Microstructural Characterization of an Irradiated RERTR-6 U-7Mo/AA4043 Alloy

Dispersion Fuel Plate Specimen Blister-Tested to a Final Temperature of 500°C

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ABSTRACT

The Material Management and Minimization (M3) Reactor Conversion Program, in the past called the Reduced Enrichment for Research and Test Reactor (RERTR) Program, is developing low-enriched uranium (LEU) fuels for application in research and test reactors. U–Mo alloy dispersion fuel is one type being developed. Blister testing has been performed on different fuel plate samples to determine the margin to failure for fuel plates irradiated to different fission densities. Microstructural characterization was performed using scanning electron microscopy and transmission electron microscopy on a sample taken from a U-7Mo/AA4043 matrix dispersion fuel plate irradiated in the RERTR-6 experiment that was blister-tested up to a final temperature of 500°C. The results indicated that two types of grain/cell boundaries were observed in the U-7Mo fuel particles, one with a relatively low Mo content and fission gas bubbles and a second type enriched in Si, due to interdiffusion from the Si-containing matrix, with little evidence of fission gas bubbles. With respect to the behavior of the major fission gas Xe, a significant amount of the Xe was still observed within the U-7Mo fuel particle, along with microns into the AA4043 matrix. For the fuel/matrix interaction layers that form during fabrication and then grow during irradiation, they change from the as-irradiated amorphous structure to one that is crystalline after blister testing. In the AA4043 matrix, the original Si-rich precipitates, which are typically observed in as-irradiated U-Mo dispersion fuel, get consumed due to interdiffusion with the U-7Mo fuel particles during the blister test. Finally, the fission gas bubbles that were originally around 2 nm in diameter and resided on a fission gas superlattice (FGS) in the intragranular regions of as-irradiated U-7Mo fuel grew in size (up to ~20 nm diameter) during blister testing and, in many areas, are no longer organized as a superlattice.

1. INTRODUCTION

The Material Management and Minimization (M3) Reactor Conversion Program, in the past called the Reduced Enrichment for Research and Test Reactor (RERTR) Program, is developing low-enriched uranium (LEU) fuels for application in research reactors, and U–Mo alloy dispersion fuel is one type being developed. [1] One fuel system that has been evaluated is a dispersion fuel comprised of U-7Mo fuel particles and Al-Si alloy matrix. Much work has been performed using SEM and TEM to characterize as-fabricated AA6061-clad U-Mo dispersion fuel with different Al-based matrices. [2-4] Further work has been performed characterizing irradiated AA6061-clad U-Mo dispersion fuel with different Al-based matrices. [5-8] Based on the microstructural characterization results reported in the literature, the microstructure of as-fabricated fuel with AA6061 cladding is comprised of U-7Mo fuel particles that primarily contain γ -(U,Mo) phase but also have some decomposed regions primarily at cell/grain boundaries and these regions contain α -U. Fuel matrix interaction (FMI) layers are present around the fuel particles that contain crystalline phases, and these phases contain U, Mo, Al, and Si. Si has been observed to exhibit localized attack along cell/grain boundaries. After a U-Mo dispersion fuel has been irradiated, the U-7Mo fuel particles only contain γ -(U,Mo) phase since irradiation causes α -U to revert back to γ -(U,Mo) phase [9,10]. The FMI layers grow and Si has been observed to migrate into the fuel from the FMI layers [11]. Fission gas bubbles grow in the U-Mo fuel particles. In the intragranular regions, a fission gas superlattice can be observed. [12,13], and at cell/grain boundaries larger fission gas bubbles can be observed. For fuel plates irradiated to intermediate fission densities ($\sim 3 \times 10^{21}$ fissions/cm³), the intra-granular regions are composed of a FGS and at the cell/grain boundaries are located larger fission gas bubbles. For higher fission density samples, fission-induced recrystallization transpires, and most regions of the U-7Mo are comprised of larger fission gas bubbles with little evidence of a FGS. [12]

In addition to stable and predictable behavior during irradiation, a nuclear fuel must exhibit adequate margin to fuel failure during irradiation. For dispersion fuels employed in research and test reactors, this has historically been determined for off-normal conditions by post-irradiation blister testing of irradiated fuel plates. [14] The blister-threshold temperature is established by post-irradiation heating of fuel plates to determine the temperature at which release of fission gas

results in a raised area on the surface of the fuel plate. It is assumed that, at this temperature, the fuel plate no longer meets requirements for operation within the reactor safety basis. The blister-threshold temperature is significantly affected by accumulation of gaseous fission products. A common blister-test method is to heat, hold at temperature, remove, visually examine, and return a fuel sample to the furnace at successively higher temperatures until visual evidence of a blister is seen. The test is then concluded.

