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OPTIMIZATION OF THE SI CONTENT IN UMO/AL(SI) FUEL PLATES

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ABSTRACT

Silicon seems to limit the formation of the undesired diffusion layer forming during in-pile irradiation of UMo/Al dispersion test fuel plates. It has been found that adding Si to the Al matrix of this fuel results in the formation of a Si-rich diffusion layer (SiRDL) covering the UMo particles. It is therefore of great interest to find the influence of the Si content on the features of this SiRDL (thickness, homogeneity composition).
Three different UMo/Al(Si) miniplates with an Al matrix containing 2wt% Si, 5wt% and 7wt% Si as well as a UMo/Al reference have been produced by AREVA-CERCA. These samples have been characterized after production and after subsequent heavy ion irradiation by XRD and SEM/EDX.
With 5 and 7wt% all UMo particles are protected with a SiRDL. However after heavy irradiation at high fluency, this SiRDL may be destroyed giving rise to the growth of the UMo/Al interdiffusion layer as it is observed in irradiated samples with no Si content. These results are in excellent agreement with those obtained on the in-pile tests RERTR-7.

1. Introduction

UMo-Al dispersion fuel is regarded as a promising candidate for a new high density fuel for research reactors. However, it has been found that during in-pile irradiation of UMo-Al dispersion fuel plates are affected by pillowing and and eventual break-away swelling. The main reason for this undesired behavior seems to be the formation of an interdiffusion layer (IDL) at the interface between the UMo particles and the surrounding Aluminum matrix. To avoid the formation of this IDL different means have been proposed [1, 2, 3, 4, 5].

It has also been shown that with swift heavy ions (e.g. Iodine at 80MeV) the effects of months of in-pile irradiation can be emulated within some hours without activation of the samples such allowing a quick and easy screening of many different kinds of samples [6, 7, 8].

Consequently, to define the most promising candidate for a future in-pile test, 20 different miniplates have been produced by TUM (Technische Universität München) and AREVA-CERCA, which combine the most promising options to avoid the formation of the IDL during irradiation [9]. From each miniplate a set of samples has been obtained which has been characterized before and after heavy ion irradiation using XRD and SEM/EDX.

This paper will focus on the effects of Silicon addition (2, 5 and 7wt%) into the Al matrix and the behavior under irradiation of the Si rich diffusion layer (SiRDL) grown around the UMo particles during fuel plate production. The results will be compared to in-pile irradiated samples (IRIS3 [10], RERTR-6 [11] and RERTR-7 [12]).

2. Mini plate production

Four miniplates are considered in this work: UMo/Al(Si2) (2wt% of Si into the Al matrix), UMo/Al(Si5) UMo/Al(Si7) as mentioned previously and a standard UMo/Al used as reference.

The UMo/Al(Si2) miniplate has been produced using commercially available ready mixed powder (Al and Si powder mixed, sintered and crushed). The UMo/Al(Si5) and UMo/Al(Si7) samples have been produced by blending very fine (-mesh 325) Al and Si powders of the highest purity. The mean particle size of the Aluminum and the Silicon powder have been determined using a laser granulometer and found to be ~20µm and ~7µm, respectively (compare Tab. 1).

Figure 1 shows a SEM picture of the blended Al(Si7) powder before sample manufacturing. The grain size of the particles is in accordance with values obtained using the laser granulometer. The Al powder has been produced by atomization, the Si powder by grinding.

All miniplates have been manufactured with a similar procedure than that used for IRIS full sized plates production. The IRIS-III plates were manufactured with the same Al2%Si pre-alloyed powder like the 2wt%Si samples presented in this study. The rolling temperature and the heat treatment were the same. However, there are certain differences concerning the process parameters and the UMo powder grain size.

Powder	Mean particle size	10% Diameter	90%Diameter
Silicon	7µm	1,2µm	15µm
Aluminum	20µm	10µm	30µm

Tab. 1: Particle size of Al and Si powder used to produce the 5wt%Si and 7wt%Si miniplates determined by a laser granulometer.

