Microstructural Changes and Chemical Analysis of Fission Products in Irradiated Uranium-7 wt.% Molybdenum Metallic Fuel Using Atom Probe Tomography

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Abstract: Understanding the microstructural and phase changes occurring during irradiation and their impact on metallic fuel behavior is integral to research and development of nuclear fuel programs. This paper reports systematic analysis of as-fabricated and irradiated low-enriched U-Mo (uranium-molybdenum metal alloy) fuel using atom probe tomography (APT). This study is carried out on U-7 wt.% Mo fuel particles coated with a ZrN layer contained within an Al matrix during irradiation. The dispersion fuel plates from which the fuel samples were extracted are irradiated at Belgian Nuclear Research Centre (SCK CEN) with burn-up of 52% and 66% in the framework of the SELENIUM (Surface Engineering of Low ENriched Uranium-Molybdenum) project. The APT studies on U-Mo particles from as-fabricated fuel plates enriched to 19.8% revealed predominantly γ-phase U-Mo, along with a network of the cell boundary decorated with α-U, γ'-U2Mo, and UC precipitates along the grain boundaries. The corresponding APT characterization of irradiated fuel samples showed formation of fission gas bubbles enriched with solid fission products. The intermediate burnup sample showed a uniform distribution of the typical bubble superlattice with a radius of 2 nm arranged in a regular lattice, while the high burnup sample showed a non-uniform distribution of bubbles in grain-refined regions. There was no evidence of remnant α-U, γ'-U2Mo, and UC phases in the irradiated U-7 wt.% Mo samples.

Keywords: atom probe tomography; nuclear fuel; microstructure; burnup; enrichment; uranium; fission products

1. Introduction

Over the last few decades, there has been purposeful effort to phase out the use of high-enriched uranium fuels used in research and test reactors using low-enriched uranium (LEU) fuel [1–6]. Maintaining reactor performance with the use of LEU dispersion fuels requires an increase in uranium (U) densities in the dispersed fuel particles. Although pure metallic U provides the highest U density, it exhibits anisotropic irradiation growth during irradiation [7], resulting in unacceptable performance relevant to irradiation behavior studies. Alloying U with molybdenum (Mo) results in a metastable body-centered cubic structure that exhibits stable swelling behavior and eliminates anisotropic growth. In particular, U-7 wt.% Mo [8–10] has been selected as a candidate alloy for dispersion fuel particles. The fuel particles studied here also incorporate a ZrN coating to limit the detrimental effect of the U-Mo interaction with the Al dispersion fuel matrix [11–15].

There have been several characterization studies carried out on U-Mo particles irradiated over a range of fission rates, temperatures, and fission densities [2,14,16–23]. As demonstrated in previous work using transmission electron microscopy (TEM) and
scanning electron microscopy (SEM), key features observed are the formation of a fission gas bubble superlattice along with decoration of grain boundaries by large fission gas bubbles [12,19]. The distribution of solid and volatile fission products species has not been well-characterized, however, because of the small scale of precipitates, they are typically assumed to form a dilute solid solutions fission products element with the fuel alloy. Atom probe tomography provides a methodology to characterize spatial localization and elemental composition of materials in three dimensions with near atomic-resolution [24–27]. This high-resolution information is achieved through ion-by-ion field evaporation from needle-shaped specimens (tip radius typically < 100 nm) subjected to high voltage or laser pulses. Moreover, the development of shielded focused ion beam (FIB) instruments presents unique opportunities for site-specific analysis from irradiated nuclear fuels using APT. This method is particularly relevant to understanding the behavior of fission gases, which drive swelling at high burnup, and may compromise fuel integrity [7,14,28,29].

Our previous APT work in isotopic quantification of uranium showed the value of this technique in analyzing local burnup [30]. In this work, APT is used to characterize the microstructural changes and chemical analysis on fission products formed in fuel during irradiation using a reference fresh fuel specimen and specimens irradiated to intermediate (52%) and high (66%) burnup. Characterization in this work is focused on detailed microstructural studies on fuel before irradiation followed by the distribution of fission gases and solid and volatile fission products through peak identification of the very complex chemistry of the irradiated fuels. APT was also employed to investigate the uniformity of enrichment in as-fabricated or fresh U-Mo samples, which is key for evaluation of the performance of fuel. Since APT has not often been applied to the characterization of irradiated fuel with high burnup, some challenges in this application are also discussed.

