the effects of the many variables on the characteristics of the IL formed. The results indicate that the IL formed in the laboratory is often very different than that formed in-reactor. The laboratory data is most relevant in understanding the formation of pre-formed IL layers that are important in low-temperature irradiations. The analysis also shows that the current information may be too diverse (too many variables), restricting comparison between the results of different researchers. This indicates that a structured experiment is needed to draw correlations between the various bodies of research.

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Appendix A

**Characterization of IRIS3 As-fabricated Plates
(Before Irradiation)**

Both analyses are in excellent agreement showing, the second enabling however to isolate fully the SiRDL from the particle core. Analysis using the Rietveld method shows that this SiRDL consist of:

- U(Al,Si)_3 ($a_0=4.16$ that is to say 44%Si assuming U(Al,Si)_3 follows a Vegard's law⁸⁵)
- U_3Si_5 or USi_{2-x} (P6/mmm, $a=b=3.95 \text{ \AA}$ $c=4.013 \text{ \AA}$) on the one hand and what we propose to be stoichiometric USi_2 (P6/mmm, $a=b=4.028 \text{ \AA}$ $c=3.89 \text{ \AA}$).