For irradiated U-Mo dispersion fuels, it is of interest to determine how microstructural changes in the fuel plate meat during blister testing contribute to the final development of blisters on the surface of the fuel plate. To this end, a sample taken from a dispersion fuel plate comprised of U-7Mo fuel particles and Al-4043 matrix (R3R030) that was irradiated in the RERTR-6 experiment in the Advanced Test Reactor was blister-tested up to a final temperature of 500°C and then characterized using scanning electron microscopy (SEM) and transmission electron microscopy (TEM). To investigate how the fuel microstructure evolved during irradiation and then during subsequent blister-testing, microstructural characterization using SEM was also performed first on a sample from an as-fabricated U-7Mo/AA4043 fuel plate (R3R020) and then on a sample taken from the as-irradiated R3R030 fuel plate before blister testing.

This paper describes the first results for detailed SEM and TEM microstructural characterization of a blister-tested U-Mo dispersion fuel plate. These results are compared to the microstructures observed for a similar fuel plate after fabrication and for R3R030 in the as-irradiated condition. Comments are made on how the microstructure of an irradiated fuel plate has changed after blister testing to 500°C, and how these changes may contribute to the eventual release of fission gas from the fuel meat, which in turn results in the eventual development of blisters on the surface of a fuel plate.

2. EXPERIMENTAL

2.1 Fabrication

The details on how the R3R030 fuel plate and other fuel plates irradiated in the RERTR-6

experiment were fabricated are discussed in [6]. In general terms, the fuel plates had a target fuel loading of 6 gU/cm^3 , used LEU-7Mo (19.7% U-235) powder produced with a rotating-disk centrifugal atomization process, and were fabricated using a standard rolling process comprised of five hot-rolling passes and further reduction using two to three cold-rolling passes. The final fabrication step was a blister test performed at around 485°C for approximately 60 minutes.

2.2 Irradiation Testing

Fuel plate R3R030 experienced the highest power density in the RERTR-6 experiment. [15] As was discussed in [6], where the microstructural characterization of an irradiated RERTR-6 U-Mo dispersion fuel plate with AA6061 matrix was described, the RERTR-6 experiment tested fuel materials to average burnups (BU) of 35–49%, where BU is the amount of U-235 that has fissioned, depending on experiment location with a peak heat flux of $<220 \text{ W/cm}^2$ and a beginning-of-life temperature of $<200^\circ\text{C}$. The experiment was irradiated in the ATR West B12 irradiation test position during cycles 134B, 135B, and 135C, and it remained in the reactor for 135.1 effective full power days (EFPD). The RERTR-6 irradiation test assembly held four capsules, axially designated as A through D from the top of the test assembly to the bottom. Each capsule had two levels, with four mini-plate positions per level, giving eight mini-plate positions per capsule. There were 32 mini-plate positions per test assembly. Due to the orientation of the fuel plates, where one edge of a fuel plate faced the core, there was a large neutron flux gradient across the width of the plates. For the R3R030 mini-plate comprised of U-7Mo particles dispersed in AA4043 alloy matrix (that has a nominal wt.% composition of 4.53Si-0.14Fe-0.09Cu-0.04Ti-0.01Zn-0.008Mg-bal Al), the average BU was 47.9% (72.3% at the edge facing the core and 36.9% on the lower flux side) [15]. Calculations were performed using the fuel performance computer code, PLATE [16], to determine more details of the irradiation conditions of the fuel plate R3R030. The calculations were based on the nominal dispersion fuel plate design, the fuel plate as-built data, and the power history calculated by Chang et al. [17]. R3R030 was determined to have a peak temperature of 97.5°C , an average fission density of $3.3 \times 10^{21} \text{ fissions/cm}^3$, an average fission rate of $2.8 \times 10^{14} \text{ fissions/cm}^3 \text{ s}$, and peak heat flux of 101.5 W/cm^2 .

2.3 Blister Testing

Blister-threshold temperature data are obtained by heating irradiated fuel plates or pieces of fuel plates (typically only for dispersion fuels) at successively higher temperatures between 350 and 550°C. For fuel plate R3R030, a 2.5 cm by 2.5 cm fuel plate section was used. A plate sample is held at temperature for a period of time (20 minutes), the furnace is turned off, and then the plate sample is removed from the furnace and visually examined for blisters. If no blister is observed, the plate sample is returned to the still warm (~70–130°C) furnace for the next anneal cycle at the next higher temperature setting (increasing in 25°C increments). This process is repeated until a blister is observed. The test is then concluded, and the temperature at which the blister was first observed is recorded as the blister-threshold temperature. The plate samples are heated at the maximum ramp rate of the furnace (~8°C/min) and cool in the ambient cell atmosphere (argon) when withdrawn for inspection, similar to the procedure used in most previous blister tests. The cell has an argon atmosphere which is typically at ~30°C.