2. Materials and Methods

The SELENIUM (Surface Engineering of Low ENriched Uranium-Molybdenum) fuel development project of SCK•CEN was initiated to tackle the issue of fuel–matrix interaction layer formation by developing coatings on U-Mo particles. The fuel particles analyzed in this work consist of an 8 g/cm³ U-7 wt.% Mo (LEU) and were fabricated using rotating disk atomization by the Korea Atomic Energy Research Institute (KAERI), with a ~1 µm thick ZrN coating applied at SCK•CEN [3,15,22,28,31–33]. Using physical vapor deposition (PVD), the fuel particles were incorporated into a dispersion fuel plate with an aluminum matrix and aluminum alloy cladding (Plate ID: U7MD1231). The fuel plate was irradiated in the BR2 reactor at SCK•CEN over 3 cycles for 69 effective full-power days (EFPD). The maximum power at the beginning of life was close to approximately 470 W/cm². The fuel plate operated under a power gradient that allows a selection of samples with estimated burnup of ~52% 235U (~4.0 × 10²¹ fissions/cm³, sample M11) and 66% (5.0 × 10²¹ fissions/cm³, sample M14). Characterization by SEM and TEM have been previously published [12] under the framework of the SELENIUM project. Detailed characterization of the ZrN coating prior to and after irradiation is provided in [13].

The cutting and mechanical polishing of metallurgical specimens was performed at SCK•CEN. APT specimens were prepared using standard lift-out and milling procedures on a FEI Quanta Ga ion FIB at Idaho National Laboratory (INL). APT data collection was performed using a Cameca LEAP 4000X HR instrument operated in voltage pulsing mode, using a pulse fraction of 20%, a pulse repetition rate of 200 kHz, a data collection rate of 5 atoms per 1000 pulses, and a specimen temperature of 50 K at the Center for Advanced Energy Studies (CAES) facility. Atom probe tips were prepared from two different regions of the same fuel plate at local burnups of 52% and 66% to study the evolution of fission products. APT analysis was performed on all samples under identical running conditions to facilitate direct comparison of the data from each condition. The CAMECA Integrated Visualization and Analysis Software (IVAS) version 3.8.4 was used for data reconstruction. The values of the image compression factor and field factor ranged between 1.3 and 1.5
and between 3.6 and 3.8, respectively. High-resolution (80,000X) SEM images were used as a reference for reconstructing APT datasets in IVAS software.

3. Results

3.1. Phases Observed in U-Mo Prior to Irradiation

Understanding of the microstructure in terms of phases present, impurities, chemical composition of phases, and uniformity in enrichment is essential to understand the irradiation behavior of the fuel particles. Scanning electron microscopy (SEM) images shown in Figure 1 provide an overview of the fuel particles dispersed in the aluminum fuel plate matrix (Figure 1a) and the particle microstructure and coating (Figure 1b) showing lamellar-like features (Figure 1c) distributed non-uniformly across the sample.

![Figure 1. SEM images at different magnification of (a) as-fabricated U-7 wt.% Mo particles dispersed in Al matrix coated with ZrN, shown with dotted lines in (b,c).](image)

Previous analysis [34] of particles produced by the same atomization process indicates that the particles are composed primarily of the body-centered cubic $\gamma$-U-Mo phase. Tomograms from the current APT analysis for an analyzed volume of $50 \times 50 \times 5$ nm are shown in Figure 2a, along with the binomial distribution of $^{235}$U and Mo confirming uniform elemental distribution in Figure 2b. The measured composition inside the $\gamma$-U-Mo phase is shown in Table 1, which matched well with the expected composition of 15.8 at.% Mo (7 wt.% Mo). The $^{235}$U enrichment value of 19.8 measured by APT by the isotopic ratio method compares well with expected values, as shown in our previous work [30].

| Elements | at.%   | 2Sigma |
|----------|--------|--------|
| U        | 84.07  | 0.57   |
| Mo       | 15.83  | 0.20   |
| Fe       | 0.02   | 0.01   |
| N        | 0.01   | 0.01   |
| Cr       | 0.01   | 0.01   |
| C        | 0.01   | 0.01   |
| O        | 0.01   | 0.01   |
Figure 2. (a) 50 × 50 × 5 nm cuboid showing distribution of U, Mo, and 235U isotope. (b) Observed and binomial distribution of Mo and 235U isotope in analyzed volume obtained using APT from as-received U-7 wt.% Mo.

