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High-Energy Synchrotron Study of In-Pile-Irradiated U-Mo Fuels

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ABSTRACT

A coordinated wide-angle and small-angle synchrotron X-ray scattering investigation was performed on in-pile-irradiated U-7wt%Mo fuel specimens at the Advanced Photon Source (APS) at Argonne National Laboratory (ANL). U-7wt%Mo samples of three different burnup conditions were studied in comparison with an unirradiated control sample. Well-developed fission gas bubble superlattice was observed to form at intermediate burnup (3.0×10^{21} fission/cm^3) according to the appearance of satellite diffraction peaks. The superlattice constant was determined to be 11.7 nm and 12.1 nm by wide-angle and small-angle scattering, respectively. Grain sub-division takes place in all three irradiated samples and causes the collapse of the superlattice at high burnup. The bubble superlattice expands the lattice constant and acts as strong sink of radiation induced defects. The evolution of dislocation loops was therefore suppressed until the bubble superlattice collapses. The lattice expansion due to the irradiation was also studied by quantifying the shift of X-ray diffraction peaks.

1. Introduction

Minimizing the risk of nuclear proliferation by replacing high-enriched uranium (HEU) with low-enriched uranium (LEU) is a major task of the US National Nuclear Security Administration (NNSA) program [1]. In order to develop practical nuclear fuel materials with high uranium density, a series of alloys and compounds containing uranium have been examined [2]. With good in-pile fuel performance, U-Mo binary alloy system has become the focus of LEU studies [3-6] for conversion of high power research reactors. Both monolithic and dispersed forms of U-Mo fuel plates have been developed and tested [7,8]. In a dispersed U-Mo fuel plate, the fuel meat U-Mo particles are embedded in Al matrix, and clad by Al alloy.

In nuclear fuel materials, gaseous fission products such as Xe cause gas swelling, altering multiple key factors that determine fuel performance [9]. Therefore, fission gas behavior is regarded as an indispensable part of the post-irradiation examination (PIE) of fuel materials. In this study, three dispersed U-7wt%Mo fuel samples irradiated in the Advanced Test Reactor (ATR) to different burnups were investigated by synchrotron X-ray scattering techniques to examine the microstructural modifications induced by neutron irradiation. Previous transmission electron microscopy (TEM) observations on the same batch of samples confirm the formation of well-developed face-centered cubic (FCC) bubble superlattice at intermediate burnup[10-13]. The superlattice was also found to collapse at high burnup. Synchrotron techniques provide the opportunity of investigating more grains and quantifying internal strain, and therefore help provide a comprehensive understanding of the microstructural responses of U-7wt%Mo dispersion fuel at different burnup levels.

2. Experiments

Three U-7wt%Mo specimens were examined in this study along with an unirradiated control specimen, as shown in Table 1. An approximately 30 μm×30 μm×30 μm cube was prepared from a U-Mo particle within each bulk sample by focused ion beam (FIB) and then mounted on a W tip. The samples were then investigated at Sector 1-ID, Advanced Photon Source (APS), Argonne National Laboratory (ANL). As illustrated in Figure 1, the samples were illuminated by a 10 μm×10 μm 71.676 keV synchrotron X-ray beam. Both wide-angle and small-angle scattering (W/SAXS) signals were collected by respective detectors.

Table 1: List of specimens investigated in this study

Sample Index	O	A	B	C
Fuel plate ID	n/a	R3R050 (low)	R3R050 (high)	R2R040
Burnup ($\times10^{21}$ f/cm^3)	0.0	3.0	5.2	6.3

The shifts of the diffraction peaks were used to measure the lattice expansion of U-7wt%Mo, while the portions of the integrated peak intensities contributed by large-sized grains were utilized to assess the volume fraction of the recrystallization. Meanwhile, the broadening of the diffraction peaks contain the information about the internal strain, and therefore interpreted by both original and modified Williamson-Hall analyses (W-H)[14]. Also analyzed were the SAXS data of those irradiated and control samples by means of standard Guinier approximation [15].