The visual examination for identification of blisters is accomplished using a telescopic camera lens, binoculars, and additional local in-cell lighting. Verification of the presence or absence of blisters is provided by at least two observers. Photographs of both surfaces of the plate are taken between each blister-anneal cycle to provide a record of blisters and other changes in the appearance of the plate.

The inherent temperature uncertainty of the system and test protocol is $\pm 13^\circ\text{C}$ (comprising accuracy of the thermocouple and controller/readout, furnace temperature uniformity and test-environment noise) and $\pm 24^\circ\text{C}$ (introduced by the temperature-increment increase), respectively.

2.4 Microstructural Characterization

Optical metallography (OM) was performed on a transverse cross section taken at the midplane of the as-irradiated sample R3R030 using a slow-speed saw (see Figure 1). In addition, a thin slice (~ 1-mm-wide) was produced adjacent to the mid-plane sample, from which two 1-mm-diameter, cylindrical punchings were taken through the nominally 1.9-cm-wide, 0.06-cm-thick fuel meat of fuel plate. One of these samples was then transferred to the Electron Microscopy Laboratory where it was mounted, polished and inserted into a ZEISS Model 960A scanning

electron microscope (SEM) that was equipped with an Oxford wavelength dispersive spectrometer (WDS) and energy dispersive spectrometer (EDS) that employed ISIS LINK software or a JEOL JSM-7000F field emission gun SEM with WDS and EDS and Oxford INCA software. The as-fabricated R3R020 sample was also characterized using this equipment. Backscattered electron (BSE) and secondary electron (SE) images were generated to examine the fuel microstructure, and the spectrometers were employed to generate x-ray maps and linescans and perform semi-quantitative point-to-point compositional analysis to determine the presence of various elements in the fuel, interaction zone, and matrix. The Oxford INCA computer software corrected for the 50 nm-thick layer of Pd coating on the sample when it determined phase compositions.

(INSERT FIGURE 1)

Characterization samples were produced from the blister-tested sample R3R030 as shown in Figure 2. Using a slow-speed saw, a small strip of the fuel plate was produced in the center region, and it was oriented longitudinally. From this small strip, small pieces were sliced. One of these samples was sent to the EML where it was then mounted and polished. SEM analysis was completed on the polished surface of the sample using a JEOL 7000F FEG SEM equipped with EDS and WDS. A FEI Quanta3D Dualbeam focused ion beam (FIB) was employed to produce multiple "lift-outs" at specific locations by coarse trenching approximately a 15- μm x 15- μm x 1- μm sample so that surfaces with little polishing damage could be obtained. For transmission electron microscopy (TEM) analysis, samples were thinned to ~100 nm of thickness to be transparent to the 200 KeV electron beam. Detailed TEM microstructural characterization was performed using a 200 kV JEOL-2010 TEM/STEM system equipped with a LaB₆ filament, a Gatan UltraSacr-1000 digital camera for imaging and a Bruker Si drift detector for composition analysis with EDS. The TEM selected area diffraction (SAD) patterns from the major zones were used for structure analysis.

(INSERT FIGURE 2)

3. RESULTS

3.1 Blister Testing

Figure 3 shows the heating profile for the R3R030 sample blister test. At 500°C, blisters were observed on both sides of the sample (see Figure 4). The blisters were observed along the fuel meat/cladding interface.

(INSERT FIGURE 3)

(INSERT FIGURE 4)

3.2 Microstructural Characterization

3.2.1 As-Fabricated

An SEM micrograph is presented in Figure 5 that shows the microstructure of the as-fabricated fuel meat for fuel plate R3R020, which was manufactured in the same manner as R3R030.

(INSERT FIGURE 5)

A more detailed characterization of the powders (see Figures 6 and 7) showed that after the five rolling passes at 500°C that were used for fabricating these plates and after the blister anneal testing that occurred after hot rolling (485°C for ~60 minutes), noticeable fuel/matrix interaction had occurred.