APT analysis was conducted from the specimen prepared from the lamellar microstructure-like features identified using electron microscopy shown in Figure 1c. The thickness of the lamellar microstructural features varied from 5 to 15 nm, as shown in Figure 3a,b, with the iso-concentration surface and the 1D plot taken across one of the lamellar-like structures indicated in Figure 3a. Concentration profiles indicate that these features contain nearly 100 at.% U, consistent with previous identification, as α-U while adjacent to these lamellar structures consists of Mo-enriched γ-U-Mo [35,36]. Note that these decompositions of γ-U-Mo into α-U did not affect enrichment levels, as expected.

Figure 3. (a) 3D distribution of network of α-U (grey color) shown with 100 at.% iso-concentration surfaces, (b) 1D profile taken across the lamellar structure by placing a cylinder (shown in (a)) across the interface. (blue: enrichment, red: Mo, grey: U).
Specimens prepared from the network of lamellar structures also revealed multiple interfaces in a single tip, as shown in Figure 4a–d. The cylinders highlighted on the left of the subfigures show where a 1D profile is taken along different interfaces. The analysis revealed that the presence of Mo enriched \( \gamma'\)-U-Mo, \( \alpha\)-U, \( \gamma'\)-U\(_2\)Mo, and grain/cell boundaries, showing segregation of Si. Note that APT analysis for these samples could not provide crystallographic orientation across the boundary, hence it is not possible to make a distinction between grain and cell boundaries. They have been demonstrated to almost always coincide in fresh fuel. The presence of a grain boundary in the analyzed volume suggests precipitation in the form of a lamellar-like microstructure consisting of different phases of U and Mo.

![Figure 4. 1D profile taken across different features observed in a single tip from corresponding cylinder. (a) Mo-enriched U-Mo and \( \alpha\)-U (b) and \( \alpha\)-U sandwiched between Mo-enriched \( \gamma'\)-U-Mo. (c) \( \gamma'\)-U\(_2\)Mo, \( \alpha\)-U, and Mo-enriched \( \gamma'\)-U-Mo, and (d) across the grain boundary, Mo-enriched \( \gamma'\)-U-Mo phase. (grey: U, red: Mo, green: Si).](image)

Specimens prepared from the grain boundary also revealed a non-uniform decoration by thin platelet-shaped U-C (carbides) phases, as shown in Figure 5a with an iso-concentration map of carbon (C). A 1D profile was taken across a grain boundary containing a carbide precipitate (Figure 5b), which shows clear enrichment of carbon, suggesting a U-C phase. Grain boundaries without U-C precipitates (shown in Figure 5c) show a slight depletion of Mo and a corresponding increase in U. Measurement of enrichment was carried out across the grain boundary to ensure that the presence of carbide phases does not affect enrichment in the fuel plate. The 1D profile indicates a uniform enrichment of 235U across the grain boundary with and without carbide precipitates. This specimen prepared from the grain boundary also showed the presence of \( \alpha\)-U phases adjacent to Mo-enriched \( \gamma'\)-U-Mo phase, reiterating previous findings.
3.2. Microstructure and Chemistry of U-Mo after Irradiation

Figure 6 provides a comparison of mass spectra obtained by APT for the three U-Mo samples (fresh, intermediate burnup—M11, and high burnup—M14). The spectra are divided into 9 segments over the range of 28 to 137 Daltons (Da) to ensure that comparison is feasible between all 3 samples. Background noise in all three cases was less than 30 ppm, which is key to ensure all peaks are accounted for in the quantification and for revealing the chemistry of the fission products labeled on top of all subfigures. Note that small traces of signal observed before 28 Da arising from H, C, N, O, OH, and OH2 are attributed to impurities arising from sample preparation and handling or from residual gases present in the analysis chamber. The combined contribution of these impurities was equal to or less than 0.3 at.% for each of the three samples. Peaks in the irradiated fuel spectra are assigned based on comparison to the mass spectrum to fresh U-Mo and by their relative isotopic abundance. For instance, signals appearing at 46, 47, 48, 49, and 50 Da were assigned as Mo\(^{2+}\) (shown in Figure 7b) since it matched the expected natural abundance. Additional peaks in the range of 44 to 50 Da were assigned to be fission products, as follows: 44 Da-88Sr\(^{2+}\), 44.5 Da-89Y\(^{2+}\), 45 Da-90Zr\(^{2+}\), 46–138Ba\(^{3+}\)/92Zr\(^{2+}\), 46.33 Da-139La\(^{3+}\), 46.5 Da-93Nb\(^{2+}\), 46.67 Da-140Ce\(^{3+}\), 47.33 Da-142Ce\(^{3+}\), and/or 147Nd\(^{3+}\) and 49.5 Da-99Tc\(^{2+}\) and/or 99Ru\(^{2+}\). A similar approach was followed for assigning chemical identity to all fission products for both irradiated U-Mo samples. Measured compositions of U, Mo (added as an alloy and fission product Mo), other fission products, and impurities from M11 and M14 samples are provided in Table 2 in at.%. The term impurities refer to C, N, Si, O, Cr, and Fe appearing in a sample due to the fabrication process and/or due to its introduction into the specimen during the milling process in FIB. 3D distribution of fission products from both irradiated samples is shown in Figure 7 by placing a cube of dimension 18 × 18 × 2 nm\(^3\) in an analyzed volume. It shows clear fission products’ enrichment into spherical clusters distributed uniformly over the analyzed volume for M11 (Figure 7a), whereas M14 (Figure 7b) showed a non-uniform distribution. Previous work with TEM already reported on the presence of a bubble superlattice structure with similar dimensions and also showed differences in distributions of fission products for both M11 and M14 samples [12]. Note that the analysis of fission products (size, shape, and spatial distribution) in APT is subject to local magnification artifacts [37]. This effect arises from differences in the evaporation field values for the different components present on the apex of the tip [38–40]. The actual location of U, Mo, and fission products can be discerned due to possible artifact overlap of trajectories of ions during the field evaporation process. The predominant presence of a local magnification effect is expected from the current system, in which clusters of fission products (radius of ~2 nm) are uniformly distributed in the U-Mo metallic matrix. Studies are planned to use advanced simulation methods to investigate
how field evaporation affects the size, shape, and spatial distribution of fuel components (U-Mo) and fission products (solids and gases).