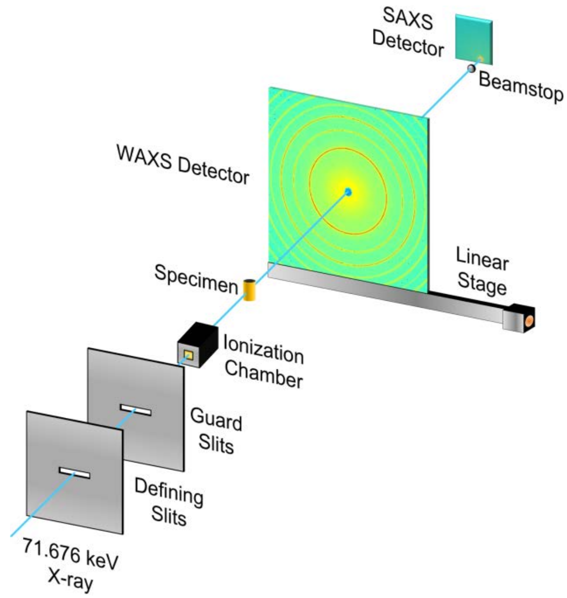

Figure 1: Experiment setup of the synchrotron X-ray scattering investigation

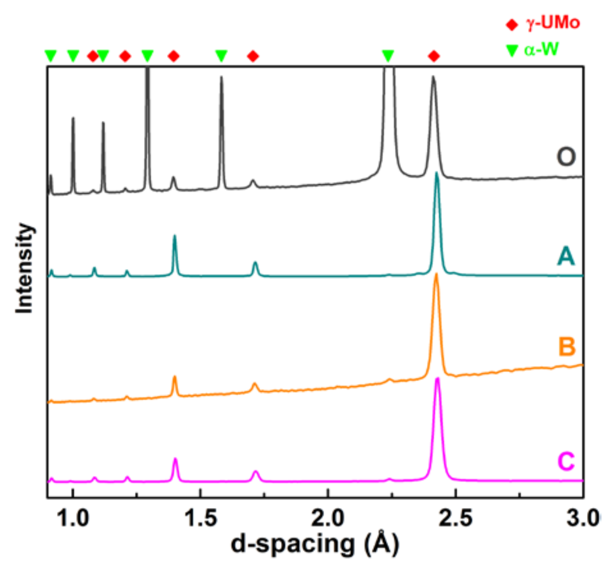

Figure 2: Azimuthally-integrated X-ray diffraction peaks

3. Results and Discussion

All of the U-Mo diffraction peaks are contributed by γ-phase (the original phase in the un-irradiated material), namely, no phase transition occurs during the in-pile irradiation based on the synchrotron data shown in Figure 2. Meanwhile, the 2-D X-ray detector captured the grain sub-division process [16]. Over 70% of the U-Mo phase experiences recrystallization as the burnup approaches 6.3×10^{21} f/cm^3. In addition, as illustrated by Figure 3, the lattice constant of γ-U-Mo rises steeply at low burnup (from O to A), and then increases slightly (from A through B to C). The results are comparable to Golosov et al.'s results [17].

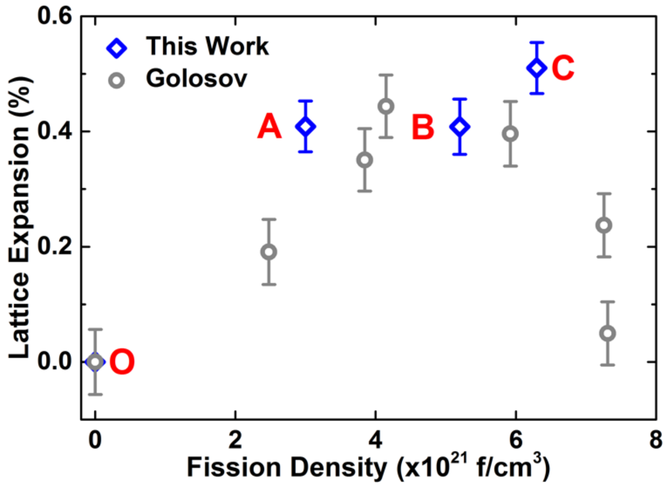

Figure 3: The evolution of lattice constant compared with Golosov's data[17]

