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MICROSTRUCTURE OF 50 YEAR OLD SCK•CEN BR1 RESEARCH REACTOR FUEL

A. Leenaers and S. Van den Berghe
Nuclear Materials Science Institute
SCK•CEN, Boeretang 200, 2400 Mol, Belgium.

ABSTRACT

The BR1 research reactor at SCK•CEN, Mol (Belgium) has a graphite core matrix loaded with fuel rods consisting of a natural uranium slug in an aluminum cladding. Fabrication reports show the application of a so-called AlSi bonding layer and an $U(Al,Si)_3$ anti-diffusion layer on the natural uranium fuel slug to limit the interaction between the uranium fuel and aluminum cladding.
The BR1 reactor is in operation since 1956 and still contains its original fuel rods. After more than 50 years irradiation at low temperature, the integrity of some of the fuel rods is investigated. The microstructure of the fuel, bonding and anti-diffusion layer and cladding is analysed using optical microscopy (OM), scanning electron microscopy (SEM) and electron microprobe analysis (EPMA).

1 Introduction

The BR1 research reactor is in operation since 1956. The reactor is mainly used as a neutron source for reactor physics experiments, neutron activation analysis, and calibration of nuclear detectors and instruments.
The reactor core consists of a graphite matrix, serving as moderator, in which the fuel rods are loaded in horizontal channels. These rods consist of a natural uranium (enrichment 0.7 % $^{235}U/U_{tot}$) cylindrical bar in an aluminum cladding.
For the past 20 years, the reactor has been operational for 3 days per week at a power of 700 kW for a maximum of 8 hours a day. BR1 is cooled by forced air convection with the help of a fan. In this way, the temperature in the reactor is kept well below 200 °C. After more than 50 years, the average fuel element burn-up is calculated to be only 0.07% FIMA.
At the manufacturing stage of the fuel rods, it was decided [1] to apply a so-called AlSi bonding layer and an $U(Al,Si)_3$ anti-diffusion layer on the natural uranium fuel slug. The bonding layer needs to ensure proper heat transfer and the anti-diffusion barrier is applied to limit the interaction between the uranium and the aluminum cladding. This type of interaction is likely to occur even at temperatures as low as 200 °C, which is also clearly observed in modern plate type fuel [2].

2 Fuel fabrication history

The ORNL graphite reactor X-10 [3], which is of similar design as the BR1 reactor, was powered by natural uranium fuel rods, clad with aluminum. The composition of the fuel rod was an uranium slug in an aluminum can, which was sealed off (by means of arc welding) with a cap as shown in fig. 2(a).

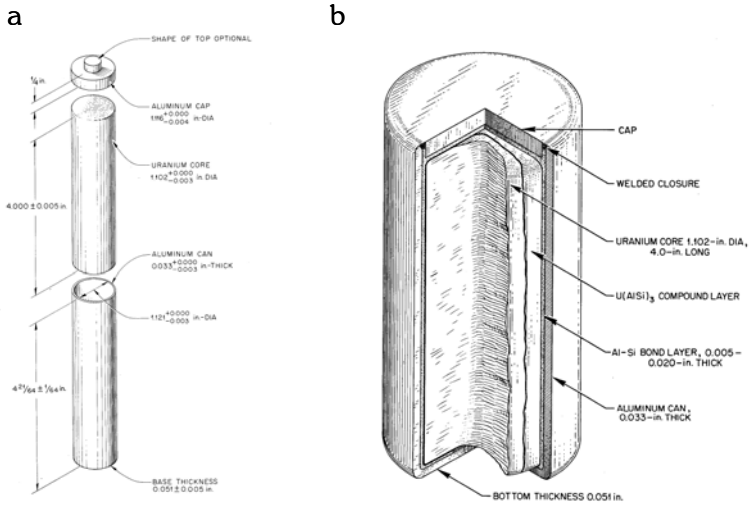

Figure 1 : Exploded view of the different components used to manufacture ORNL natural uranium fuel rods (a) and a schematic section of the canned fuel rod (b).(Figures scanned in from [3])

The original U slugs for the ORNL reactor were simply canned by forcing the slug in the can. However, the interaction between the uranium and aluminum, as well as air pockets in the cap weld caused failure of several of the rods. This has lead to the idea of using diffusion barriers and bond aids during the production of the fuel rods. The barriers were intended to inhibit the interdiffusion of uranium and aluminum at temperatures around 200 °C, whereas bonding layers were needed to enhance the heat flow across the interface.
Adequate bonding and impediment of the U-Al interaction could be achieved at the same time by using an aluminum-silicon alloy. By dipping the heated uranium slug in a molten aluminum-silicon alloy of eutectic composition, an adherent layer of $U(Al,Si)_3$ would form at the interface, acting as a deterrent to diffusion (in this paper this layer is further referred to as anti-diffusion layer). The use of a liquid metal during the subsequent canning of the slug, not only ensures a good bonding between the uranium rod and the aluminum can but also an intimate contact between cap and can (in this paper this layer is further referred to as bonding layer). The developed procedure to fabricated such a fuel rod was called the flux-alpha canning process. Details on this process can be found in [4].

