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Kr implantation into heavy ion irradiated monolithic U–Mo/Al systems: SIMS and SEM investigations

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HIGHLIGHTS

- Uranium–Molybdenum alloys were PVD sputtered on an Al substrate.
- The samples were exposed to both Iodine (at 80 MeV) and Krypton (at 45 MeV) irradiation.
- Secondary ion mass spectrometry (SIMS) and scanning electron microscopy (SEM) was performed to study the microstructure.
- A Kr accumulation in large μm-sized bubbles was observed.
- I and Kr irradiation offer an time- and cost-efficient method for the U–Mo development process.

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ABSTRACT

Worldwide, high performance research and material test reactors are aiming to convert their fuel from high enriched uranium towards low enriched ones. High density U–Mo/Al based nuclear fuels are considered as a promising candidate for this conversion. However, during in-pile test irradiations, the formation of an interdiffusion layer (IDL) between the U–Mo and the Al matrix is observed, caused by irradiation enhanced U–Al interdiffusion processes. This IDL accumulates fission gases at the IDL/matrix interfaces. Together, these two effects strongly reduce the performance of this new fuel type. Recently, the out-of-pile technique of heavy ion irradiation (^{127}I) on U–Mo/Al layer systems proved to be an alternative to time-consuming in-pile test irradiations for certain fuel behaviour aspects. Here we present SIMS and SEM investigations of non-conventional ^{82}Kr implantation into previously heavy ion irradiated U–Mo/Al layer systems. It is shown that Kr accumulates inside μm large porosities at the IDL/matrix interfaces. This critical accumulation of μm-sized large gas bubbles is directly related to the presence of the irradiation induced IDL. Without IDL no critical accumulation of fission gas bubbles occurs.

1. Introduction

In an international collaboration high flux research and test reactors are working actively to convert from fuels consisting of highly enriched uranium towards fuels with lower enrichment. As the scientific performance, i.e. the high neutron fluxes, of these reactors has to be preserved, a new kind of high density fuel is needed. One very promising candidate is an alloy of uranium-molybdenum (U–Mo with 7 to 10wt% Mo) [1]. This alloy can be incorporated in fuel assemblies either in powder form and successively dispersed in an Al matrix (dispersion fuel) [2,3], or as a massive bulk material (monolithic fuel) [4,5]. Both types are further encased by an Al cladding forming the resulting fuel plate.

Past test irradiations showed the growth of an interdiffusion layer (IDL) between the U–Mo and the surrounding Al matrix in case of dispersed powder [6,7], or between the U–Mo bulk material and the cladding in case of monolithic plates [4]. Scanning electron microscopy (SEM), neutron diffraction and transmission electron microscopy (TEM) of the IDL indicate its amorphous nature at irradiation temperatures below 200 °C [8–12]. Generated fission

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gases like Xe and Kr eventually accumulate at the IDL/Al interfaces in large bubbles with diameters of more than 1 μm [9,10], which might become the origin of plate cracking. Therefore this layer strongly limits these fuels' irradiation performance.

The understanding of the IDL growth behaviour and fission gas dynamics inside this layer is one major topic of U–Mo/Al fuel development. Recent studies have proven that heavy ion irradiation by ^{127}Iodine at 80 MeV on U–Mo/Al systems results in the formation of an IDL whose physical properties are similar to those obtained after in-pile irradiation, i.e. both elemental composition of U:Al and its amorphous nature are identical to the observations made after in-pile tests [13–15].

This work reports about the investigation of Kr implantation into U–Mo/IDL/Al layer systems previously generated by iodine irradiation. The Kr deposition was then analysed by scanning electron microscopy (SEM) and secondary ion mass spectrometry (SIMS) after the implantation step. It is to be verified whether this Kr implantation technique also results in the Kr accumulation inside large μm-sized porosities as observed after in-pile irradiation. The implantation of typical fission gases will help to elucidate the mechanism of gas accumulation at the IDL and thereby help to develop strategies to overcome critical gas accumulation.

2. Experimental methods

2.1. Sample manufacturing

All samples used for this study were created by physical vapour deposition of a 4 μm thick U-8wt%Mo layer on a 99.9995% pure and 500 μm thick Al substrate. The depleted U-8wt%Mo ingot was provided by AREVA-CERCA.

