Heavy-Ion Irradiation for Rapid Evaluation of In-Reactor Nuclear Fuel Behavior

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Abstract. Heavy-ion irradiation provides a powerful surrogate technique to study and quantify in-reactor fuel behavior under well-controlled laboratory conditions. This work presents results from uranium-molybdenum (UMo) fuel studies conducted at the Maier-Leibnitz Laboratory (MLL) of the Technical University of Munich, where energetic heavy ions are used to simulate fission fragment effects. By comparing ion-irradiation data with in-reactor results, a strong quantitative correlation is demonstrated for the growth kinetics of the interdiffusion layer (IDL) between UMo and Al, its temperature-dependent microstructural evolution, and the performance of diffusion barrier coatings. Furthermore, high-dose ion irradiation reproduces irradiation-induced restructuring in UMo, confirming the capability of the method to simulate high-burnup phenomena. These results highlight how accelerator-based nuclear physics techniques can be applied to materials research, directly reflecting the HIAS 2025 theme of *Nuclear Physics and its Applications*. Ongoing and future collaborations, including test irradiations at the Heavy Ion Accelerator Facility (HIAF) at the Australian National University, aim to expand this approach to new accelerator environments and strengthen its role in the qualification of next-generation research reactor fuels.

1 Introduction

The conversion of research reactors from highly enriched uranium (HEU) to low enriched uranium (LEU) fuels is a central objective of the international *Reduced Enrichment for Research and Test Reactors* (RERTR) program. This global non-proliferation effort requires the replacement of existing fuels with high-density LEU alternatives while maintaining reactor performance. Uranium-molybdenum (UMo) alloys, typically containing 7–10 wt.% Mo, have been identified as promising candidates for high-density LEU fuels due to their high uranium density and favorable irradiation behavior, which is largely attributed to the retention of the cubic γ-phase at moderate temperatures [1–3].

This work demonstrates how accelerator-based ion beam techniques can bridge nuclear physics and materials science by applying controlled irradiation to replicate reactor conditions. Heavy-ion accelerators enable fundamental studies of radiation-matter interactions that are directly relevant to the development of advanced nuclear fuels.

A key challenge in the deployment of UMo fuels, whether in dispersion or monolithic form, is their interaction with aluminum during irradiation. In dispersion fuels, fine UMo particles are embedded in an aluminum matrix, while in monolithic fuels, a solid UMo foil is bonded to aluminum cladding. In both systems, interdiffusion between UMo and Al during irradiation leads to the formation of an interaction or interdiffusion layer (IDL), composed mainly of UAl_x phases [4]. These phases are mechanically weak and prone to irradiation-induced swelling, which can promote plate failure if not controlled [5]. Mitigating this reaction is therefore critical to ensure the long-term stability of LEU fuels.

In-reactor irradiation experiments remain the most direct means of assessing fuel performance, yet they are costly, time-consuming, and limited by strongly coupled parameters such as temperature, fission rate, and burnup. Each reactor test typically spans several years and results in highly radioactive specimens that require complex post-irradiation handling. As a complement, accelerator-based heavy-ion irradiation offers a powerful and flexible alternative. By using energetic ions as surrogates for fission fragments, this method reproduces key in-reactor effects such as ballistic mixing, radiation-enhanced diffusion, and microstructural evolution, but at greatly accelerated timescales and without introducing additional radioactivity [6, 7].

A long-term research program has been established to apply accelerator-based heavy-ion irradiation to UMo fuels, enabling the study of radiation-enhanced diffusion, microstructural evolution, and coating performance under well-controlled laboratory conditions. Building on these developments, the present work summarizes recent experiments using energetic iodine and xenon ions to reproduce key in-reactor phenomena and to evaluate the effectiveness of various diffusion barrier materials.