The Al cans were deep drawn from commercially pure Aluminum 2S (currently called alloy 1100) sheet stock, while the caps were machined from wrought stock, free from porosity. The uranium slugs were machined from rods, degassed and heat treated to provide a randomly oriented grain structure.
In the canning operation, 3 molten metal baths were used :

1) The Al-Si coating bath to deposit the anti-diffusion layer on the U slug
   Bath temperature : 590 °C- 615 °C
   Bath composition (3 layers) :
   a) Lead layer to preheat the slugs
   b) Intermediate layer of Al-Si alloy (11.2 - 11.5 wt% Si)
   c) Flux layer to provide protective cover of the bath
2) Al-Si alloy (11.2 - 11.5 wt% Si) rinse bath : to rinse the slugs so the final Al-Si canning bath did not get contaminated.
   Bath temperature : (595 °C ± 5 °C)

3) Al-Si (11.2 - 11.5 wt% Si) canning bath to apply a bonding layer between fuel rod and aluminum can and cap.
   Bath temperature : (593 °C ± 3 °C)
   Canning procedure :
   a) The Al can was submerged and filled with Al-Si
   b) The U slug was submerged
   c) The U slug was inserted into the can
   d) The cap was submerged and inserted into the top of the can and pressed against the uranium slug

The complete assembly was then taken out of the bath and the excess Al-Si was removed. The fuel rod is thus composed of an uranium slug coated with an $U(Al,Si)_3$ anti-diffusion layer and tightly bonded to the Al can and cap through an AlSi bonding layer. Finally, a weld bead was run around the exposed braze line at the top of the rod using an argon shielded arc, without filler rod. The several features of a U slug canned using this technique are illustrated in fig.2(b).

3 Microstructure

The microstructure of an irradiated BR1 fuel rod, which was used for over 50 years, is analysed. For reason of comparison, the microstructure of an unirradiated rod (referred to as "fresh" fuel rod and originating from the same production batch as the irradiated rod) has also been investigated.
The top part of a fuel rod, containing the Al cap, part of the fuel slug and the Al cladding can, was cut off. A segment of the sample is embedded in an epoxy resin in such a way that a complete section of the rod (fuel, cap and cladding) can be observed (fig. 2). The mount is polished with SiC paper of successively finer grain size, finishing on cloth with diamond paste of 3 µm and 1 µm.

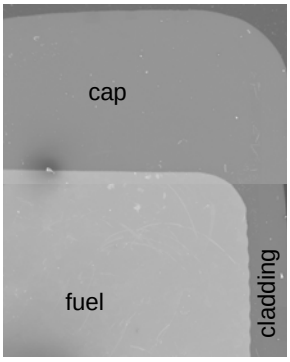

Figure 2 Collage of secondary electron images showing the top part of the fresh fuel rod in cross section.

The samples were analyzed using a shielded JEOL 6310 scanning electron microscopy (SEM) combined with an energy dispersive X-ray system (EDX).
The electronprobe micro analysis (EPMA) was performed on a shielded CAMEBAX-R Microbeam, upgraded with digital image and X-ray acquisition hard- and software. X-ray mappings were recorded to obtain the lateral distribution of several elements and the elemental composition was quantified using wavelength dispersive X-ray analysis (WDX). Prior to each measurement, a calibration was performed using the appropriate standards (table 1).

Element	X-ray line	Wavelength (pm)	Crystal	Standard
O	Kα	2362.0	PC1	UO_2
Al	Kβ	794.0	TAP	Al_2O_3
Si	Kα	712.7	PET	Si (pure)
U	Mα	391.0	PET	UO_2

Table 1 Crystals used for X-ray map and quantitative EPMA analysis. Prior to quantification of the elements calibration is performed using the appropriate standards.

The main features investigated, of both samples, are the bonding layer and the anti-diffusion layer.