2.2. Heavy ion irradiation

Heavy ion irradiation was performed at the tandem accelerator of the Maier-Leibnitz Acceleration Laboratory (MLL) with ^{127}Iodine at 80 MeV with a particle flux of $7.0 \cdot 10^{11}$ particles/s$\cdot$cm^2 and an irradiation temperature of 140 ± 2 °C. The beam had a perpendicular incident vector on the sample surface with an according footprint of 6 mm in diameter. After 20 h of irradiation, a fluence of $1.0 \cdot 10^{17}$ particles/cm^2 was reached. According Monte Carlo calculations of the stopping range of ions in matter (SRIM [17]) under these irradiation conditions indicate two iodine stopping peaks (see Fig. 1A). The first one with an iodine concentration of $3.9 \cdot 10^{20}$ atoms/cm^3 is located at the U–Mo/Al interface, while the second one with a peak concentration of $5.1 \cdot 10^{20}$ atoms/cm^3 is found at a depth of 7 μm inside the Al substrate.

This iodine fluence has been previously calculated to be equivalent to an in-pile burnup of around 10% which corresponds to $4 \bullet 10^{20}$ f/cm^3 [16]. Nano-X-ray diffraction and transmission electron microscopy measurements were performed on identically treated samples [15]. There it has been demonstrated that these irradiation conditions result in the growth of a 1 μm thick interaction layer between the U–Mo and the Al substrate which is fully amorphous.

2.3. Krypton irradiation

After the iodine irradiation step, the samples were stored under air. As a consequence, the growth of a 0.5 μm-sized UO_2 layer on top of the U–Mo layer is expected [15]. According transmission electron microscopy measurements revealed this layer's nanocrystalline property, while nano-X-ray diffraction indicated some slight over-oxidation towards U_4O_9 or U_3O_7 [15]. According SRIM calculations were undertaken for Kr as well. To indicate the energy needed for a maximum Kr concentration at the U–Mo/IDL interface, the target material was simulated as a four layer system. First, a 0.5 μm thick UO_2 layer on top of the 4 μm thick U–Mo layer has been modelled, followed by a 1 μm thick IDL and the Al bulk material (see Fig. 1B). The crystallographic UAl_3 phase was used to simulate the IDL, as this phase is most commonly found after previous heavy ion irradiation experiments with temperatures above 200 °C and particle fluxes of $2.3 \cdot 10^{12}$ atoms/s$\bullet$cm^2, i.e. higher than those applied in this study [13,14]. Considering these thicknesses, under perpendicular incidence, a Kr energy of 45 MeV is necessary to fulfil the peak stopping criteria of 0.7 μm before the U–Mo/IDL interface.[1]

The Kr irradiation experiment was carried out at the IRRSUD beamline at the Grand Accelerateur National d'Ions Lourds (GANIL) in Caen, France. After 40 h of irradiation at a particle flux of $1.8 \cdot 10^{10}$ particles/s cm^2 and an irradiation temperature of 141 ± 2 °C, a fluence of $(2.56 \pm 0.13) \bullet 10^{15}$ particles/cm^2 was reached. Considering this fluence, SRIM predicts a maximum Kr concentration of $2.68 \cdot 10^{19}$ atoms/cm^3 at the peak stopping position right before the U-MO/IDL interface (see Fig. 1B).

In total, 6 samples were irradiated during one beamtime. The Kr beam itself was swept so that the whole sample surface was homogeneously exposed, in contrast to the 6 mm beam diameter during the iodine irradiation (see Fig. 2A). As a result, the samples exhibit regions which were implanted with both I and Kr, and regions which were only exposed to Kr.

To compensate slight U–Mo layer thickness variations, some samples were covered with a 0.8 μm thin 99.9995% pure Al foil to adjust the beam energy accordingly and to guarantee a peak Kr deposition at the U–Mo/IDL interfaces for all samples.

2.4. Analysis methods

The results presented in the following were acquired on an Al foil covered sample. This sample is considered representative for the five other implanted samples. As indicated in Fig. 2A, this sample was cut in two pieces of the same size along the red dashed line. One half was embedded in resin to enable SEM cross-section examination. The other half was kept for successive SIMS measurements.

2.4.1. SEM

SEM images were acquired in both secondary electron mode (SE) and backscatter electron detection mode (BSE). Accordingly, Fig. 2B shows a SE image of the sample's transversal cross-section of an area irradiated with both iodine and krypton, while Fig. 2C shows a BSE image of an area which was not exposed to the iodine beam, but only to krypton.

2.4.2. SIMS

As SEM can only deliver sample surface information, SIMS was utilized to quantify the Kr amount inside the irradiated layer systems. According dynamic-SIMS (D-SIMS) measurements were performed at the “Luxembourg Institute of Science and Technology” (LIST) in Belvaux, Luxembourg. To achieve a depth resolution in the range of a few nanometers, a CAMECA SC-Ultra instrument was utilized with a Cs^+ beam at 4 kV and a current of 60 nA.

[1] Among the available Kr energies, 45 MeV enables the closest possible stopping peak to the U–Mo/Al interface.