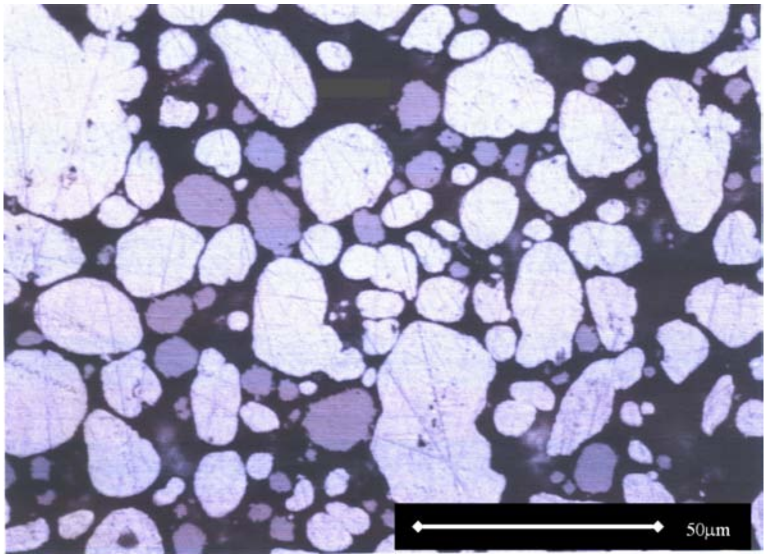

Figure 1: SEM picture of blended Al 7wt%Si powder before sample manufacturing. Si grains appear grey, Al grains appear white. The grain size is in accordance with values obtained with a laser granulometer. The Al powder has been produced by atomizing, the Si powder by grinding.

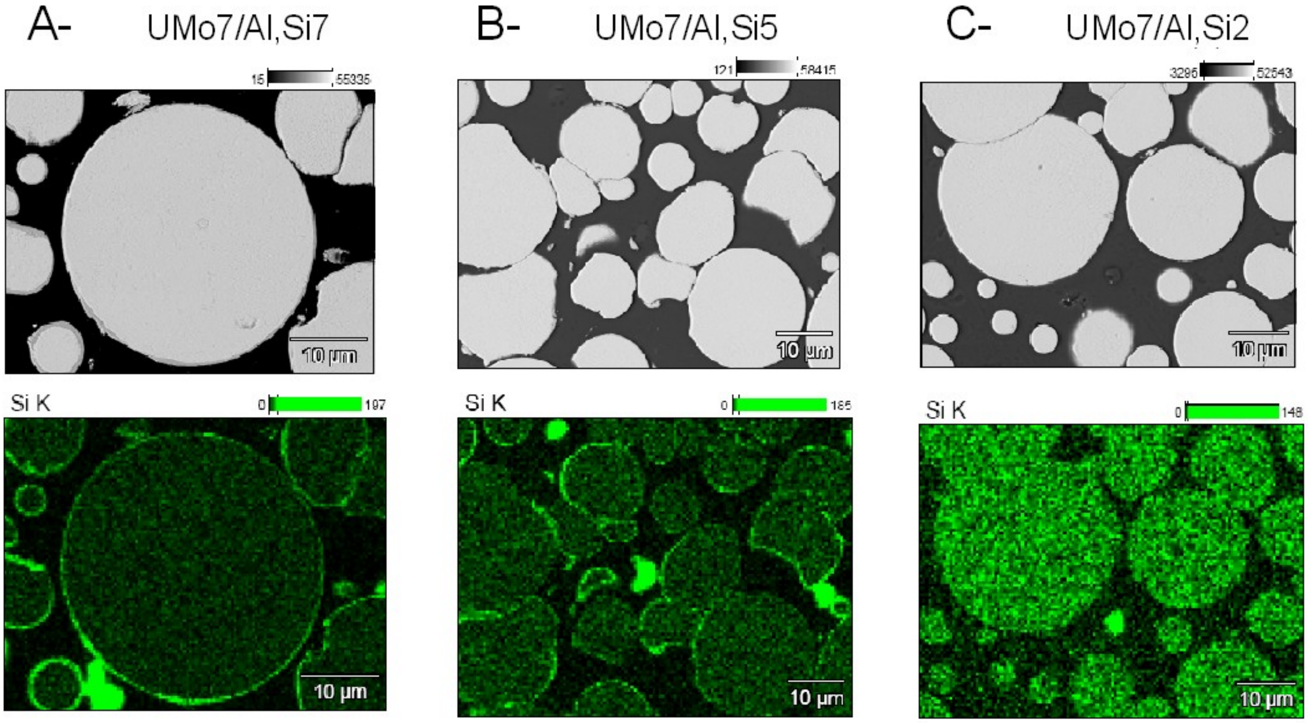

Figure 2: Comparison between Si repartition in two mini plates UMo7/Al(Si7) (A-), UMo7/Al(Si5) (B-) and UMo7/Al(Si2) (C-).

