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Journal of Nuclear Materials 357 (2006) 191–197
journal of nuclear materials
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Heavy ion irradiation of U–Mo/Al dispersion fuel

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Received 10 October 2005; accepted 13 June 2006

Abstract

The usage of high-density U–Mo/Al dispersion fuel for high burn up in research and test reactors seems to be limited by the unfavourable interdiffusion layer between the fuel and the Al-matrix, which develops during irradiation. This interdiffusion layer was observed up to now only after costly and time consuming in-pile irradiation and could not be created in out-of-pile experiments. This paper presents a new approach of creating such an interdiffusion layer out-of-pile by irradiation with heavy ions. An appropriate choice of heavy-ion irradiation simulates irradiation damage and deposition of fission fragments as it happens during in-pile irradiation and induces a diffusion process between the fuel and the Al matrix. An irradiation experiment and post-irradiation examinations are presented.

1. Introduction

To minimize proliferation risks it is desirable to use – whenever possible – low enriched uranium (LEU) instead of highly enriched uranium (HEU) fuel for research and material test reactors. In order to achieve this goal high density fuels are required. For high performance reactors, U_3Si_2/Al dispersion fuel represents up to now the highest qualified uranium density of up to 4.8 gU/cm^3 [1]. But for high flux research reactors even a higher uranium density is required in order to convert the fuel from HEU to LEU. One of the most promising candidates is the metallic U–Mo/Al dispersion fuel. This fuel would allow a uranium density of around 8.5 gU/cm^3 and is currently under worldwide development [2].

However, since the year 2003 some concerns came up due to anomalous swelling of full size plates during irradiation at high neutron flux and

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doi:10.1016/j.jnucmat.2006.06.006

heat load values. Post-irradiation examinations (PIE) of these plates revealed an Al-rich interdiffusion layer building up at the interface between U–Mo grains and the Al-matrix with a thickness of some 10 μm [3]. Cracks which are regarded as the reason of the breakaway swelling have been observed exactly between the interdiffusion layer and the matrix [4].

Therefore the build-up and the composition of the interdiffusion layer are of main interest. But because of the strong activation after the in-pile irradiation of the samples only a few types of examinations have been carried out like optical microscopy and scanning electron microscopy (SEM). Here we show an approach to generate an interdiffusion layer by out-of-pile irradiations with heavy ions thereby simulating the radiation damage created by fission fragments during reactor irradiation without the drawback of creating difficult to handle strong radioactive samples.

2. Sample preparation

Fuel plates containing atomized (spherical shape) U–6 wt%Mo and U–10 wt%Mo powder dispersed in aluminium were provided by the RERTR-team (reduced enrichment for research and test reactors) at ANL (Argonne National Laboratory) where they have been manufactured like miniplates for in-pile irradiation. The U-density in these specimens is about 8 gU/cm^3 and the maximum diameter of the spherical U–Mo grains was around 120 μm.

Initially specimens were cut into pieces of 5×5 mm^2. Afterwards they were polished in order to get rid of the cladding and to make the meat layer accessible for irradiation with heavy ions. Since the penetration depth, and therefore the modification by irradiation with heavy ions, is in the order of some micrometers (see Fig. 1) a good polishing is of essential importance.

3. Irradiation condition

The irradiation experiment has been carried out at the Munich 14 MV tandem accelerator in Garching near Munich, Germany. All specimens were irradiated with I-127 at 120 MeV under high vacuum. Iodine was chosen because it allows on the one hand a high particle flux and on the other hand it represents a typical fission fragment. Since cooling of the specimen has to be achieved mainly by thermal conductivity, the specimens were screwed on