3.1 Bonding layer

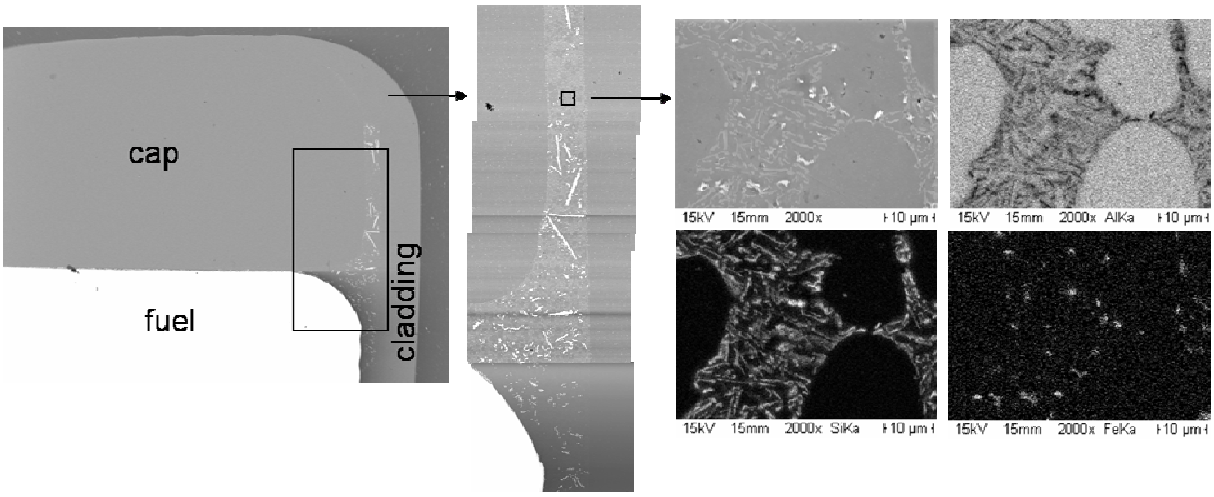

Figure 3 Backscattered electron images over the complete section of top of the fuel rod revealing the Al-Si bonding layer. The eutectic composition of the alloy is clearly seen in the X-ray maps recorded.

The backscattered electron image of the top of the fresh fuel rod (fig. 3a,b) reveals the bonding layer between the aluminum cladding/cap and uranium. This bonding layer is reported to consist of an Al-Si alloy with an eutectic composition. The EDX maps, recorded on a small area in the bonding layer, confirm this (fig. 3c).
It should be noted that the white needle-shaped inclusions observed in the bonding layer (fig.3a,b) are probably peeled off flakes of the U-Al-Si anti-diffusion layer. This layer is reported to have a very brittle nature [3]. Probably the flakes peeled off during the canning process (inserting of the $U(Al,Si)_3$ coated U slugs in the Al can, while being submerged in the AlSi bath).

3.2 Anti-diffusion layer

The detailed EDX maps (fig. 4) reveals the anti-diffusion layer (ADL) in-between the uranium slug (U) and the bonding layer (BL). Also note the large pure aluminum zones at the interface bonding layer / anti-diffusion layer.

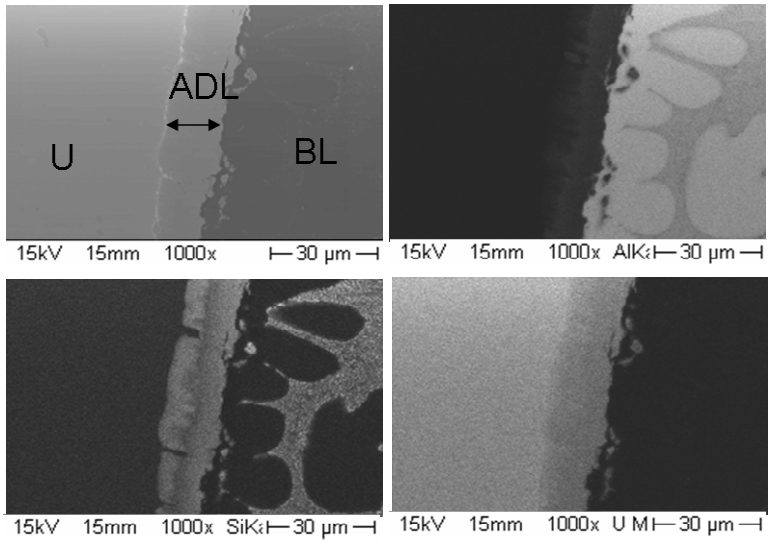

Figure 4 SE image and Al Kα, Si Kα, and U Mα X-ray map of an area covering the uranium (U), the anti-diffusion layer (ADL) and the bonding layer (BL) of the fresh fuel rod.