Figure 6. Mass spectrum in the range of 28 to 137 Da from unirradiated U-7 wt.% Mo alloy (black, lower spectrum), M11 at 52% LEU burnup (red, center spectrum), and M14 at 66% LEU burnup (blue, top spectrum). All plots from 3 samples are normalized to 238U$^{3+}$ peak. Sub figures from (a–i) are from different part of mass spectrum. Different color codes identified above each sub figures are used to identify U, Mo, and individual fission products.
Figure 7. $18 \times 18 \times 2 \text{ nm}^3$ slice showing distribution of fission products observed from APT data in (a) M11 and (b) M14 samples, respectively. Dotted circles are used to highlight enriched segregation of fission products.

Table 2. Measured composition from M11 and M14 samples using APT (impurities: C, N, Si, and O).

| Elements       | M11  | M14  |
|----------------|------|------|
| U              | 74.22| 65.6 |
| Mo             | 19.80| 26.8 |
| Fission Products | 5.66 | 7.1  |
| Impurities     | 0.30 | 0.30 |

4. Discussion

4.1. Evolution of U-Mo Matrix

The as-received U-7 wt.% Mo sample showed the presence of $\gamma'$-$U_2Mo$, $\alpha$-$U$ phases, and Mo-alloyed $\gamma$-$U$-$Mo$ from the parent $\gamma$-$U$-$Mo$ matrix, along with presence of U-$C$ phase along the grain boundaries. It has been shown previously that discontinuous precipitation (DP) plays a major role during eutectoid transformation in U-10 wt.% Mo at 500 °C and higher [41]. DP results in the formation of a lamellar microstructure consisting of $\alpha$-$U$ and Mo-enriched bcc $\gamma$-$U$-$Mo$ phase along the prior $\gamma$-$U$-$Mo$ grain boundaries. Lower Mo contents, such as 7 wt.% in the present case, can also lead to a thermally induced phase decomposition from $\gamma$-$U$ to $\alpha$-$U$, $U_2Mo$, and Mo-alloyed $\gamma$-$U$ during fuel plate fabrication, which involves several heat treatments. Such behavior has already been reported on plates produced in the SELENIUM project [14,23,34]. However, after irradiation, the U-Mo sample at intermediate (M11) and high burnup (M14) did not show remnants of $\alpha$-$U$, $U_2Mo$, and UC phase. This has also been widely reported for a wide range of irradiated U-Mo fuels [17,23,33].

Irradiation promotes $\gamma$-$U$-$Mo$ because the fission spikes melt the material locally, after which it is quenched. This leads to stabilization of the high $\gamma$ phase independent of Mo content. Recrystallization is likely caused by the lattice pressure due to fission products and defects. The lower Mo content near grain/cell boundaries reduces the stability of that crystal structure, but even in heat-treated U-Mo, where no Mo gradient is present, recrystallization starts at the grain boundary.