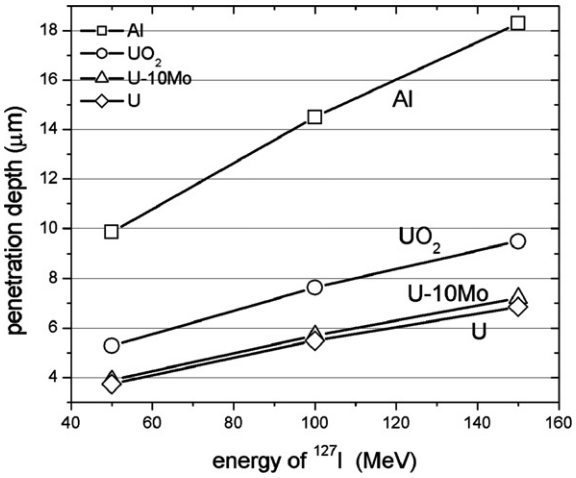

Fig. 1. Penetration depth of iodine into selected materials versus kinetic energy [5].

an aluminum block. A maximum heavy ion flux as high as 4.4×10^{11} s^{-1} (i.e., 10 W) was chosen in order to limit the specimen temperature to a maximum of 200 °C, which was measured *in situ* by a thermocouple on the rear side of the specimen. This temperature covers conservatively the temperature conditions in the meat of high flux reactors and is low enough to avoid a thermally activated diffusion or decomposition of the metastable γ-phase of the U–Mo fuel particles. After up to 13 h of irradiation a final fluence of 1×10^{17} ions/cm^2 on the surface of the specimen was achieved in an area of approximately 2×2 mm^2. This ion (or fission fragment) density could be compared to a low or even medium burn-up. Fig. 2 shows the sample during irradiation under an angle of 30°. Since the energy of the iodine projectile is less than 1 MeV per nucleon, nuclear reactions (especially activation of the specimen) can be excluded.

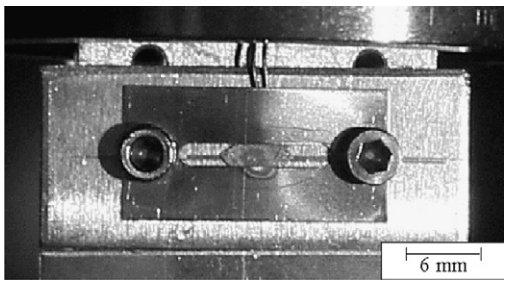

Fig. 2. Specimen – located in the slot between the two screws – during irradiation with heavy ions.

4. Post-irradiation examination

Since the specimens were not activated, post-irradiation examinations can be conducted much easier than for in-pile irradiated fuel plates. In particular no manipulations in hot cells is needed.

4.1. Optical microscopy

After irradiation an optical inspection of the specimen took place, which revealed first that the irradiated areas are very dark. The obtained optical images for U–6 wt%Mo/Al and U–10 wt%Mo/Al (two times for different fluence) specimen are shown in Fig. 3. They show that each spherical fuel particle is surrounded by a layer. This layer has a distinct border and a light blue color, presumably due to the deposition of iodine. Assuming a penetration depth of the iodine perpendicular to the surface of the specimen of 2 μm and a fluence of 10^{17} ions/cm^2, the iodine concentration would be as high as 5×10^{20} ions/cm^3. It means approximately one order of magnitude less than the atom density of the fuel. The blue color is not visible in the matrix (presumably due to the much higher penetration depth of the projectile) and not in the fuel since it becomes black due to oxidation in air. Independent of the particular irradiation conditions it has been observed that the thickness of the new phase depends on many parameters but is always in the range of some 10 μm. This observation fits to the interdiffusion layer, which has been observed after in-pile irradiation [3]. Thermal diffusion can be excluded because only some μm away from the irradiated area, where the temperature is almost the same due to the high thermal conductivity of the Al-matrix ($\lambda \approx 230$ W/m/K), no interdiffusion layer is visible.