From the Si kα map in figure 4 and 5, it is clearly seen that the anti-diffusion layer in-between the uranium slug and the bonding layer consist of two layers.

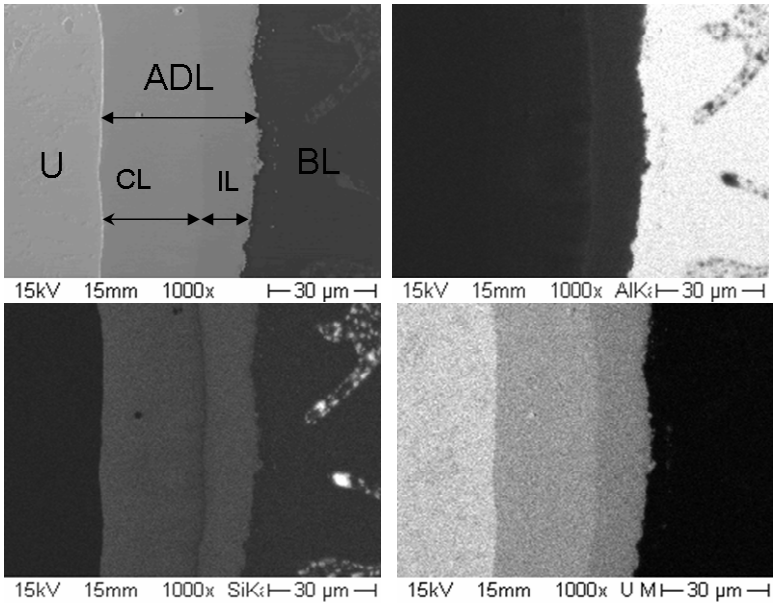

Figure 5 SE image and Al Kα, Si Kα, and U Mα X-ray map of an area covering the uranium (U), the anti diffusion layer (ADL) (consisting of a coating layer and interaction layer) and the bonding layer (BL) of the irradiated fuel rod.

For clarity reasons : the layer adjacent to the U slug is referred to as the coating layer (CL), while the layer between the coating layer and the bonding layer is referred to as interaction layer (IL).
Comparing the thickness of the anti-diffusion layer formed in the fresh fuel (fig.4) and in the irradiated one (Fig.5), it is found that the coating layer (CL) adjacent to the uranium slug has tripled in size during irradiation, while the interaction layer (IL), bordering the bonding layer, has kept the same size.

The increase in anti-diffusion layer thickness is also clearly observed in the optical micrographs taken of the fresh and irradiated fuel (fig.6a,b).

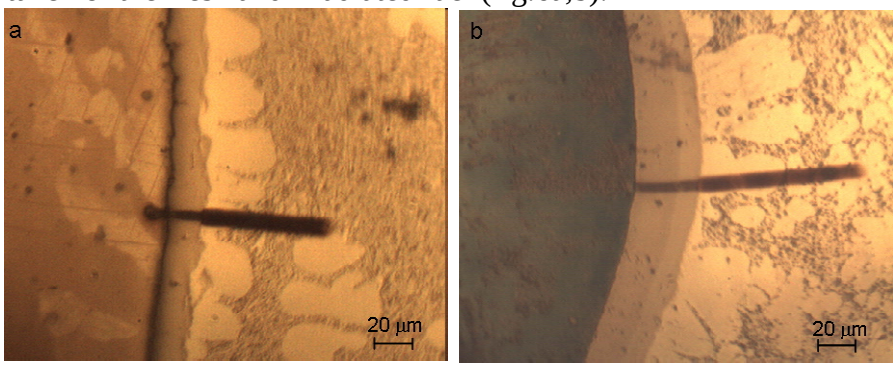

Figure 6 Optical micrograph of an area covering the fuel, anti-diffusion layer and the AlSi bonding layer in the fresh fuel(a) and irradiated rod (b). The dark line observed is a contamination trace created by the electron beam during the recording of a linescan with EPMA.

The optical micrographs also demonstrate that the pure Al zones, at the anti-diffusion layer/AlSi bonding layer interface, in the irradiated fuel rod are noticeably larger than in the fresh fuel.