The present paper summarizes selected results from these studies, focusing on the correlation between ion- and reactor-irradiation data on IDL growth kinetics, temperature-dependent microstructural evolution, diffu-

sion barrier effectiveness, and irradiation-induced restructuring in UMo. The outlook of this work extends toward new collaborative opportunities at international accelerator facilities, where similar techniques can further advance the application of nuclear physics to materials research.

2 Experimental Approach

Heavy-ion irradiation experiments were conducted at two accelerator facilities. Iodine irradiations were performed at the 14 MV tandem accelerator of the Maier-Leibnitz Laboratory (MLL) in Garching, Germany, while xenon irradiations were carried out at the Argonne Tandem Linac Accelerator System (ATLAS), USA. The complementary use of both facilities enabled cross-validation of results and, more importantly, allowed simulation of different classes of fission products: iodine ions represent solid fission fragments that readily form stable compounds within the fuel, whereas xenon ions represent gaseous fission products responsible for bubble formation and gas-driven swelling. Together, these experiments capture the combined effects of solid and gaseous fission products on UMo/Al fuel systems.

Both accelerators provided stable, high-current ion beams with precise control of energy, current, and target temperature, essential for reproducing reactor-irradiation conditions under well-defined laboratory settings. Iodine-127 and xenon-132 ions were chosen to simulate the kinetic energy range (70-100 MeV) of fission fragments. Ion fluxes ranged from 10^{12} to 10^{13} ions/cm^2/s, corresponding to effective fission-rate equivalents of 10^{14}-10^{15} fissions/cm^3/s, and total fluences reached up to 7×10^{17} ions/cm^2 [6, 7]. Target temperatures between 50 °C and 275 °C were applied to systematically study temperature-dependent irradiation effects. A static, approximately Gaussian beam profile was used to generate a controlled fluence gradient across the exposed area, enabling simultaneous investigation of regions subjected to different local doses and dose rates within a single irradiation.

Several model sample configurations were designed to address different aspects of UMo fuel behavior. UMo/Al bilayers were used to study interdiffusion kinetics and derive activation parameters for the IDL. Al/X/UMo trilayers (X = Mo, Zr, W) were employed to evaluate the effectiveness of metallic coatings as diffusion barriers. All samples were fabricated by physical vapor deposition (PVD) at TUM. Layer thicknesses were selected based on SRIM/TRIM simulations to position the peak damage region near the interface of interest.

Post-irradiation analysis combined mechanical preparation with complementary microscopic techniques. Samples were mounted in epoxy, cross-sectioned, and polished to expose the beam centre and peripheral regions of lower dose. The polished sections were examined by scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) to determine the thickness, composition, and morphology of the intermixing layer.

The IDL thickness was determined from backscattered-electron SEM images, supported by EDS line scans to identify the chemically intermixed region. The IDL boundaries were defined based on the contrast and compositional transition between the unreacted UMo and Al layers and the irradiation-induced intermixing zone. For each irradiation condition, thickness values were measured at multiple positions along the interface, and the reported values correspond to the mean thickness.

Electron backscatter diffraction (EBSD) was used to assess irradiation-induced microstructural modifications in UMo. Site-specific lamellae were further prepared by focused ion beam (FIB) milling for high-resolution transmission electron microscopy (TEM) of interface structure. SRIM/TRIM simulations provided ion-range and displacement profiles, enabling correlation of the local damage distribution with the observed interfacial evolution. However, it should be noted that SRIM/TRIM simulations are based on a binary collision approximation and do not account for dynamic processes such as defect recombination, diffusion, or phase formation during irradiation. In particular, time- and dose-dependent changes in material composition, such as the development of interdiffusion layers, are not captured. Furthermore, quantitative discrepancies in predicted damage levels (e.g. dpa) have been reported when compared to standard models and more advanced simulations. Therefore, SRIM/TRIM results provide an approximate description of irradiation damage and must be interpreted with care when compared to experimental observations [6, 8].