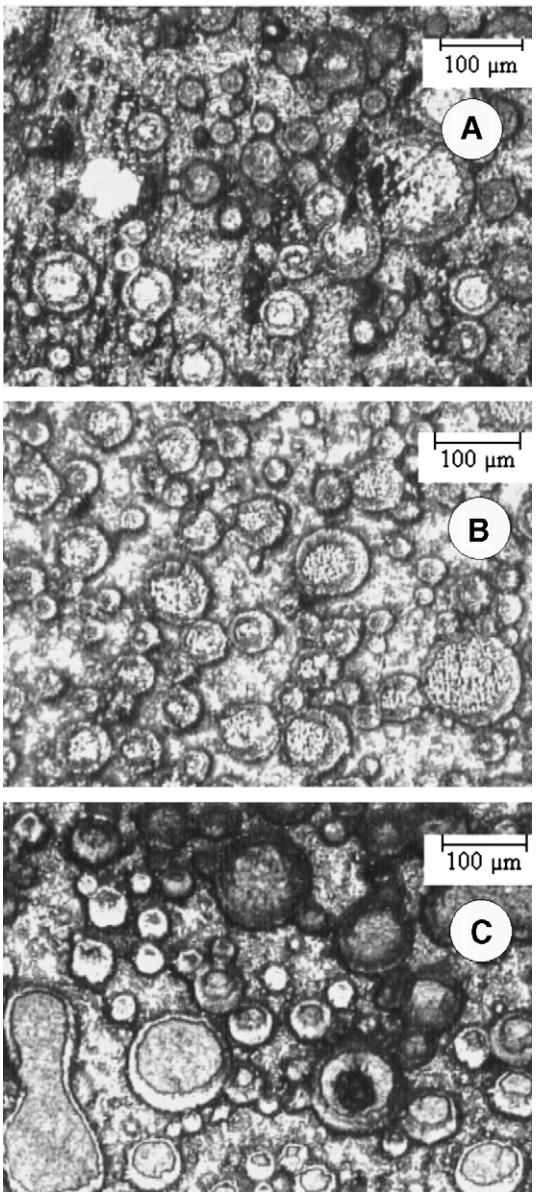

Fig. 3. Light-optical image of U–6 wt%Mo/Al (A) and U–10 wt%Mo/Al (B and C) specimens, irradiated to a fluence of 10^{17} ions/cm^2 (A and C) and 5×10^{16} ions/cm^2 (B).

4.2. Scanning electron microscopy

Scanning electron microscope images were taken at a Hitachi model s-4000 field emission scanning electron microscope, where the accelerating voltage was set to 20 keV. Specimen were mounted on a sample holder, which had a tilt of 45°.

A transition between an irradiated and a non-irradiated area is shown in Fig. 4. At first glance a significant change due to the irradiation is visible like in the optical images.

Fig. 5 shows an irradiated U–Mo/Al specimen. The image shows – like the light-optical image – around each spherical U–Mo particle a new layer, which is particularly pronounced in the direction of the irradiation and has a thickness of around 10 μm. While its thickness depends on the direction of irradiation, it seems to be independent of the fuel particle diameter. The latter is in agreement with the observation at in-pile irradiated samples.

Also to mention, Fig. 3 (optical image; part C) shows a part of Fig. 4 (SEM image) of an U–10 wt%Mo/Al sample, just mirrored. Both show

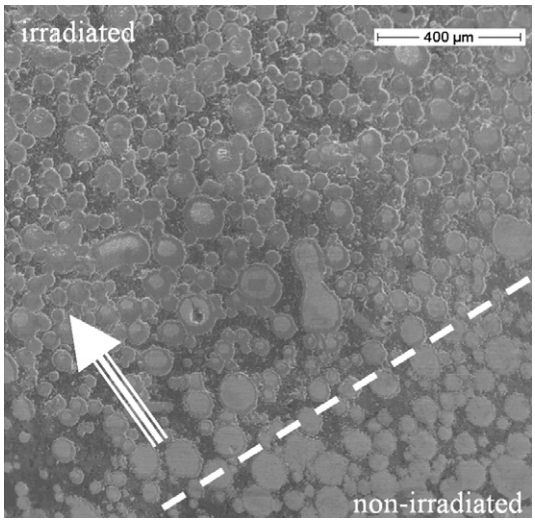

Fig. 4. SEM image of a U–10 wt%Mo/Al specimen, partly irradiated to a fluence of 10^{17} ions/cm^2, the arrow marks the direction of the incident heavy ion beam.

