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HEAVY ION IRRADIATION OF U-MO/AL DISPERSION FUEL

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Abstract

High-density U-6wt%Mo/Al and U-10wt%Mo/Al dispersion fuel samples have been irradiated by heavy ions in order to simulate fission fragments. The dose was set to 10^{17}Ion/cm², which corresponds to a low burn-up as compared to in-pile irradiation. The aim of the irradiation is to investigate the cause of the interdiffusion layer, which develops between the U-Mo fuel particles and the surrounding Al-matrix. Post irradiation analysis has shown an interdiffusion layer due to irradiation by heavy ions. This interdiffusion layer has similar properties as found by reactor irradiation.

Introduction

The high density U-Mo/Al dispersion fuel suffers mainly from the unfavourable interdiffusion layer (IDL) between the fuel and the Al-matrix, which tends to break up at the transition to the Al-matrix. Up to now this IDL was observed only by in-pile reactor irradiation and could not be created in out-of-pile experiments. Annealing experiments showed a diffusion behaviour as well, see for instance [1,2], but thermally activated diffusion should be excluded as a reason for in-pile diffusion, because a much higher temperature is required (at least 400°C) and diffusion would be much faster than in-pile.

Why an irradiation by heavy ions?

During in-pile irradiation of nuclear fuels, fission takes place. Per thermal fission of Uranium-235 energy of around 204MeV is released. The main part (167±5MeV, more than 80%) of energy is carried away by the kinetic energy of two fission fragments [3]. These two fission fragments have an atomic mass number of around $A \approx 95$ respectively $A \approx 137$ (heavy ion)[4]. Fission fragments are usually not stable and therefore cause a high radiation and dose rate after in-pile irradiation, which hampers the post irradiation examination significantly.

Our approach is to irradiate U-Mo/Al dispersion fuel with typical fission fragments which are stable. For practical purposes we have chosen I-127 as projectile at 120MeV. This projectile has several advantages in comparison to others: first it is a typical fission fragment, secondly it can be accelerated to a typical energy of fission. And thirdly the Munich tandem accelerator at Garching, Germany can provide a high flux with this isotope.

Experiments

Three samples have been investigated which have been provided by the RERTR-team (Argonne, USA), see table. Before irradiation the samples have been polished in order to uncover the meat-layer and to obtain a clean surface of 5 x 5 mm². The beam size was set to 2 x 1 mm² and the heavy ion flux was limited in order to not exceed a meat temperature of 200°C, measured close to the irradiation position by a thermal couple. A final fluence of 1 x 10^{17} ion/cm² was achieved after max. 13h of irradiation. This fluence corresponds to a low burn-up as compared to in-pile irradiation. While fission fragments are emitted isotropically, the

Sample (label)	ANL#2	ANL#6	ANL#7
Fuel	U-6wt%Mo	U-10wt%Mo	U-10wt%Mo
Matrix	AL	AL	AL
Fluence	$1x10^{17}$ ion/cm²	$0,5x10^{17}$ ion/cm²	$1x10^{17}$ ion/cm²
Flux (maximum)	$3x10^{11}$ ion/s		
Temperature (maximum)	200°C		

heavy ion beam is vectored [7]. Alike in vacuum the divergence of the beam in matter is still marginal, but the penetration depth differs, see diagram 1. Because fuel particles and matrix are hit by the heavy ion beam in the same way an effective penetration depth for the meat could be defined in order to compare fission density and fluence. Henceforth a comparison of orders of magnitudes is possible. For instance the final fission density in the fuel meat of the FRM-II research reactor in Germany is $2x10^{21}$fission/cc [6] i.e. $4x10^{21}$ fission fragments/cc. Assuming that the effective penetration depth perpendicular to the surface is 5µm (for an irradiation angle of 30° between beam and surface) a fluence of 10^{17} ion/cm² corresponds to a fission fragment density of $2x10^{20}$ fission fragments/cc. However, the latter value has to be corrected at least by the volume fraction of the fuel in the meat and the influence of the direction.

Discussion

Since the samples are not activated, post irradiation examination can be done easily and immediately after the irradiation.