Although a layered geometry was used, the samples are representative of both monolithic and dispersion UMo fuels. The ion penetration depth is much smaller than the characteristic UMo particle size, so each irradiated region effectively reproduces the local UMo/Al interface environment within a single particle-matrix zone. The resulting interdiffusion and microstructural evolution are therefore relevant to both fuel types.

3 Results and Discussion

3.1 IDL growth kinetics

IDL between UMo and Al grows as a function of irradiation time, temperature, and fission rate. To compare ion- and reactor-irradiation conditions, the thickness evolution was evaluated using an empirical correlation for IDL growth derived from reactor-irradiation data by Kim and Hofman [5]:

$$Y_{\mathrm{IDL}}^2 = A\, \dot{f}^{0.5} \exp\!\left(-\frac{q}{T}\right) t\, f_{\mathrm{Mo}}, \tag{1}$$

where Y_{IDL} is the IDL thickness, $\dot{f}$ the fission rate (or equivalent fission-rate density), t the irradiation time, T the absolute temperature, A and q are empirical parameters, and f_{Mo} accounts for the Mo concentration in the fuel matrix.

For comparison with ion-beam mixing formalisms, the IDL thickness can be related to the classical mixing parameter describing irradiation-induced interfacial broadening. Under diffusion-like conditions, the square of the

IDL thickness scales with the product of diffusion coefficient and irradiation time. Consequently, the fourth power of the thickness scales with the square of this quantity. The fourth power of the IDL thickness is therefore used to represent the scaling behavior of irradiation-induced mixing in a form that enables direct comparison with established ion-beam mixing descriptions [9].

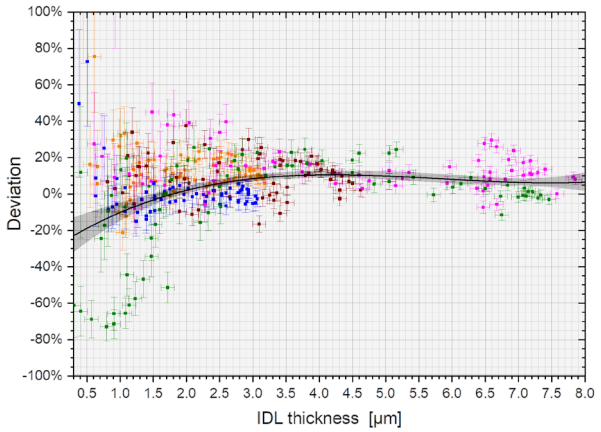

Figure 1. Deviation between measured and calculated IDL thickness using Eq. 1. The relative deviation is defined as $(Y_{\mathrm{calc}} - Y_{\mathrm{meas}})/Y_{\mathrm{meas}}$. Different colours represent datasets obtained under distinct irradiation conditions. The error bars indicate the standard deviation of IDL thickness measurements obtained from multiple positions along the interface, reflecting local variations in layer thickness and measurement uncertainty. The solid line is a cubic fit used only as a guide to the eye to visualize trends, and the shaded grey region represents the estimated uncertainty of the empirical model. Ion-irradiated data show good agreement with the reactor-based empirical model, confirming comparable IDL growth behavior.

Equation 1 was applied to predict the expected IDL growth under equivalent fission-rate conditions, using activation parameters derived from reactor-irradiation data. The calculated values were then compared with measured IDL thicknesses from ion-irradiated UMo/Al samples. As shown in Fig. 1, the predicted and measured data exhibit strong quantitative agreement, demonstrating that heavy-ion irradiation reliably reproduces the kinetics of IDL formation observed in in-reactor experiments.

The temperature dependence of IDL growth, summarized in Fig. 2, shows a clear transition near 180 °C. At lower temperatures, IDL growth is mainly driven by ballistic mixing, whereas above this threshold thermally activated radiation-enhanced diffusion dominates, resulting in a pronounced increase in growth rate. This transition marks a shift in the dominant atomic transport mechanism from ballistic to diffusion-controlled behavior. Comparable trends have been observed in reactor-irradiated UMo fuels and in Xe-ion irradiation experiments at ATLAS, where a similar transition temperature of approximately 170 °C was reported [10]. The agreement between solid- (I) and gaseous-fission-product (Xe) analogues demonstrates the robustness of the ion-beam approach in reproducing in-reactor diffusion kinetics.