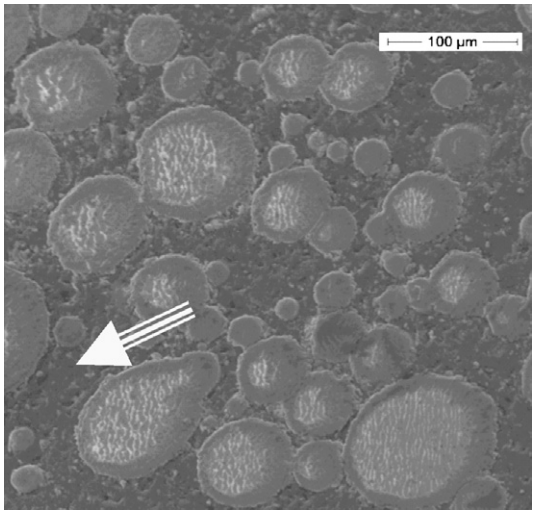

Fig. 5. SEM image of a U–6 wt%Mo/Al specimen, irradiated to a fluence of 10^{17} ions/cm^2.

similar dimensions of fuel, matrix and the new layer surrounding the fuel particles.

On the fuel particles themselves ripples have been formed, which are perpendicular to the incoming heavy ion beam [7]. They are a hint for surface amorphisation due to the irradiation by heavy ions. Ripples on a fuel particle are shown in detail in Fig. 6.

4.3. Energy dispersive X-ray analysis

A qualitative analysis of the element distribution by EDX (energy dispersive X-ray analysis) was performed on an EDR288, where the emitted X-ray radiation was analyzed by a Si(Li) detector. Fig. 6 (SEM image) shows irradiated fuel particles (U–6 wt%Mo) in an aluminium matrix. While the arrow marks the direction of the incoming heavy ion beam, the encircled numbers present the positions where EDX-measurements have been carried out. Results of these measurements are shown in Fig. 7. Measuring time for each EDX analysis was in the range of 200 s – dependent on the dead time of the detector and position at the specimen.

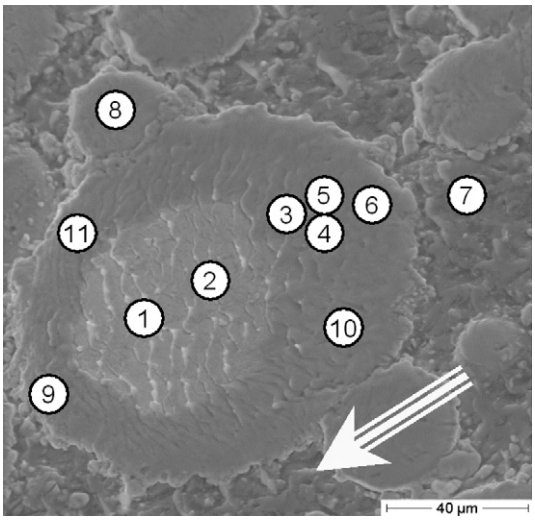

Fig. 6. SEM image of an irradiated U–10 wt%Mo fuel grain, the arrow indicates the direction of the heavy ion beam.

The EDX data show – beside the iodine projectile – a homogeneous distribution of uranium and molybdenum in the fuel, and pure aluminum in the matrix. In spite of the distinct borders of the new layer between the fuel and the matrix, there is a gradient of the elements. While the uranium and molybdenum concentrations decrease, the aluminum concentration increases in the direction from the fuel to the matrix. The new layer is also much thicker in the direction of the incoming heavy ion beam, but during in-pile irradiation the direction for fission fragments is the opposite way (from the fuel into the matrix), where the new layer is thinnest. Therefore a comparison with the interdiffusion layer of in-pile irradiated specimens has to be done with the smallest thickness value of this new layer.