Optical microscopy

Because of the irradiation by iodine and the penetration depth of only some micrometers, the irradiated surface appears very dark. Furthermore due to the viscose wavelike pattern which develops on the surface, optical microscopy is difficult. Nevertheless pictures 1, 2, and 3 show irradiated samples ANL#2, ANL#6, and ANL#7 respectively. Each picture shows the centre of the irradiated area, and clearly three separated phases – fuel particles, matrix and an interdiffusion layer in-between. While the diameter of the fuel particles varies between some µm and approximately 80µm, the thickness of the interdiffusion layer seems to be constant at about 10µm to 30µm depending on the direction of the beam and on the sample.

Scanning electron microscopy and EDX

Picture 4 shows a SEM image of an U-6wt%Mo particle in Al (sample ANL#2), which was irradiated under an angle of 30°. Like the optical microscopy three phases are visible, again. They are separated by sharp borders. The U-6wt%Mo fuel particle in the middle has a diameter of around 60µm and some ripples on the surface are visible. This wavelike pattern is an irradiation effect and did not exist before the irradiation. Surrounding the fuel particle, an interdiffusion layer is observable, which is particularly pronounced in the direction of the heavy ion beam (from bottom-left to top-right). The maximum thickness is around 30µm. Within the interdiffusion layer the chemical composition is almost homogeneous as found by EDX (energy dispersive X-ray spectroscopy), see diagram 2.

Picture 5 shows two SEM images of the samples ANL#6 and ANL#7. In principal they can be characterised in the same way as the sample ANL#2. But both samples show a significant smaller interdiffusion layer (max. 16µm for ANL#6 and max. 21µm for ANL#7) of its thickness with a strongly reduced dependence on the ion beam direction. EDX has shown also a better homogeneity of the interdiffusion layer compared to sample ANL#2.

Conclusion

Up to now only three samples have been irradiated in the same manner and they show promising results, but nevertheless further irradiations and examinations have to be done.

The observed new interdiffusion layer has properties in common with the interdiffusion layer, which was found by in-pile irradiation. Optical properties, chemical composition and dimensions are comparable. Optical and SEM-pictures of in-pile irradiated U-Mo/Al dispersion fuel can be found for instance in [5].

Assuming that the interdiffusion layer which has been found by irradiation with heavy ions and the interdiffusion layer, which was observed after in-pile reactor irradiation are the same, this technique will give a possibility to support the development of U-Mo/Al dispersion fuels.

The advantages compared to in-pile irradiation would be:

- very quick results (some hours of irradiation by heavy ions are sufficient to simulate a moderate burn-up)
- no activation of the sample (handling and post irradiation examinations will be much easier)
- less expensive

Acknowledgement

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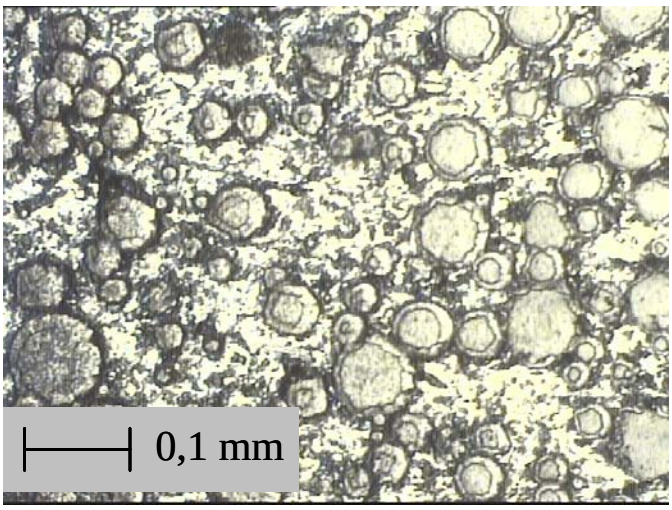
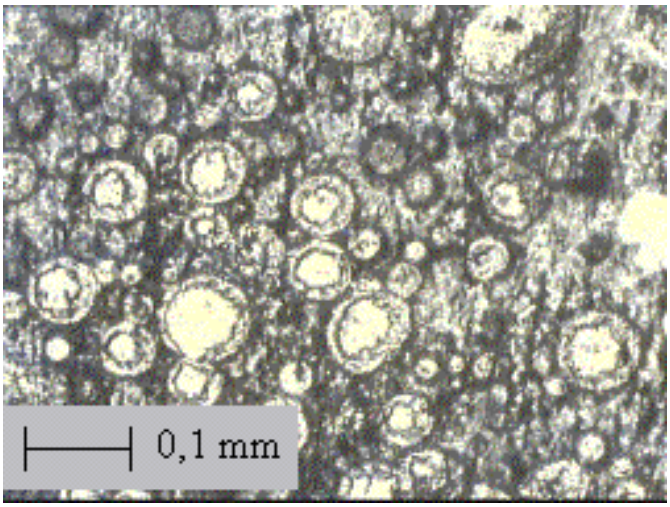