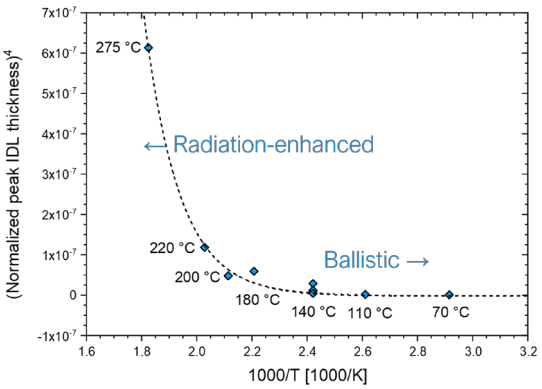

Figure 2. Temperature dependence of normalized IDL growth under 80 MeV I irradiation at MLL. The fourth power of the IDL thickness is used to represent the scaling behavior of irradiation-induced interfacial mixing and to enable direct comparison with ion-beam mixing formalisms, consistent with diffusion-like growth of the intermixing layer. A transition near 180 °C separates ballistic from diffusion-controlled growth regimes.

3.2 Microstructural and compositional evolution

The transition in growth kinetics described above is accompanied by corresponding changes in the microstructure and composition of the IDL. TEM analyses revealed a gradual structural transformation with increasing temperature. At 140 °C, the IDL remained amorphous and uniform. At 220 °C, nanosized crystallites of 10-20 nm emerged, and at 275 °C the layer became fully nanocrystalline. EDS and electron diffraction identified UAl_3 as the predominant reaction product. Complementary SEM/EDS and STEM/EDS measurements revealed a progressive Al enrichment with temperature, consistent with the structural transition from amorphous to crystalline IDLs. Compositional profiles further show that amorphous layers maintain near-stoichiometric U/Al ratios, whereas crystalline layers become increasingly Al-rich owing to enhanced solute mobility. These trends are consistent with post-irradiation examinations of reactor-irradiated UMo dispersion fuels, which likewise exhibited temperature-dependent thickening of the interaction layer and enrichment in Al-rich phases such as UAl_3 [12].

Quantitative EDS analyses confirm this compositional evolution. As shown in Fig. 4, the Al/(U+Mo) ratio increases with temperature from about 5 in the amorphous regime to nearly 8 in the crystalline regime. The consistent SEM/EDS and STEM/EDS results indicate that this Al enrichment arises from increasing non-stoichiometry of the IDL at elevated temperatures, which enables greater Al incorporation.

3.3 Performance of diffusion barriers

To mitigate UMo-Al interdiffusion, interlayers of Mo, Zr, and W were evaluated as diffusion barriers under ion irradiation. Figure 5 shows the total thickness of the irradiation-induced intermixing layers as a function of ion