4.4. X-ray powder diffraction

A first X-ray powder diffraction (XRD) measurement was performed with a STOE-STADIP

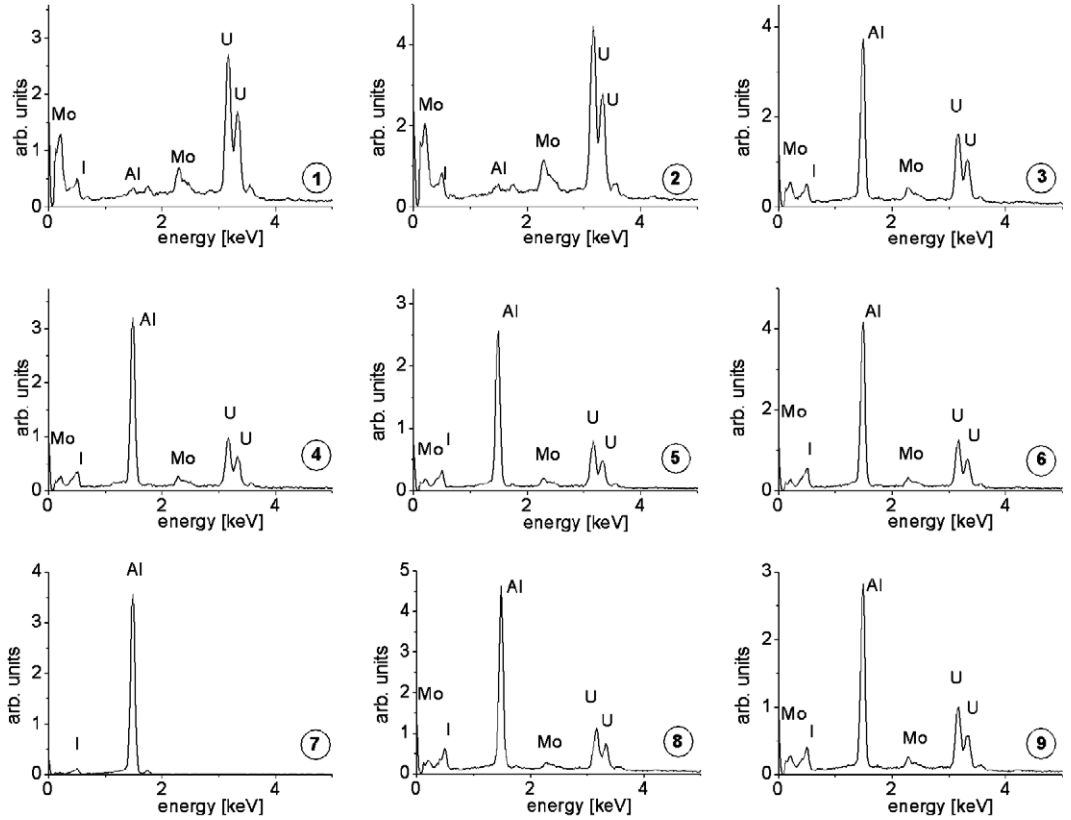

Fig. 7. EDX-measurement diagrams corresponding to Fig. 6.

diffractometer, where a curved, primary Ge(1 1 1) crystal monochromator produces strictly monochromatic Mo–$K_{\alpha 1}$-radiation ($\lambda = 0.07093$ nm, penetration depth into U–6 wt%Mo: 6.3 μm [6]). Three specimens (one non-irradiated and two irradiated with 120 MeV iodine to a fluence of 10^{17} ions per cm^2) were analyzed in reflection geometry and a linear position sensitive detector (acceptance 6°, channel width 0.02°) coupled in 1:2 mode was used for data acquisition. Up to 2700 data points in the angular range of 6°–60° were taken for each specimen in 24 h runs at an incoming beam diameter of approximately 1 mm. Hereby the specimen was rotated. Irradiated U–6 wt%Mo/Al and U–10 wt%Mo/Al specimens showed the presence of new binary phases like UAl_3 compared to the non-irradiated specimen. Qualitative phase analysis indicates the presence of up to five crystalline phases, whose reflection positions are marked in the lower part of Fig. 8. We have to point out that no decomposition of the metastable γ U–Mo phase was observed. Neither a ternary (Al, Mo, and U) nor binary compound of Al and Mo could be evidenced.