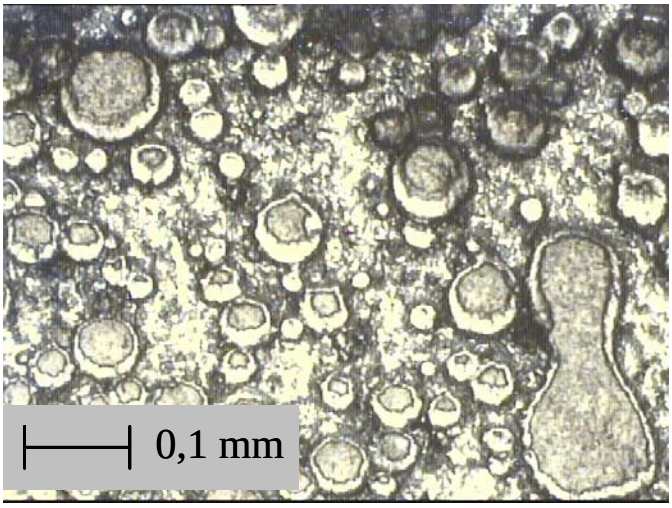
Picture 1:

optical microscopy of U-6wt%Mo/Al after irradiation (ANL#2)



Picture 2:

optical microscopy of U-10wt%Mo/Al after irradiation (ANL#6)



Picture 3:

optical microscopy of U-10wt%Mo/Al after irradiation (ANL#7)

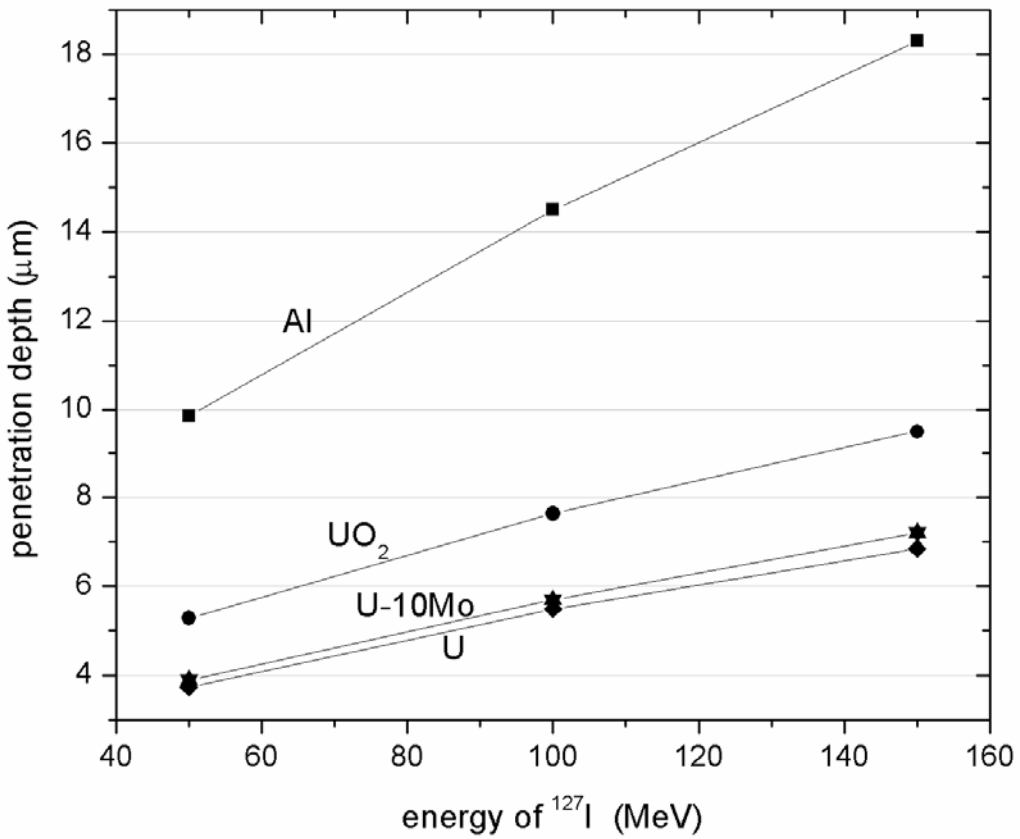
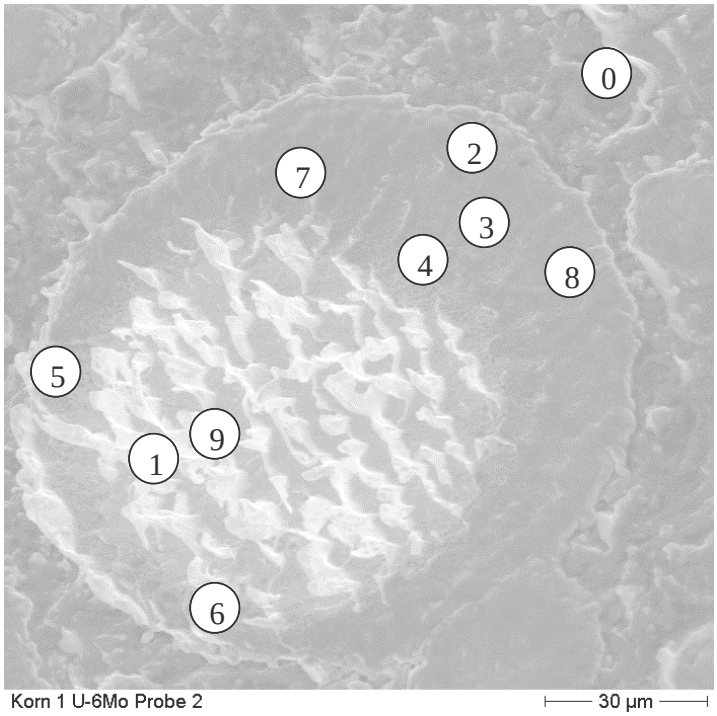