3. Characterization before irradiation

3.1. SEM/EDX analyses

Figure 2 compares the three miniplates containing 2wt%Si, 5wt%Si and 7wt%Si using SEM and EDX mapping. Three different features can be discussed:

- The growth of a Si rich layer around the UMo particles in the 5wt% and 7wt%Si case during production of the miniplates
- The presence of Si precipitates in the Al-matrix
- The occurrence of USiAl particles inside the matrix

In the 2wt%Si case usually no Si rich layer has been observed around the UMo particles after hot rolling, even if a Si precipitate is located very close to the UMo (compare Fig. 3). Rarely a very thin Si rich layer (<0.5µm), limited to only a few grains and a very small fraction of the affected UMo grain surface has been observed. This behavior is consistent with data that has been presented before [13]. However, in the case of the IRIS-III plate (Al2w%Si), which was manufactured with the same AlSi powder used for the 2wt%Si sample in our study, a SiRDL was observed but not on all the UMo particles (heterogeneous presence). The difference between the IRIS-III plates and the case regarded here may be related to the difference in process parameters and the grain size of the UMo powder.
In the 5wt%Si case each particle is at least partially covered by a Si rich layer (compare Fig. 1 and 4a). However, smaller particles reveal a thicker and more homogenous layer than bigger particles. The biggest particles are frequently only partially covered by the Si-rich layer. The thickness of the layer varies from 0.4µm to 1.2µm.
In the 7wt%Si case each particle is completely covered by the SiRDL (compare Fig 1 and 4b). However, also here the layer around the smaller particles is thicker than around the bigger particles. In general the thickness of the Si-rich layer is not very homogenous varying between 0.4µm and 2µm.
The Si rich layer can also be found around particles very close to each other (<1µm) – compare Fig. 5.
Table 2 summarizes the thickness of the Si rich layer found in the three different miniplates. Note that in general the layer is thicker for smaller particles (i.e. when looking at the top or the bottom of a cut through particle). That behavior has been taken into account with Table 2.

Si content wt%	Thickness of the Si rich diffusion layer (SiRDL)		
	Small sections (d<25µm)	Medium sized	Large particles (d>70µm)
2	-------		
5	~1µm	Irregular	
7	~1µm	Irregular	

Table 2: Influence of Si content in the Al matrix on the thickness of the silicon SiRDL . Quantitative values take into account the diameter of the UMo particle section considered.

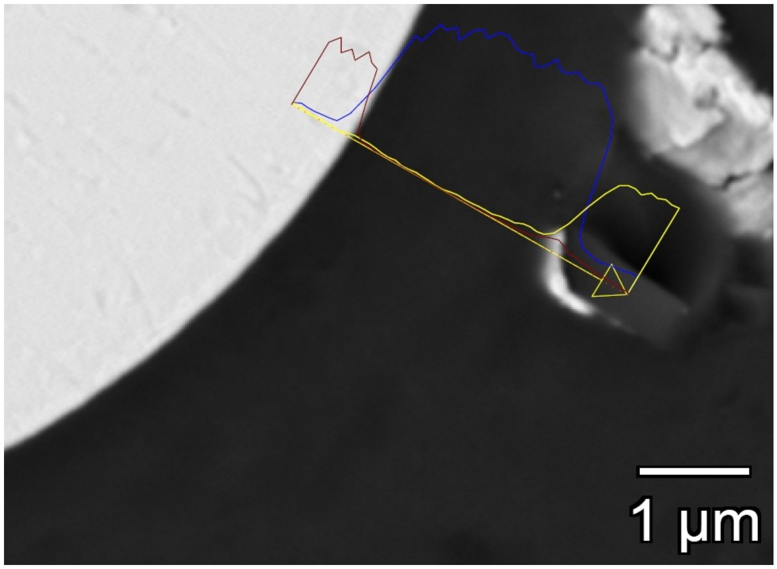

Figure 3: Si precipitate close to a UMo particle. The blue line indicates the Al content, the red line the U content and the yellow line the Si content. No Si rich layer has formed around the UMo particle.