Quantitative multiphase Rietveld analysis yields the respective weight fractions of the specimen as shown in Table 1. Both irradiated specimens thus contain a significant portion of UAl_3. Further, UAl_4 is present. In the U–10 wt%Mo/Al specimen also UAl_2 exists. These compounds did not exist before the irradiation with heavy ions and therefore they ought to be products of the irradiation process.

5. Discussion

Post-irradiation examination has shown that a new layer around the spherical fuel particles is produced by irradiation with heavy ions and thereby simulating fission fragments. This new layer has similar properties like the one which was created by in-pile irradiation, compare [3].

Nevertheless, since this new layer consists of binary alloys like UAl_3 which are already used in reactor fuels, there is no hint of the abnormal swelling of the U–Mo/Al fuel. But during the heavy-ion irradiation one of the specimens (U–6 wt%Mo/Al) was broken into at least four pieces. Fig. 9 shows an

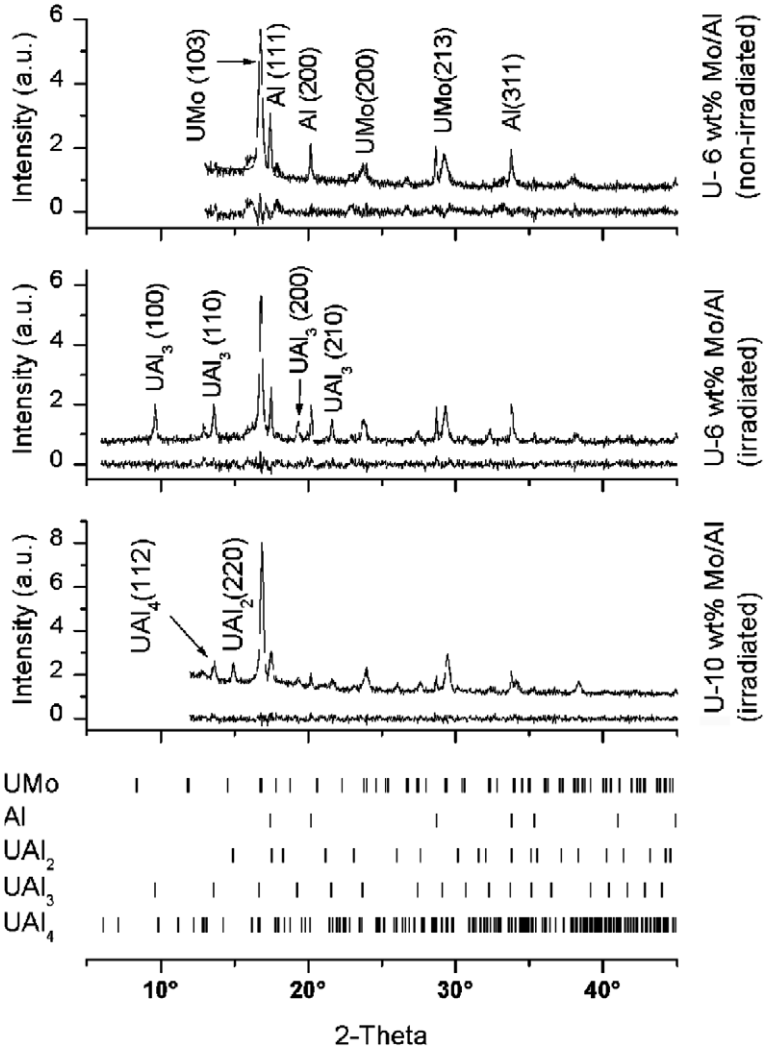

Fig. 8. X-ray powder diffraction measurement and multiphase Rietveld analysis of three specimen. The difference between observed and calculated data is depicted around the zero intensity line. Reflection positions of the identified phases are shown in the lower part.