4.1.1. M11 (Intermediate Burnup, 52%)

At the intermediate burnup level, it was shown in previous work that the fission gas bubbles are present on the cell boundaries and in the cells of U-Mo fuel [23,33]. The fracture surfaces at these burnups clearly show formation of small sub-grains near the cell boundaries. The microstructural changes taking place in U-Mo at the intermediate burnup
level are comparable to the low-dose sample \cite{23,33}. The presence of nano-sized ordered bubbles was reported based on TEM findings. However, APT allows to chemically identify the fission products, which is extremely challenging from EDS-TEM analysis due to the small size of the bubbles in comparison to the thickness of the lamella. Previous attempts to study the nature of fission products in the nanometric bubble superlattice could identify only Sr, Y, and Ba \cite{23,33}. APT analysis on the other hand revealed the presence of Zr, Y, Nb, Mo, Tc, Ru, Pd, La, Ce, Nd, Sr, Ag, Pr, Sm, Gd, Xe, Cs, and Ba for the intermediate burnup sample. Note that the possibility exists that overlap of peaks in the mass spectrum could mask the presence of other fission products. The mass spectrum also revealed a trace amount of Xe in the specimen prepared from the matrix region. It is well-known that the gaseous fission products are located on the cell boundaries and at the interface of the interaction layer and matrix due to its poor solubility in the U-Mo alloy \cite{23}. The relative contribution of fission products in the measured composition of the fuel at this burnup was close to 5.6 at.%, however, the local magnification effect can affect the absolute quantification of fission products due to the difference in the evaporation field of the U-Mo matrix and the bubble superlattice structures.

4.1.2. M14 (High Burnup, 66%)

Running the M14 specimen in APT was challenging due to a low survivability of tips consisting of numerous bubbles/voids, as shown in SEM images in Figure 8 with different magnification. In the current study, 2 out of 11 tips showed statistically significant data. Previous studies on high burnup structures using SEM and TEM have shown fully recrystallized fuel along with increased porosity, with no significant change in bubble size, at least up to full recrystallization \cite{23,33}. Fission products found in this specimen were predominantly solids, similar to the intermediate burnup specimen along with some trace amounts of Xe. The relative contribution of fission products in the analyzed volume for the M14 specimen was higher (~7 at.%) than in the M11 specimen (5.6 at.%), which is consistent with higher burnup.

![Figure 8](image_url)

**Figure 8.** (a) SEM images showing porosity in high burnup structure of the M14 sample. (b,c) Tip images at different stages of milling for APT analysis.

4.2. Evolution of Fission Products

For intermediate burnup (M11 sample 52%, $4.0 \times 10^{21}$ f/cm$^3$), TEM studies have shown the formation of a fission gas bubble superlattice with FCC structure coherent with the U(Mo) lattice BCC structure \cite{12,15,20,42}. However, it is very challenging to quantify the EDS signal from TEM due to the small size of the bubbles (<2 nm) embedded in thin foil lamella (50–100 nm). It was shown that the bubble lattice is present in all grains, and no lattice distortion was observed. Only near the grain boundaries was the bubble superlattice destroyed in this sample. APT data show that the bubble superlattice structure contains the following fission products: Y, Zr, Nb, Mo, Tc, Ru, Pd, Sr, La, Nb, Ce, Nd, Tc, Rh, Pd,
Ag, Pr, Sm, and Gd. Mass spectrum also indicated trace amounts (<0.001 at.%) of Xe (labeled at 131Xe\(^{1+}\), 132Xe\(^{1+}\), and 133Xe\(^{1+}\)), which also had overlap with one of the isotopes of Cs (133Cs\(^{1+}\)). All the detected fission products that could be labeled based on their mass-to-charge ratios were predominantly solid elements. Note that the fission products are labeled to the best of our ability, with potential issues of overlap of isotopes of different elements. For this reason, mass spectrum has been labeled with all possible options of elements wherever there is a possibility of overlap.

For the high burnup sample (M14, 66%, 5.0 × 10^{21} \text{ f/cm}^3), TEM studies have shown that the U(Mo) grains subdivided, reducing the size of the grains from a few micrometers to sub-micron-sized grains. Although the superlattice of fission gas bubbles was destroyed at most of the locations, there were some regions where the bubble superlattice remained or newly developed in the subdivided grains. APT on M14 revealed similar fission products as observed for the M11 sample, but with relatively higher concentrations. Both the M11 and M14 samples showed no clear evidence of Kr, although very low amounts of Xe were measured. No measurable Kr signal was detected in the mass spectrum for both samples, which could mean that Kr is too low (10 times lower production than Xe) and probably lower than the background noise limit of the APT instrument. Additionally, sample preparation using FIB could impact the location of Kr and Xe inside bubbles. Since APT is based on the field evaporation behavior of atoms, artifacts may hinder the detection of Xe and Kr from small bubbles due to differences in field evaporation with the surrounding matrix of U and Mo.