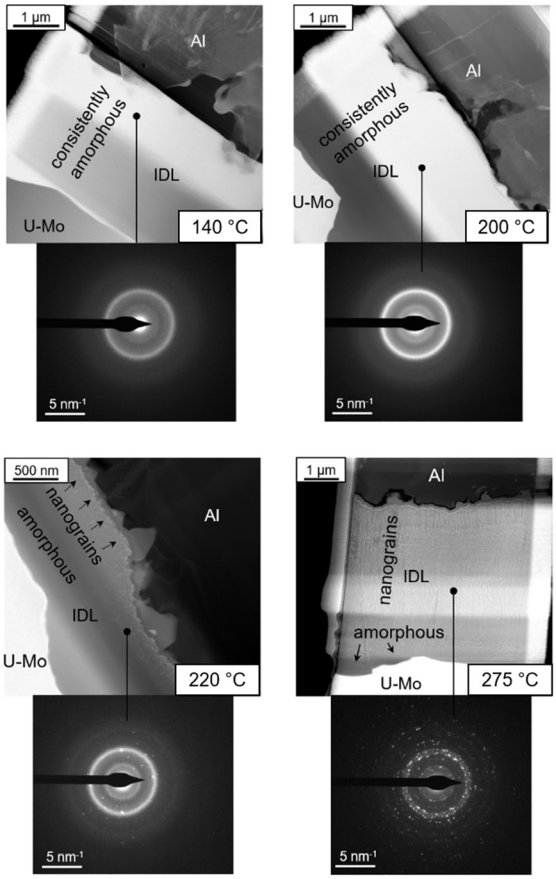

Figure 3. Evolution of IDL microstructure with temperature under 80 MeV I irradiation at MLL. TEM cross-sections and corresponding selected-area diffraction patterns show the transition from a fully amorphous structure at 140 °C to a nanocrystalline layer dominated by UAl_3 at 275 °C.

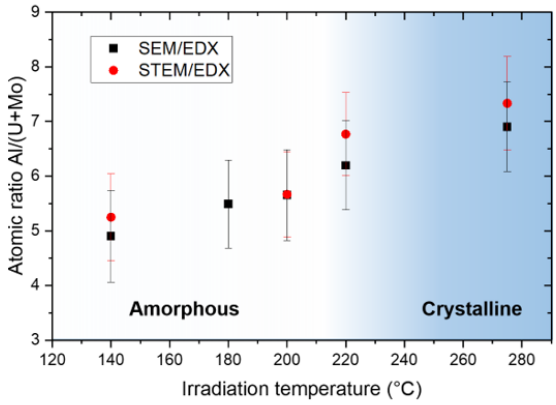

Figure 4. Al/(U+Mo) atomic ratio in the IDL as a function of irradiation temperature. Both SEM/EDS and STEM/EDS analyses show increasing Al enrichment with temperature, corresponding to the transition from amorphous to crystalline IDLs.

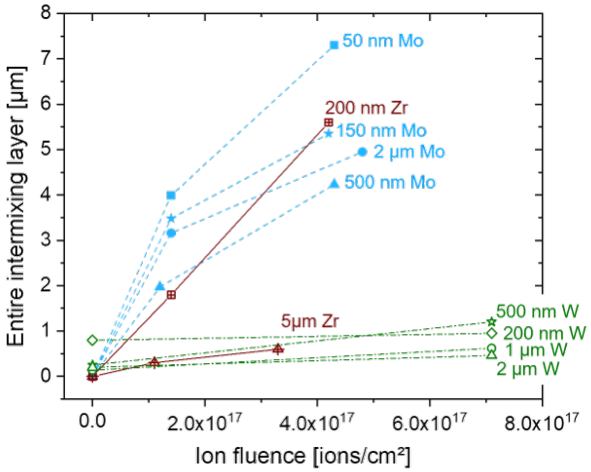

Figure 5. Thickness of the irradiation-induced intermixing layer as a function of ion fluence for Mo-, Zr-, and W-coated samples. W coatings exhibit the highest irradiation stability, while Mo and thin Zr coatings show pronounced intermixing under similar conditions.

fluence for different materials and coating thicknesses. Clear performance differences were observed.

Mo coatings exhibited extensive irradiation-enhanced intermixing with Al, as the intermixing layer thickened rapidly with fluence, suggesting that Al-Mo compound formation promotes continued atomic transport across the interface. Zr coatings showed intermediate behavior: thin layers (<200 nm) were readily consumed, while thicker ones (5 μm) offered improved resistance. W coatings demonstrated the highest stability, with negligible layer growth even at the highest fluences, owing to the lower thermodynamic driving force for Al-W compound formation compared with Al-Mo and Al-Zr systems.