Diagram 1: penetration depth of iodine –127 into different materials [7]



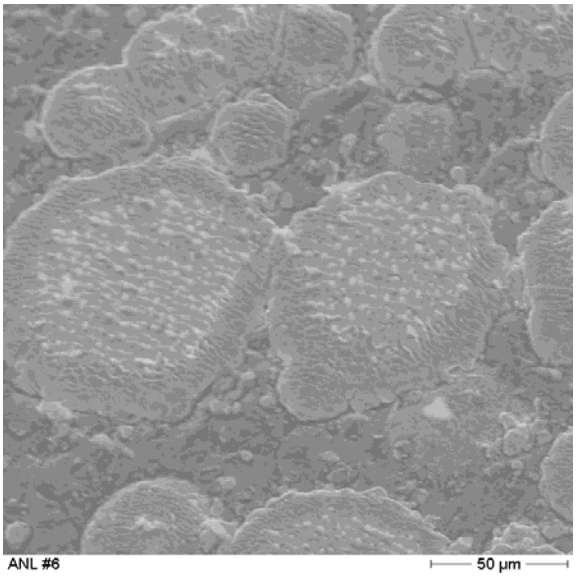
Picture 4:

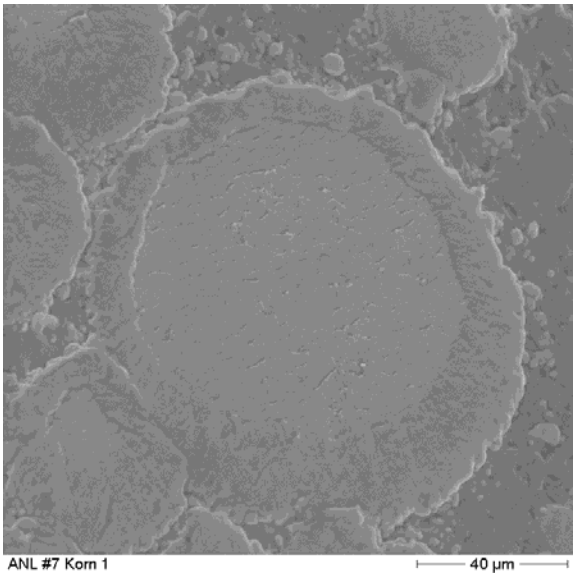
SEM of irradiated U-6wt%Mo/Al sample (ANL#2)

at position #1..9 EDX measurements have been carried out, see diagram 2

at position 0 pure Al has been found

the ion beam direction was from bottom-left to top-right





Picture 5: SEM-Pictures of sample ANL #6 (left hand side) and ANL#7 (right hand side)