5. Conclusions

Systematic analysis of fresh and irradiated U-Mo dispersion fuel was carried out using APT. Specimens of different burnup were studied to elucidate microstructural changes taking place in U-Mo fuel due to irradiation. The as-received or as-fabricated U-Mo showed a network of \(\gamma'\)-U\(_2\)Mo, \(\alpha\)-U phases, and Mo-enriched \(\gamma\)-U-Mo from the parent \(\gamma\)-U-Mo matrix, along with the presence of U-C phase along the grain boundaries with uniform uranium enrichment across all phases. However, irradiation led to predominantly Mo-enriched \(\gamma\)-U-Mo phase with the presence of the bubble superlattice structures enriched with fission products. This work is the first of its kind in providing an in-depth chemical analysis on the localization of fission products formed in LEU metallic fuel irradiated to different burnups using APT. The local distribution of the bubble superlattice structures observed in APT complement previous TEM findings. The measured content of fission products increased with the increase in the burnup, as expected from fuel behavior. Mass spectrum from both samples showed that the bubbles are decorated with solid fission products along with some trace amounts of gaseous Xe elements. This study revealed the chemical identity of the fission products, which can provide fundamental insight on fission products’ formation in metallic fuels at high burnup.

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References

1. Snelgrove, J.; Hofman, G.; Meyer, M.; Trybus, C.; Wieneck, T. Development of very-high-density low-enriched-uranium fuels. Nucl. Eng. Des. 1997, 178, 119–126. [CrossRef]
2. Keiser, D.D.; Hayes, S.L.; Meyer, M.K.; Clark, C.R. High-density, low-enriched uranium fuel for nuclear research reactors. JOM 2003, 55, 55–58. [CrossRef]
3. Berghe, S.V.D.; Leenaers, A.; Koonen, E.; Sannen, L. From High to Low Enriched Uranium Fuel in Research Reactors. Adv. Sci. Technol. 2010, 73, 78–90. [CrossRef]
4. Wachs, D.M.; Clark, C.R.; Dunavant, R.J. Conceptual Process Description for the Manufacture of Low-Enriched Uranium-Molybdenum Fuel; No. INL/EXT-08-13840; Idaho National Laboratory (INL): Idaho Falls, ID, USA, 2008.
5. Cahn, R.; Haasen, P.; Kramer, E.J.; Frost, B.R. Materials Science and Technology: A Comprehensive Treatment. Int. J. Mater. Res. 1993, 84, 90. [CrossRef]
6. US Nuclear Regulatory Commission. Limiting the Use of Highly Enriched Uranium in Domestically Licensed Research and Test Reactors. Fed. Regist. 1986, 51, 6514.
7. Harrison, J. The growth of gas bubbles in a stressed medium and the application to stress enhanced swelling in alpha uranium. J. Nucl. Mater. 1967, 23, 139–153. [CrossRef]
8. Collette, R.; King, J.; Buesch, C.; Keiser, D.; Williams, W.; Miller, B.; Schulthess, J. Analysis of irradiated U-7wt%Mo dispersion fuel microstructures using automated image processing. J. Nucl. Mater. 2016, 475, 94–104. [CrossRef]
9. Oh, S.J.; Kim, K.H.; Jang, S.J.; Lee, D.B.; Lee, Y.S.; Park, J.M.; Park, H.D.; Kim, C.K. The Development of U-7W%Mo for Large Particle Powders by Centrifugal Atomization; International Atomic Energy Agency (IAEA): Vienna, Austria, 2006; pp. 126–130.
10. Mirandou, M.; Balart, S.; Ortiz, M.; Granovsky, M. Characterization of the reaction layer in U-7wt% Mo/Al diffusion couples. J. Nucl. Mater. 2003, 323, 29–35. [CrossRef]