4 Quantitative analysis

4.1 Fresh fuel rod:

A linescan starting in the fuel slug, covering the anti-diffusion layer and ending in the bonding layer has been defined (fig. 6a.). In each point (step) of the linescan the amount of U,Al and Si is measured.

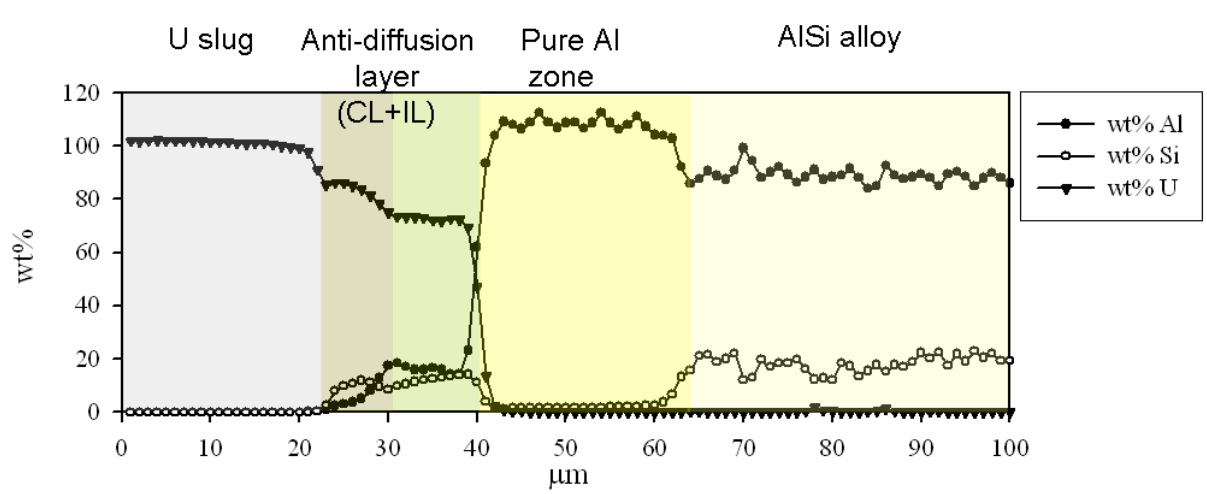

Figure 7. Quantitative linescan over de U slug, anti-diffusion layer and bonding layer observed in the fresh fuel rod.

The quantitative data in fig. 7 clearly show that the anti-diffusion layer consists of two different layers. The first layer, i.e. the coating layer, adjacent to the uranium slug is approximately 5 µm thick and is composed of 84 wt% U, 10 wt% Si and 4 wt% Al (mean values). The second interaction layer, in-between the coating layer and the bonding layer, is approximately 7 µm thick and contains 76 wt% U, 11 wt% Si and 16 wt% Al (mean values).
After the anti-diffusion layer, a nearly pure aluminum zone is measured preceding the AlSi alloy.

4.2 Irradiated fuel rod :

A similar linescan as defined for the fresh fuel, has been made on the irradiated rod (fig. 6b).

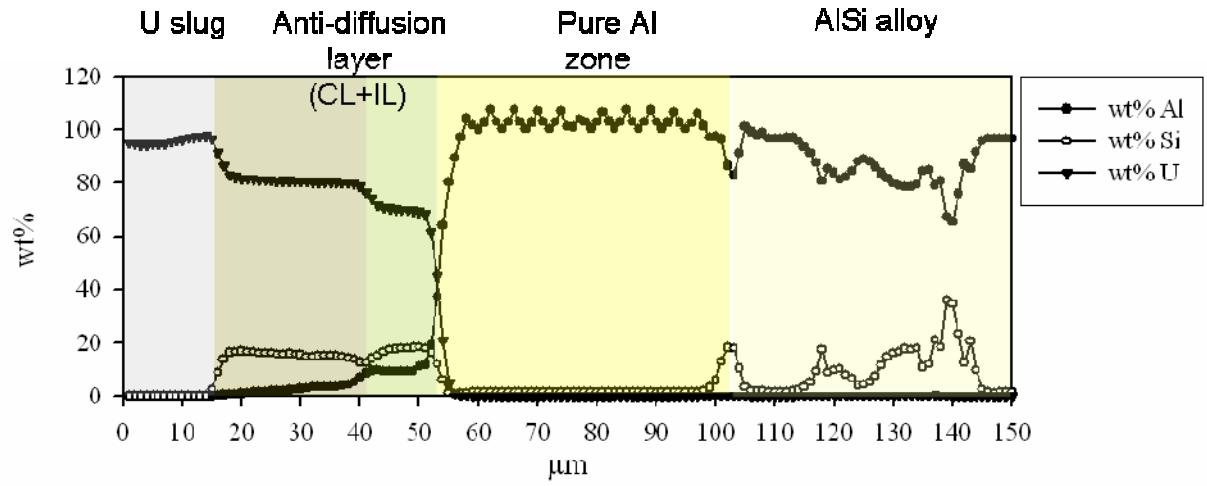

Figure 8 Quantitative linescan over de U slug, anti-diffusion layer and bonding layer observed in the irradiated fuel rod.