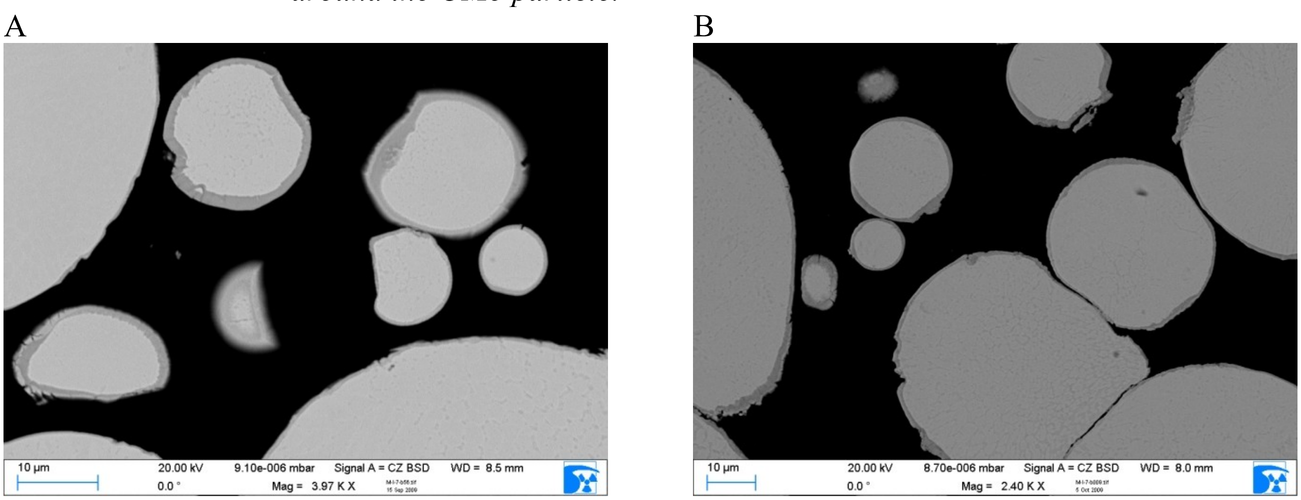

Figure 4: Si rich layer around UMo particles, BSE image. 3-A shows the 5wt%Si case, 3-B shows the 7wt% Si case

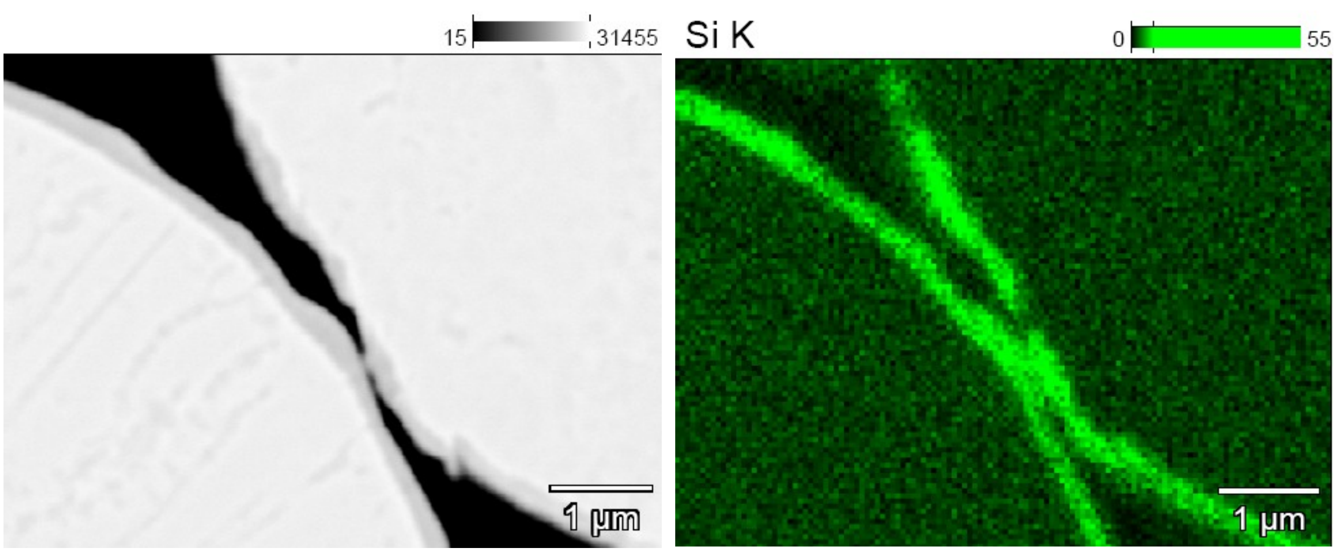

Figure 5 A Si rich layer is also present at spots where two UMo particles are very close to each other.