11. Jungwirth, R.; Zweifel, T.; Chiang, H.-Y.; Petry, W.; Berghe, S.V.D.; Leenaers, A. Heavy ion irradiation of UMo/Al samples PVD coated with Si and ZrN layers. J. Nucl. Mater. 2013, 434, 296–302. [CrossRef]
12. Van Renterghem, W.; Miller, B.; Leenaers, A.; Berghe, S.V.D.; Gan, J.; Madden, J.; Keiser, D. Transmission electron microscopy investigation of neutron irradiated Si and ZrN coated UMo particles prepared using FIB. J. Nucl. Mater. 2018, 498, 60–70. [CrossRef]
13. He, L.; Bachhav, M.; Keiser, D.; Madden, J.; Perez, E.; Miller, B.; Gan, J.; Van Renterghem, W.; Leenaers, A.; Berghe, S.V.D. STEM-EDS/EELS and APT characterization of ZrN coatings on UMo fuel kernels. J. Nucl. Mater. 2018, 517, 174–182. [CrossRef]
14. Leenaers, A.; Berghe, S.V.D.; Koonen, E.; Kuzminov, V.; Detavernier, C. Fuel swelling and interaction layer formation in the SELENIUM Si and ZrN coated U(Mo) dispersion fuel plates irradiated at high power in BR2. J. Nucl. Mater. 2015, 458, 380–393. [CrossRef]
15. Van Renterghem, W.; Leenaers, A.; Van den Berghe, S.; Miller, B.; Gan, J.; Madden, J.; Keiser, D.; Palancher, H.; Hofman, G.; Breitkreuz, H. Post Irradiation TEM Investigation of ZrN Coated U (Mo) Particles Prepared with FIB; Idaho National Lab (INL): Idaho Falls, ID, USA, 2015.
16. Gan, J.; Keiser, D.; Miller, B.; Robinson, A.; Wachs, D.; Meyer, M. Thermal stability of fission gas bubble superlattice in irradiated U-10Mo fuel. J. Nucl. Mater. 2015, 464, 1–5. [CrossRef]
17. Gan, J.; Miller, B.; Rosenblum, A.; Wachs, D.; Meyer, M. Microstructural characterization of irradiated U-7Mo/Al-5Si dispersion fuel to high fission density. J. Nucl. Mater. 2014, 434–445. [CrossRef]
18. Keiser, D.D.; Perez, E.; Wieneck, T.; Leenaers, A.; Berghe, S.V.D. Microstructural characterization of a thin film ZrN diffusion barrier in an As-fabricated U-7Mo/Al matrix dispersion fuel plate. J. Nucl. Mater. 2015, 458, 406–418. [CrossRef]
19. Miller, B.; Gan, J.; Keiser, D.; Robinson, A.; Jue, J.; Madden, J.; Medvedev, P. Transmission electron microscopy characterization of the fission gas bubble superlattice in irradiated U–7 wt%Mo dispersion fuels. J. Nucl. Mater. 2015, 458, 115–121. [CrossRef]
20. Meyer, M.; Gan, J.; Jue, J.; Keiser, D.; Perez, E.; Robinson, A.; Wachs, D.; Woolstenhulme, N.; Hofman, G.; Kim, Y. Irradiation performance of U-Mo monolithic fuel. Nucl. Eng. Technol. 2014, 46, 169–182. [CrossRef]
21. Meyer, M.; Hofman, G.; Hayes, S.; Clark, C.; Wieneck, J.; Snelgrove, J.; Strain, R.; Kim, K.H. Low-temperature irradiation behavior of uranium–molybdenum alloy dispersion fuel. J. Nucl. Mater. 2002, 304, 221–236. [CrossRef]
22. Leenaers, A.; Berghe, S.V.D.; Detavernier, C. Surface engineering of low enriched uranium–molybdenum. J. Nucl. Mater. 2013, 440, 220–228. [CrossRef]
23. Leenaers, A.; Van Renterghem, W.; Berghe, S.V.D. High burn-up structure of U(Mo) dispersion fuel. J. Nucl. Mater. 2016, 476, 218–230. [CrossRef]
24. Gault, B.; Moody, M.P.; Cairney, J.M.; Ringer, S.P. Atom Probe Microscopy; Springer Science and Business Media LLC: New York, NY, USA, 2012.
25. Kelly, T.F.; Miller, M.K. Atom probe tomography. Rev. Sci. Instrum. 2007, 78, 031101. [CrossRef]
26. Lefebvre, W.; Vurpillot, F.; Sauvage, X. Atom Probe Tomography: Put Theory into Practice; Academic Press: Cambridge, MA, USA, 2016.
27. Saxey, D.; Moser, D.; Piazolo, S.; Reddy, S.; Valley, J. Atomic worlds: Current state and future of atom probe tomography in geoscience. *Scr. Mater.* 2018, 148, 115–121. [CrossRef]