The quantitative linescan on the irradiated fuel (fig. 8) immediately confirms that the width of the anti-diffusion layer has substantially increased in comparison to the width of the ADL in the fresh fuel.

The coating layer adjacent to the uranium slug is approximately 21 µm thick and contains 80 wt% U, 16 wt% Si and 3 wt% Al (mean values). The interaction layer is approximately 6 µm thick and is composed of 70 wt% U, 18 wt %Si and 10 wt% Al (mean values).

Also in this linescan the anti-diffusion layer is followed by respectively a pure Al zone and the AlSi alloy.

5 Discussion

The irradiation of a natural uranium fuel rod with an aluminum cladding for more than 50 years, up to a burnup of 0.07% FIMA appears to had no detrimental effect on the fuel nor the cladding. The only found particularity is the growth of the anti-diffusion layer on the fuel slug, which was already applied during fabrication.

The first step of the production process of the fuel elements consists of the deposition of an anti-diffusion layer (ADL) on the uranium slug by submerging the preheated slug in an AlSi eutectic melt. The ADL is clearly visible in the fresh fuel examinations. We assumed that, after the initial deposition, the ADL would be a single layer, but the current observations reveal a double structure with two different compositions. This is thought to be the consequence of the third step in the fabrication, namely the canning process itself in which the coated slugs are submerged (this time without preheating) in another AlSi melt bath and inserted in their capsules. During this process, the deposited ADL will react with the AlSi melt and form an interaction layer (IL). The canning bath is intended to ensure a good thermal conduction between aluminum can and coated slug by providing a bonding layer (BL) between them.

The formation of a double anti-diffusion layer was also observed in an out-of-pile diffusion experiment. In an U-AlSi diffusion couple, annealed at 400 °C, the diffusion zone appeared metallographically as two layers. It is measured by XRD that the 2 compounds correspond to UAl_3 (near the U interface) and USi_3 (near the AlSi interface) [5]. Building on those results, the CL neighboring the U slug was presumed to have a $U(Al,Si)_3$ [3] or UAl_3 [5] composition, while the IL was assumed to have an USi_3 composition [5]. However, the previously reported

compositions of the two layers observed, do not agree with the quantification obtained by EPMA in this study.

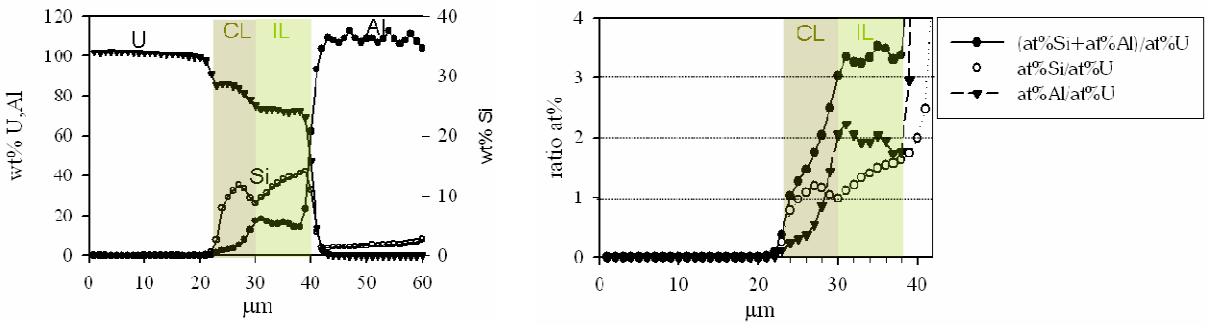

Figure 9 Quantification of the double interaction layer observed in the fresh fuel.