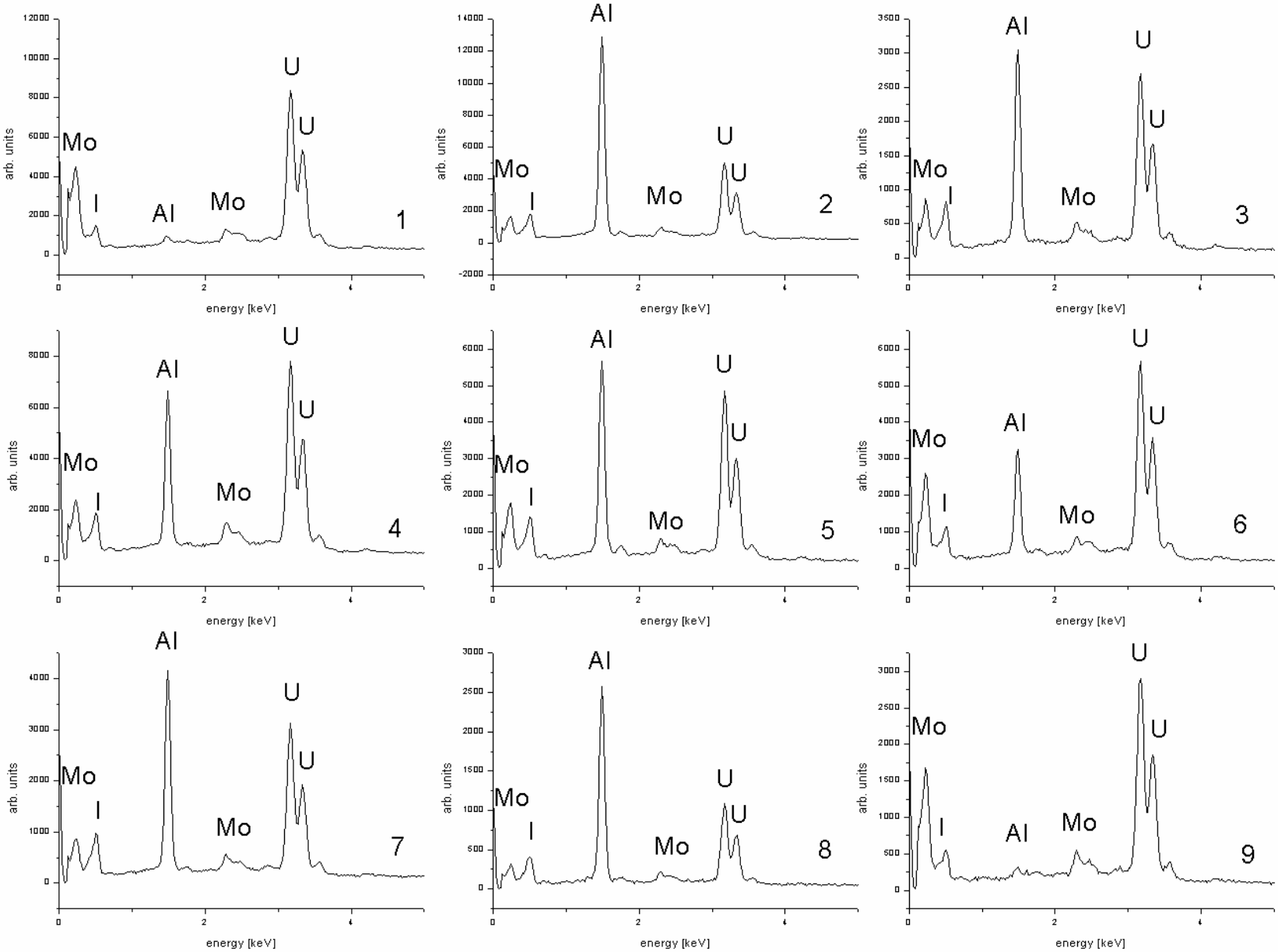


Diagram 2: EDX- measurements of sample #2 (U-6wt%Mo/Al) at different position, see Picture 4