28. Berghe, S.V.D.; Parthoens, Y.; Cornelis, G.; Leenaers, A.; Koonen, E.; Kuzminov, V.; Detavernier, C. Swelling of U(Mo) dispersion fuel under irradiation—Non-destructive analyses of the SELENIUM plates. *J. Nucl. Mater.* 2013, 442, 60–68. [CrossRef]

29. Kim, Y.S.; Hofman, G. Fission product induced swelling of U–Mo alloy fuel. *J. Nucl. Mater.* 2011, 419, 291–301. [CrossRef]

30. Bachhav, M.; Gan, J.; Keiser, D.; Giglio, J.; Jädermås, D.; Leenaers, A.; Berghe, S.V.D. A novel approach to determine the local burnup in irradiated fuels using Atom Probe Tomography (APT). *J. Nucl. Mater.* 2020, 528, 151853. [CrossRef]

31. Van den Berghe, S.; Leenaers, A.; Detavernier, C. SELENIUM Fuel: Surface Engineering of U (Mo) Particles to Optimise Fuel Performance; European Nuclear Society: Brussels, Belgium, 2010.

32. Berghe, S.V.D.; Lemoine, P. Review of 15 years of high-density low-enriched UMo dispersion fuel development for research reactors in Europe. *Nucl. Eng. Technol.* 2014, 46, 125–146. [CrossRef]

33. Berghe, S.V.D.; Van Renterghem, W.; Leenaers, A. Transmission electron microscopy investigation of irradiated U–7wt%Mo dispersion fuel. *J. Nucl. Mater.* 2008, 375, 340–346. [CrossRef]

34. Leenaers, A. Surface-Engineered Low-Enriched Uranium-Molybdenum Fuel for Research Reactors. Ph.D. Thesis, University of Ghent, Ghent, Belgium, 2014.

35. Leenaers, A.; Berghe, S.V.D.; Koonen, E.; Jarousse, C.; Huet, F.; Trotabas, M.; Boyard, M.; Guillot, S.; Sannen, L.; Verwerft, M. Post-irradiation examination of uranium–7wt% molybdenum atomized dispersion fuel. *J. Nucl. Mater.* 2004, 335, 39–47. [CrossRef]

36. Iltis, X.; Zacharie-Aubrun, I.; Ryu, H.; Park, J.; Leenaers, A.; Yacout, A.; Keiser, D.; Vanni, F.; Stepnik, B.; Blay, T.; et al. Microstructure of as atomized and annealed U-Mo7 particles: A SEM/EBSD study of grain growth. *J. Nucl. Mater.* 2017, 495, 249–266. [CrossRef]

37. Miller, M.; Hetherington, M. Local magnification effects in the atom probe. *Surf. Sci.* 1991, 246, 442–449. [CrossRef]

38. De Geuser, F.; Lefebvre, W.; Danoix, F.; Vurpillot, F.; Forbord, B.; Blavette, D. An improved reconstruction procedure for the correction of local magnification effects in three-dimensional atom-probe. Surface and Interface Analysis: An International Journal devoted to the development and application of techniques for the analysis of surfaces. *Interfaces Thin Film.* 2007, 39, 268–272.

39. Vurpillot, F.; Bostel, A.; Blavette, D. Trajectory overlaps and local magnification in three-dimensional atom probe. *Appl. Phys. Lett.* 2000, 76, 3127–3129. [CrossRef]

40. Wang, X.; Hatzoglou, C.; Sneed, B.; Fan, Z.; Guo, W.; Jin, K.; Chen, D.; Bei, H.; Wang, Y.; Weber, W.J.; et al. Interpreting nanovoids in atom probe tomography data for accurate local compositional measurements. *Nat. Commun.* 2020, 11, 1–11. [CrossRef] [PubMed]

41. Jana, S.; Overman, N.; Devaraj, A.; Sweet, L.; Lavender, C.; Joshi, V. Discontinuous Precipitation in U-10 wt.%Mo Alloy: Reaction Kinetics, Effect of Prior γ-Um Microstructure, the Role of Grain-Boundary Misorientation, and the Effect of Ternary Alloying Addition. *JOM* 2019, 71, 2770–2779. [CrossRef]

42. Miller, B.D.; Keiser, D., Jr.; Abir, M.; Aitkaliyeva, A.; Leenaers, A.; Hernandez, B.J.; Van Renterghem, W.; Winston, A. 3D reconstructions of irradiated UMo fuel to understand breaching effects in ZrN diffusion barriers. *J. Nucl. Mater.* 2018, 510, 431–436. [CrossRef]