According to previous TEM observation of the same batch of fuel plates, nanoscale fission gas bubbles form FCC superlattice at intermediate burnup and then collapse into microscale fission bubbles due to grain sub-division at high burnup, which reduces the grain size from micron level to 100-500 nm [13]. As predicted by diffraction theory, long-distance ordered structures in crystalline matrix such as bubble superlattice contribute to the satellite diffraction peaks near the matrix's diffraction peaks as well as the transmitted beam. The satellite diffraction peaks were identified near all the diffraction peaks of γ-U-Mo phase in Sample A, and absent in Samples B and C as well as the control sample, as shown in Figure 4. The positions of these satellite peaks are consistent with the FCC superlattice structure, and give a superlattice constant around 11.7 nm, which is comparable with the previous TEM measurement (12.0 nm)[13]. SAXS detector also captured the satellite peaks near the transmitted beam, corresponding to similar superlattice constant (12.1 nm).

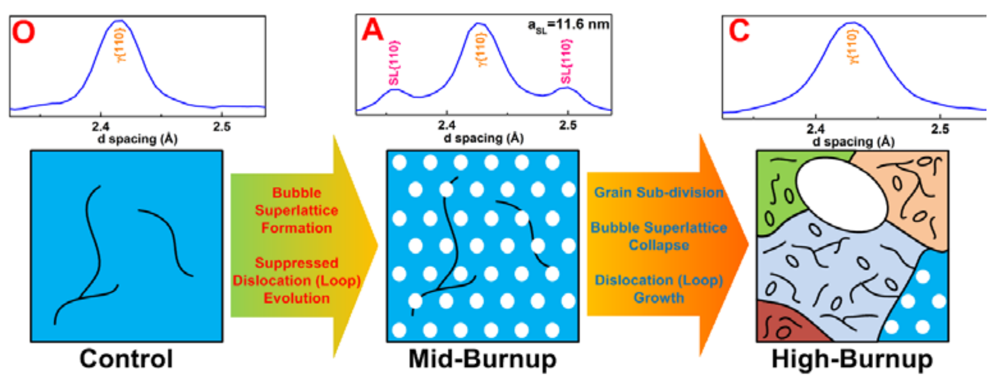

Figure 4: the observation of satellite peaks and the interpretation of the microstructure evolution

The original W-H analysis showed that the internal strain almost remains constant until high burnup. At high burnup, the internal strain steeply rises. The modified W-H analysis further confirmed that the major contribution to the internal strain comes from dislocations or dislocation loops, especially for Samples B and C. Meanwhile, at intermediate burnup, due to the existence of the bubble superlattice, no Guinier zone was found in the SAXS data. As the superlattice collapses in Samples B and C, Guinier approximation gives an average bubble size of 36.6 nm. Thus, aside from those microscale huge bubbles, large-sized nanoscale bubbles also form at high burnup.

By combining all the results mentioned above, the microstructure evolution within the in-pile-irradiated U-7wt%Mo fuel can be deduced, as shown in Figure 4. The formation of the gas bubble superlattice during the intermediate burnup expands the lattice and provides densely distributed sinks for radiation-induced defects, suppressing the evolution of dislocation loops. As recrystallization-induced grain sub-division takes place, it eventually makes those superlattice structures to collapse at high burnup and therefore accelerates the nucleation and growth of dislocation loops.

4. Conclusions

In summary, both synchrotron WAXS and SAXS were utilized to investigate irradiation-induced microstructural modifications within in-pile-irradiated U-7wt%Mo dispersion fuels with different burnup levels. A series of microstructural features, including gas bubble superlattice, lattice expansion, and dislocation density, were quantitatively analyzed so that the microstructural evolution mechanism of irradiated U-7wt%Mo can be clarified. The results of this study develop a better understanding of the fuel performance of U-7wt%Mo, especially at high burnup levels.

5. Acknowledgement

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