From fig. 9 it is seen that the CL on the U slug mainly consists of silicon and uranium. The ratio of the at% shows that the composition of the layer is close to USi. This means that when the uranium slug was submerged in the molten eutectic AlSi coating bath (at ~ 600 °C), U has preferentially interacted with the Si of the melt, forming a thin (~ 5 µm) USi layer. During the canning process, this USi layer will then have reacted with the Al-Si melt in the canning bath and formed the IL consisting of U, Si and Al. From the measured composition, this interaction layer can be identified as $U(Al,Si)_3$, with a Al/Si ratio of 1.3. This reaction is indicated on the ternary diagram of fig. 10 and lies on the dotted line connecting USi and Al.

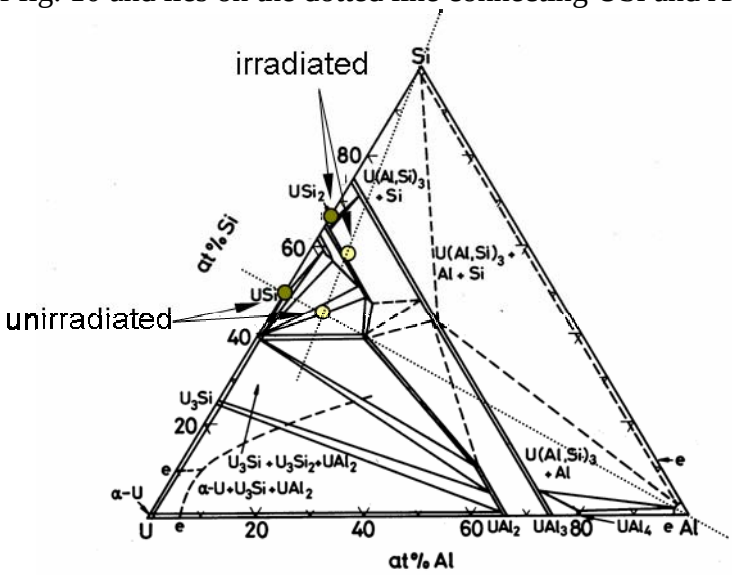

Figure 10 Isothermal section at 400 °C of the ternary system U-Al-Si [6]. The composition of the coating layer (CL●) and the interaction layer (IL○) found in the fresh an irradiated fuel rod examined in this study is indicated by the dots.

The microstructure analyses further show that at the interface between the anti-diffusion layer and the AlSi bonding layer, large pure Al zones have formed. A possible explanation for this is that silicon is preferentially consumed by the anti-diffusion layer during the early stages of the intermetallic bonding of the two materials. The AlSi bonding layer will get locally depleted in Si and therefore yield the observed larger Al zones [7].

Observations on the irradiated specimen show that irradiation for 50 years at moderately low temperature (~ 140 °C) has caused the anti-diffusion layer to grow. The quantification of the composition of the double ADL in the irradiated fuel rod (fig. 11) shows that the CL is ~ 21

μm thick and no longer has a USi composition but changed to USi_2. Again, only very low amounts of Al are found in this layer, increasing towards the interface with the IL. This layer still has a $U(Al,Si)_3$ composition but compared to the measurements on the fresh fuel rod, the Al/Si ratio has changed to 0.5.

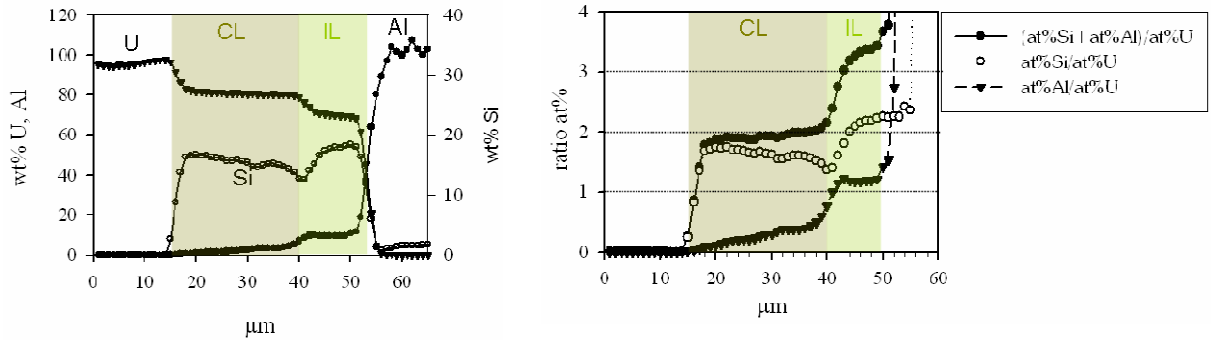

Figure 11 Quantification of the double interaction layer observed in the irradiated fuel.