The composition of the Si rich layer has been determined by EDX analyses. Values are given in Table 3. The composition of the layer appears to be stable and seems to be the same for the 5wt%Si and 7wt%Si case. Note that since the layer is sometimes very thin the probed volume may be influenced by the surrounding Aluminum matrix such indicating a too high Al content inside the layer. However, the exact phase composition can be found using µXRD (for the method see [14].

Si content wt%	**Elementary composition of the SiRDL**				
	O	Si	Al	Mo	U
2	--------				
5	13,7 ± 2,1	**41,9 ± 2,3**	7,9 ± 1,7	5,0 ± 0,5	**31,8 ± 1,9**
7	14,1 ± 2,7	**39,0 ± 0,3**	12,6 ± 4,1	4,7 ± 0,6	**29,7 ± 1,7**

Table 3: Influence of Si content in the Al matrix on the thickness of the SiRDL . Quantitative values take into account the diameter of the UMo particle section considered.

EDX mapping makes large Si precipitates inside the Al matrix visible (compare Fig. 2). Their mean size is:

- ~8,5x4,3µm² for 2wt% Si
- ~8,7x6,1µm² for 5wt% Si
- ~9,1x6,9µm² for 7wt% Si

3.2. XRD analyses

A XRD measurement with a full Rietveld analysis has been performed with all samples before irradiation with heavy ions. The following phases have been found:

- γ-UMo
- Al
- UO_2 slight contamination at the sample surface
- traces of α-Uranium

Note that no traces of Si or USiAl phases have been found such indicating that the global amount of the Si rich layer is very low.

3.3. Discussion

Protecting the UMo particles with a SiRDL appears to be one of the most promising solutions for improving the in-pile performances of the UMo/Al nuclear fuels. Therefore, many other samples with very close composition have been produced worldwide. In the following a comparison is proposed.
Since the temperatures involved in the fuel plate production at AREVA-CERCA are known to be lower than those used for RERTR fuel plate production [11], it had to be investigated whether SiRDLs could be grown during the fuel plate production step of AREVA-CERCA. This work, even if performed on miniplates, demonstrates that this is the case for 5 and 7 wt% Si. Note that experience shows that SiRDLs are more difficult to grow in miniplates than in full sized plates.
If one compares the features of the SiRDL obtained in the UMo7/Al(Si5) miniplates with that of the very close UMo7/Al(Si4.8) full sized plate (included in the RERTR-7 test experiment) [12], some differences may be found in terms of thickness and Si content. Indeed the Si concentrations appear to be higher in the SiRDL that ranges between 45 and 69 %Si and the size of the SiRDL slightly higher (between 1 and 2µm) in RERTR-7, confirming the influence of the hot-rolling and blister test temperature.
The production of very few samples with a Si concentration higher 5wt% has been reported in literature. To our best knowledge, only one sample i.e. UMo7/Al(Si8) has been produced. This fuel rod has been

included in the KOMO-4 irradiation [15], but very little information is available on it. Moreover it is likely that the elevated size of the UMo particles as well the high temperatures chosen in the manufacturing process should make considerable differences.

4. Characterization after heavy ion irradiation

One part of the four available mini-plates (UMo/Al, UMo/Al,(Si2), UMo/Al(Si5) and UMo/Al(Si7)) has been irradiated at the tandem accelerator in Garching with a 80MeV ^{127}I beam. For each sample, the irradiation conditions were chosen identical: the total integral fluency is 1×10^{17} ions/cm² and the irradiation angle was 30° thus corresponding to a maximum of about 1000 dpa and 1at% of ^{127}I in UMo7 (SRIM calculation). Sample temperature, as measured at the rear of the miniplates, ranged between 150 and 300°C. Further experimental details on this technique can be found elsewhere [6, 7].
In these conditions, the SRIM2008 code gives an iodine penetration depth of about 2.5±0.7 in UMo7 and of 6.2 ±0.9µm in Al. Since the Al matrix is not 100% dense, this penetration depth into Al will be slightly higher: the presence of an IL can be observed up to at least 7µm below the sample surface.

4.1. XRD analysis

The samples have been examined after irradiation with heavy ions using XRD with Rietveld analysis. Beside the phases that were already present before irradiation, UAl_3 has been found. The lattice parameter of UAl_3 in the UMo/Al miniplate is ~4.23Å which is in agreement with the results obtained previously on heavy ion irradiated UMo7/Al samples [7]. Note that the UAl_3 cell parameter measured on the three other miniplates (UMo/Al(Si2), UMo/Al(Si5) and UMo/Al(Si7) also equals to 4.23 Å thus indicating that there is almost no Si present inside this phase i.e. no Si in the IDL grown by heavy ion irradiation.
As expected the α-Uranium content decreased, the UO_2 contamination on the sample surface increased during irradiation with heavy ions [8].