These results highlight the importance of coating thickness and chemical stability in determining barrier performance. W coatings of 1 μm or greater effectively suppress UMo-Al interdiffusion, whereas thinner or more reactive coatings promote interfacial mixing. This result provides practical guidance for designing robust UMo fuel architectures.

3.4 Irradiation-induced restructuring of UMo

At high burnups, UMo fuels undergo microstructural restructuring in addition to IDL formation. Grains subdivide into subgrains and fission gas bubbles accumulate along new boundaries, contributing to swelling in high-burnup fuel [12]. To determine whether heavy-ion irradiation can reproduce this effect and identify its underlying mechanism, UMo samples were irradiated to increasing doses and analyzed by EBSD.

Figure 6 shows the progressive evolution of the UMo microstructure with ion fluence. At low doses, i.e. below the threshold for restructuring, the grains remain unchanged. With increasing fluence, subgrain networks develop, and at the highest doses the irradiated region be-

comes fully restructured, closely resembling the high-burnup structure observed in reactor-irradiated fuel.

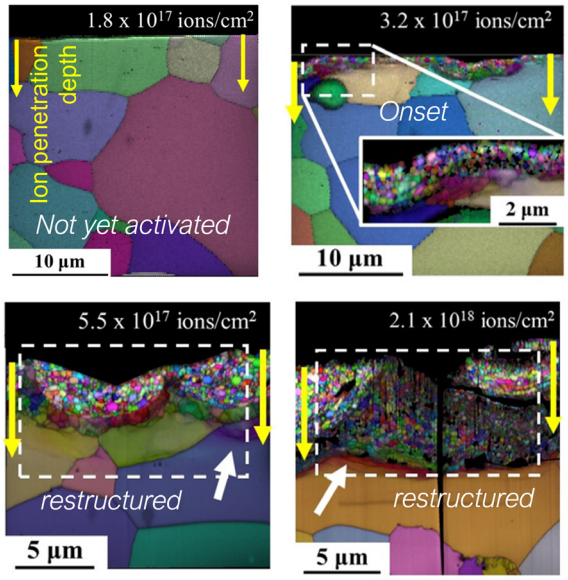

Figure 6. EBSD maps showing progressive restructuring of UMo with increasing ion fluence. The formation of subgrain boundaries and polygonized regions reproduces the high-burnup structure seen in reactor-irradiated fuels.

Misorientation analysis confirmed polygonization as the dominant mechanism, and the onset dose for restructuring under ion irradiation conditions was determined. These results demonstrate that heavy-ion irradiation can not only reproduce the microstructural evolution observed in-reactor but also quantify its progression under accelerated conditions.

4 Summary and Outlook

Heavy-ion irradiation experiments conducted in this work have established a reliable methodology for reproducing in-reactor phenomena under well-controlled laboratory conditions. Ion irradiation successfully replicated the growth kinetics of the UMo-Al interdiffusion layer, showing a clear transition behavior consistent with in-reactor observations. The temperature-dependent evolution from amorphous to nanocrystalline IDLs confirmed the shift from ballistic to diffusion-controlled growth. Diffusion barriers, particularly tungsten, effectively suppressed UMo-Al interdiffusion, providing valuable guidance for optimizing future UMo fuel architectures. At higher doses, UMo restructuring was also reproduced, demonstrating the capability of ion irradiation to quantify high-burnup effects under accelerated conditions.

Building on this foundation, new test irradiations are being initiated at the Heavy Ion Accelerator Facility (HIAF) at the Australian National University (ANU). Compared to the facilities used in the present work, HIAF will provide access to a broader range of ion species, beam energies, and flux conditions, enabling systematic studies beyond the constraints of MLL and ATLAS. In particular, future experiments will focus on decoupling temperature and dose-rate effects and assessing the reproducibility of IDL growth and microstructural evolution across different irradiation environments. In addition, more realistic material conditions will be considered, including alloying effects and manufacturing-related impurities. Finally, the insights gained from these studies will contribute to the development and qualification of advanced research reactor fuels.

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