These results thus indicate that the irradiation has caused the diffusion of Si into the ADL. The proposed mechanism for the Si diffusion (fig. 12), and according to the binary diagram [8], is that at low temperature the USi layer (CL), will react with the silicon of the $U(Al,Si)_3$ (IL) and forms an USi_2 layer. The Si deficit in the $U(Al,Si)_3$ layer will be equalised by the supply of silicon coming from the AlSi BL (this reaction is indicated on the ternary diagram of fig. 10 and lies on the dotted line connecting $U(Al,Si)_3$ and Si). As the BL is a rich source of free silicon, the diffusion mechanism will continue. As a result the USi_2 layer will continuously grow and the BL will get more depleted in Si, resulting in the growth of the pure Al zones observed.

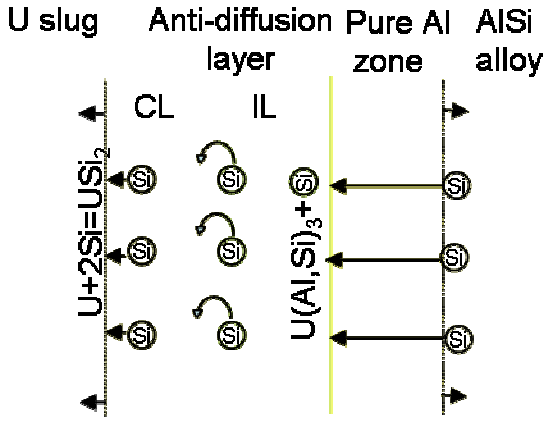

Figure 12 Proposed diffusion mechanism of silicon through the anti-diffusion layer

For the IL, the simultaneous supply of Si from the BL and the supply of Si to the CL, causes the width of the IL to remain virtually unchanged. The Al from the IL will only partly diffuse into the CL as can be seen in the EPMA results. On average, the composition of the IL remains the same ($U(Al,Si)_3$) but the ratio Al/Si changes in favor of Si.
As it turns out, it has been the unintended growth of a thin USi layer on the uranium slug during the fabrication of the rods, that has mostly limited the interaction between the uranium and the aluminum cladding.

The obtained results can be related to modern MTR fuel (mainly UMo but also U_3Si_2), where during irradiation an interaction occurs between the fuel and the aluminum matrix (dispersion fuel) or cladding (monolithic fuel), both resulting in the formation of an $(U,Mo)Al_x$ layer.

This layer continues to grow during irradiation and it is suggested [9] and measured [10] that it becomes amorphous during irradiation at moderate temperature. As a result, the $(U,Mo)Al_x$ layer will no longer pin the fission gas and releases it to the interface of the $(U,Mo)Al_x$ layer with the aluminum matrix or cladding. Depending on the irradiation condition (high burn-up), these gas filled pores continue to grow, linkup and finally will cause unacceptable swelling of the fuel plate [2].
Based on thermodynamic and metallographic analysis, it is suggested [11] that addition of silicon to the Al matrix would stabilize the growth of the $(U,Mo)Al_x$ interaction layer. The first irradiation tests indeed show that the average plate swelling decreases with increasing Si content in the matrix [11]. The underlying mechanism responsible for this effect is not fully understood. However, based on the result found in current study it can be assumed that the addition of a larger amount of silicon to the aluminum matrix, causes the creation of a thin USi layer on the fuel particles. It is shown in this paper that such a thin USi coating is an effective aluminum diffusion barrier.
It should be noted however that, considering the low burnup (0.07% FIMA) of the currently investigated fuel rods, hardly any fission gas has been created. The behavior of fission gas in the USi layer is thus not known.

6 Conclusion

After being irradiated for more than 50 years in the BR1 reactor, the natural uranium aluminum cladded fuel rods appears to be in very good condition.
It is found that the AlSi bonding layer does indeed provides a tight connection between fuel and cladding. The applied anti-diffusion layer however is not homogenously composed of $U(Al, Si)_3$. The dipping of the uranium slug in the molten eutectic AlSi coating bath has created a thin USi layer. During the canning process, this USi layer has reacted with the molten AlSi bath and formed a second $U(Al,Si)_3$ layer. It has been shown that it is the inadvertently created USi layer that acts as the anti-diffusion barrier for aluminum.
Irradiation at moderate temperature, causes that Si to diffuse through the interaction layer and to interact with the uranium slug. The source of this additional Si is identified as being the AlSi bonding layer.

7 References

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