4.2. SEM/EDX analysis

The irradiated surface of each miniplate has been examined by SEM. However more accurate characterizations (i.e. elementary composition analysis or thickness measurements) require the study of transversal cross sections (Figure 6) [8]. This section shows the results of preliminary investigations. Figure 6 compares SEM observations performed on transversal cross sections of UMo/Al, UMo/Al(Si2) and UMo/Al(Si7) heavy ion irradiated miniplates.

A- UMo/Al(Si7) B- UMo/Al(Si2) C- UMo/Al

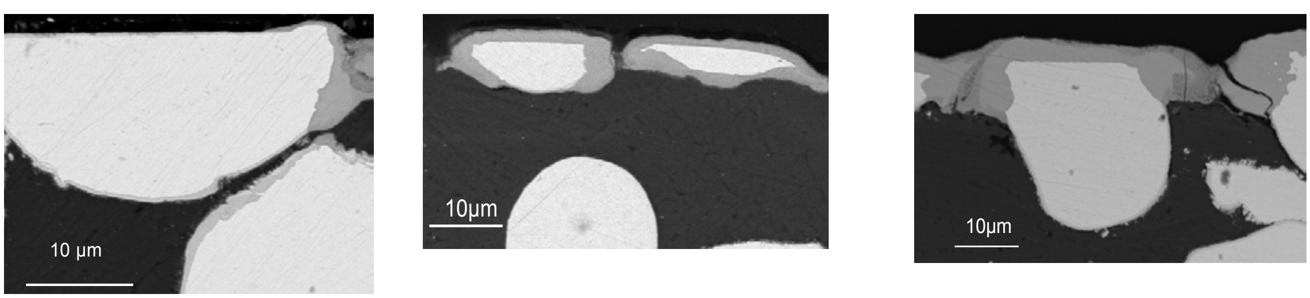

Figure 6: SEM observations of UMo/Al(Si7), UMo/A(,Si2) and UMo/Al heavy ion irradiated miniplates. The porous part of the IDL is the result of oxidation during the irradiation. Note that because of the irradiation conditions and the important IDL production, some IDL has flowed above the particles at the sample surface (B- and C-).

4.2.1. UMo/Al miniplate

The high flux and high temperature used during this irradiation have lead to the growth of a thick IDL (about 5µm). Moreover since the elementary composition of the IDL is directly linked to these two experimental parameters [17] (as this is the case for in-pile irradiated UMo/Al fuel [18]), a Al/(U+Mo) atomic ratio close to 3 was expected. An average value of 3.4±0.1 has been measured.

4.2.2. UMo/AlSi2 miniplates

The features of the IDL appeared in the UMo/Al(Si2) miniplate are very close to that observed in the UMo/Al reference: no Si has been found in the IDL and the IDL is just slightly thinner. Only when a Si precipitate was located close to the UMo/Al interface, a significant reduction of the size of the IL could be found. Compared to a very similar sample irradiated however at a higher dose and flux [16], the influence of the addition of 2%wt Si into the matrix is less obvious.

4.2.3. UMo/AlSi5 and UMo/AlSi7 miniplates

As in the fresh fuel, very few differences between the UMo/Al(Si5) and the UMo/Al(Si7) miniplates could be found after heavy ion irradiation. Since all UMo particles were covered by a SiRDL in these miniplates, the behaviour under heavy ion irradiation of this SiRDL could be investigated.
Observations of transversal cross sections show that the SiRDL has vanished in the zones concerned by the highest fluencies and a conventional IDL has formed. EDX measurements in the IDL (in the areas larger than 1 µm²) do not show the presence of measurable Si quantities except when a Si precipitate is close to this area (cf. Figure 6-A).
Considering first the irradiation conditions and in particular the value of the irradiation angle (30°) and second the different iodine penetration depth into UMo and Al (shadowing effect by UMo particles), each UMo/Al interface has not seen the same final fluency. Moreover, at the periphery of the irradiated zone, the obtained final fluency is less high.
Therefore from the irradiation of one sample, the influence of the fluency on the growth of the IDL can be deduced. It appears that the interaction process may be divided into two main parts (cf. Figure 7):

- First, an interaction occurs between the SiRDL and the Al matrix (cf. Figure 7-A) until the SiRDL has disappeared (cf. Figure 7-B). In this case, the IDL is often too thin for being probed with EDX, however it is likely that with the growth of the IDL, the Si concentration decreases (if no Si is located in the irradiated areas, close enough to the IDL as shown in Figure 8.). At this step, the UMo core of the particle is preserved from any interaction with Al.
- Afterwards, a conventional UMo/Al interaction appears (cf. Figure 7-C). The UMo particle is consumed now.