Table 1
Phase abundance of the heavy ion irradiated specimen resulting from multiphase Rietveld analysis of X-ray powder data

Phase specimen	Al (wt%)	γUMo (wt%)	UAl_2 (wt%)	UAl_3 (wt%)	UAl_4 (wt%)
U–6 wt%Mo/Al	41.3 ± 2.3	23.5 ± 1.1	–	19.8 ± 1.0	15.4 ± 3.4
U–10 wt%Mo/Al	27.7 ± 1.2	36.3 ± 0.8	11.0 ± 0.4	13.4 ± 0.5	11.6 ± 1.8

light-optical microscope image of one part of this specimen. Like the other specimens, it was irradiated by iodine, but the energy of the projectile was reduced to 80 MeV and the flux increased to 2.3×10^{12} ions/s in order to reach higher deposition rates and to find the limits of the instrument, at which the specimen can be irradiated.

While a maximum temperature of less than 100 °C was measured – much lower than for the others due to a better water-cooled sample holder – the irradiated area was similar in size (0.9×3 mm^2). After only 14 min, which corresponds to a fluence of 7.2×10^{16} ion/cm^2, irradiation had to be stopped because the specimen was broken.

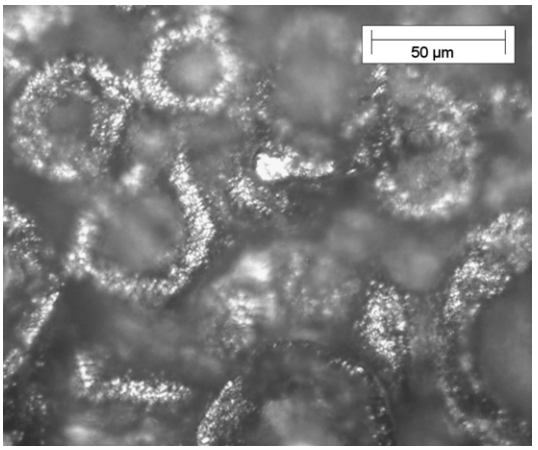

Fig. 9. Light-optical image of a destroyed specimen.

Optical microscopy of the broken parts revealed that there is a new protruding phase around each spherical fuel particle. These local elevations around the spherical fuel particles could be caused by either a volume increase of the new phase (interdiffusion layer), the fuel particles or a combination of both. As a reason for the swelling a transformation from crystalline to amorphous could be taken into account, because amorphous materials swell significantly under irradiation [8]. A first hint of such a transition was found in the XRD-pattern of the irradiated U–10 wt%Mo specimen which shows an amorphous hump under small angles (not shown in Fig. 8). Also the formation of ripples on the U–Mo particles indicates an amorphous surface. Such a transition from crystalline to amorphous was also discovered for U_3Si fuel for ion and in-pile irradiation [9]. A further examination of the broken U–Mo/Al specimen may be difficult since only small pieces are available for X-ray diffraction measurements which could prove an amorphous phase. This model could show the reason for the abnormal swelling of the in-pile irradiated fuel plates.

6. Summary

In this paper we showed the similarity or even equivalence of the interdiffusion layer of U–Mo/Al fuel, that is found in-pile and alternatively generated by heavy ion irradiation. As well a similar failure under irradiation has been reported. Since the resistance against swift heavy ions should be a basic requirement for nuclear fuels, this new approach can help to measure and to understand properties of the unfavorable interdiffusion layer – facilitating the development of U–Mo fuel considerably.

Acknowledgements

We would like to thank the RERTR-Team at ANL (USA) for providing the specimen and the French U–Mo Group for much technical aid. Especially we would like to thank G. Hofman (Argonne National Laboratory, USA), C. Jarousse (CERCA, France) and J. Noirot (CEA-Cadarache, France) for fruitful discussions. And we thank A. Lümkemann (TU-München, Germany) for the support at the SEM. We also thank the operator team of the Munich tandem accelerator laboratory and the support from the Maier-Leibnitz Laboratorium of LMU and TUM.

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