As a conclusion, the presence of the SiRDL around an UMo particle does not prevent by itself from the growth of a conventional IDL (generally silicon free) with however a lower thickness.
This result is in agreement with those obtained on the IRIS 3 and RERTR7 test fuel plates [10, 12]. Main conclusions of these experiments were that:

- the SiRDLs, present in the fresh fuel have been destroyed,
- Si is generally not present in the IDL except when Si precipitates were found to be in close contact with the UMo particles

This result would underline the important role played by Si precipitates located close to the UMo particle: they would act as a Si source definitely needed during the in-pile irradiation since it is shown here that the Si present inside the SiRDL will be not be sufficient for stabilizing the IDL. As a consequence, the AlSi alloy used as a matrix should be characterized by the presence of a high number of homogeneously dispersed small Si precipitates rather than large Si precipitates.
Note however that only one experimental condition (high temperature and final fluency) has been tested in this section. Further investigations are needed to confirm these results.

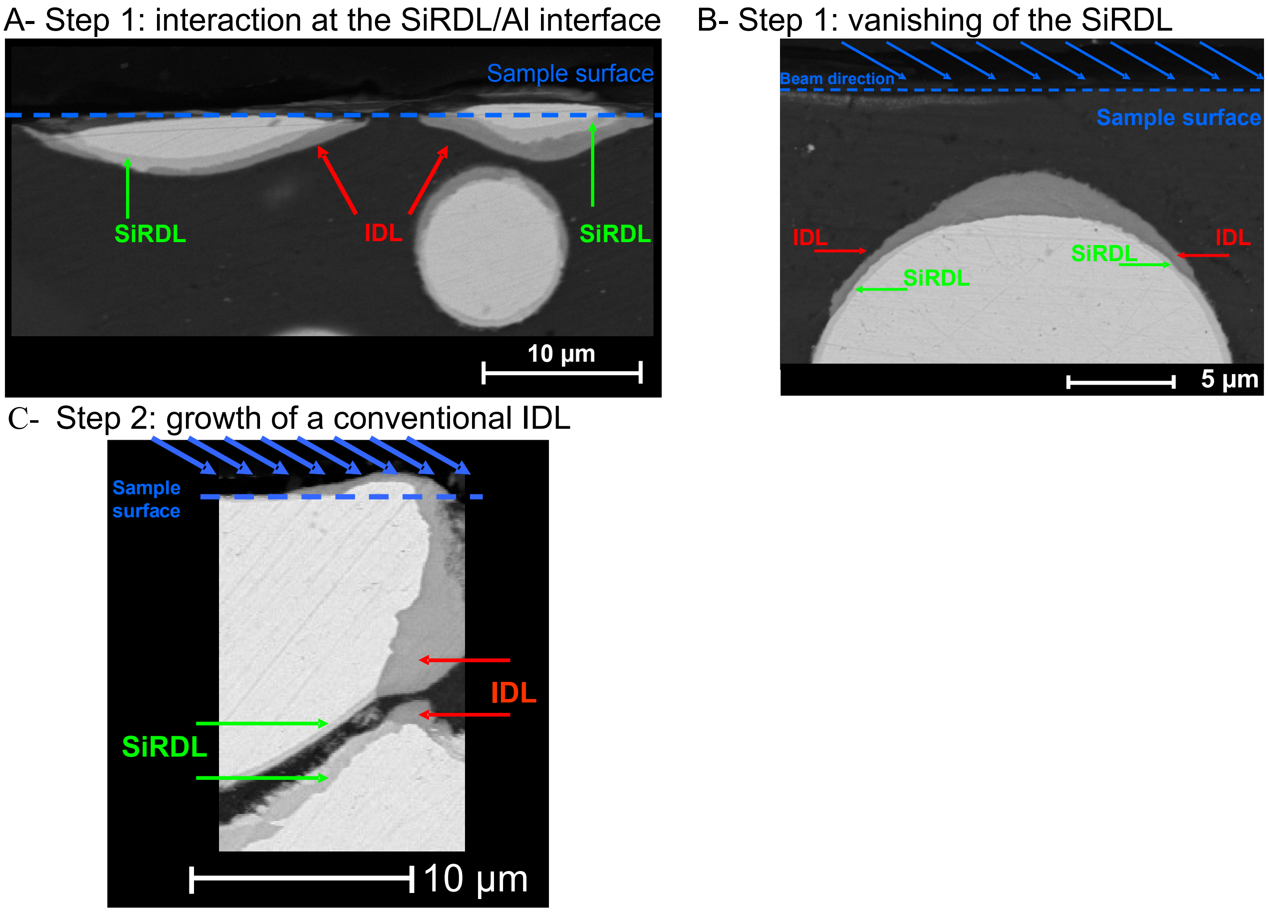

Figure 7: IDL growth around UMo particles protected by a SiRDL into two steps. Details of the transversal cross section of the heavy ion irradiated part of the UMo/AlSi7 miniplate. An IDL has formed according to the penetration depth of the heavy ions (red arrow). A SiRDL is visible around the UMo particle (green arrows).
A-The two UMo particles located at the sample surfaces are surrounded by first a SiRDL and then an IDL. Area taken at the periphery of the irradiated zone i.e. where the final fluency is lower.
B- In the upper part of UMo particle embedded in the Al matrix, the SiRDL has been destroyed by the heavy ion irradiation and the IDL is directly in contact with the UMo particle core.
C- A standard UMo/Al interaction occurs and the shape of the UMo particle indicates clearly that UMo has been consumed in the interaction.

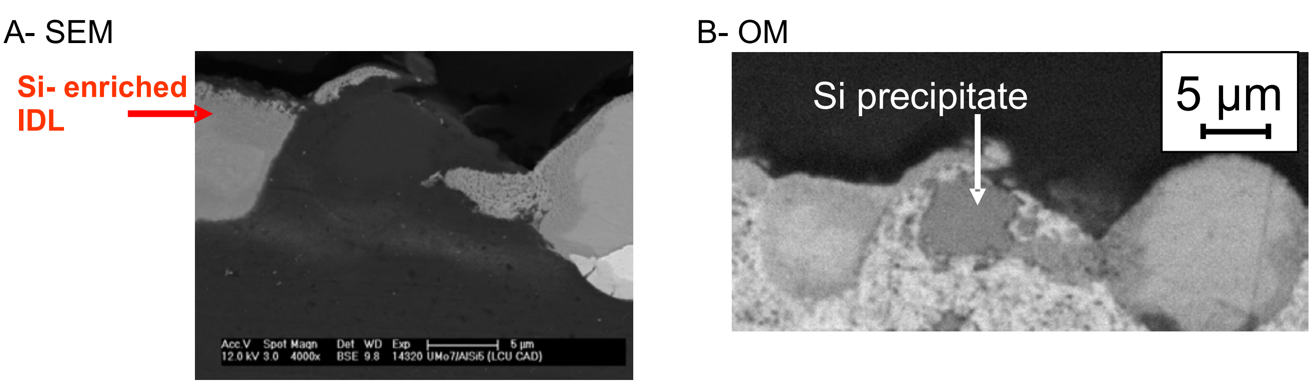

Figure 8: Presence of a significant Si concentration inside the IDL when a Si precipitate is located close to this IDL. SEM (A-) and optical micrographs (B-) of the same irradiated area of the UMo/Al(Si5) miniplate.

5. Conclusions

- In addition to the standard UMo/Al, three different miniplates containing 2wt%, 5wt% and 7wt% of Si inside the Al matrix one have been examined before and after irradiation with heavy ions
- A Si rich layer has formed around the UMo particles during the production of the miniplates in case of 5wt% and 7wt%Si. This is not the case for 2wt%Si.
- The SiRDL does not by itself protect the UMo particle from the growth of an IDL during heavy ion irradiation since the SiRDL is dissolved inside the IDL. The degree of decomposition is dose-dependent. When the SiRDL has been completely dissolved, a normal UMo/Al diffusion occurs in case there is no Si particle nearby.
- In case that a Si particle is in close contact to UMo, the resulting IDL is Si rich. Furthermore, the thickness of the IDL is significantly reduced in case that the UMo particle and the Si particle are in contact.

6. Acknowledgments

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