(INSERT FIGURE 6)

(INSERT FIGURE 7)

(INSERT FIGURE 8)

The fuel/matrix interaction that occurred resulted in the formation of a fairly uniform layer around each particle, along with localized areas where the rate of interaction was greatly enhanced. There appeared to be interaction product that formed along boundaries into the fuel particles. Figure 6(e) shows that Si particles were present in the as-fabricated fuel. Si can show up in the fuel particles as an impurity. Point-to-point SEM/EDS analysis was performed on the interaction layer that surrounded the fuel particle. The results of this analysis indicated that the interaction layer had a Si content of up to 45 at%; the maximum (Al + Si) concentration was measured to be 69 at%; and the (Al+Si)/(U+Mo) ratio varied between 1.7 and 2.2.

3.2.2 As-Irradiated

3.2.2.1 Optical Metallography

In Figure 8 is presented a low-magnification optical micrograph of the as-irradiated R3R030 fuel plate microstructure at the transverse cross section taken at the mid-plane, along with higher-magnification images taken at different locations. U-7Mo fuel particles, a narrow (1-2 microns thick) interaction layer around the fuel particles, and AA4043 alloy matrix constitute the fuel meat microstructure.

(INSERT FIGURE 9)

3.2.2.2 Scanning Electron Microscopy

Figure 10 is an optical micrograph that approximately delineates where a sample punched from the irradiated R3R030 fuel plate for SEM analysis would have passed through the fuel meat microstructure. Figure 11 shows SEM micrographs of that sample after mounting and polishing.

(INSERT FIGURE 10)

(INSERT FIGURE 11)

X-ray maps for U, Mo, Al, and Si are presented in Figures 12 and 13. The presence of a Si-rich interaction layer around the fuel particles is apparent, along with Si-rich precipitates in the AA4043 alloy matrix. Figure 12 (e) shows where regions close to some fuel particles are devoid of the Si-rich precipitates in the matrix.

(INSERT FIGURE 12)

(INSERT FIGURE 13)

3.2.3 As Blister-Tested

3.2.3.1 Scanning Electron Microscopy

SEM micrographs of the sample taken from the blister-tested fuel plate section are presented in Figure 14. The general microstructure consisted of regions bright in contrast along cell/grain boundaries that exhibited fission gas bubbles. The cell/grain interiors were of darker contrast due to the higher Mo concentration. One isolated feature of interest is shown in Figure 14 (f) where, due to the indistinct morphology of the features observed, it is possible that this material at this very localized location was liquid at some point in time during the blister test. As reported in [18], multiple ternary Al-Mg-Si compositions exist that will be liquid at temperatures below 500°C (For example, 64Al-39Mg-0.1Si is a liquid at 451°C), and AA4043 contains Al, Mg, and Si. No other regions in the sample microstructure were observed to have similar morphologies.

Figure 15 shows BSE and SE images where crack-like features are present in some fuel particles. Also presented in this figure are x-ray maps for Xe, Al, U, Mo, and Si. The Xe x-ray map shows that even though the sample was exposed to high temperatures up to 500°C appreciable Xe remains in the microstructure, including the AA4043 matrix. The Si x-ray map indicates that the "crack-like" features shown in (a) and (b) are enriched in Si. The Si x-ray map also shows that almost all the Si precipitates that were present in the AA4043 matrix after fabrication and

irradiation are no longer present after blister testing. They appear to have been consumed during the FMI interaction process that occurred during the exposure to high temperatures, whereby Si-rich interaction layers changed in thickness and concentration around the fuel particles, and Si penetration extends into the fuel particles from the interaction layer along cell/grain boundaries.

(INSERT FIGURE 15)

Composition analysis was performed along a line that extended from the matrix across the FMI layer and into the fuel across some of the "crack-like" features. The location of the line is presented in Figure 16, along with the results for U, Mo, Al, and Si in (b) and for Zr, Ba, Xe, Nd, and Si in (c). Si is enriched in the FMI layer and in the location of the "crack-like" features. Xe is observed in the matrix and the fuel particle. Ba, Nd, and Zr can also be measured.

(INSERT FIGURE 16)

Some fuel particles with extensive Si penetration along cell boundaries were observed (see Figure 17). These regions look different compared to boundaries without this penetration. SEM/EDS indicated approximately 5 wt. % Si is present on the dark contrast cell/grain boundaries. It appears that there are fewer fission gas bubbles in these regions, and the Si-enriched phase is interconnected through the microstructure of some of the fuel particles and extends to the fuel/matrix interface.

(INSERT FIGURE 17)

FIB samples were produced from a U-7Mo fuel particle that exhibited no evidence of Si-attack along the cell/grain boundaries. These samples were produced for SEM and TEM analysis to determine how the fission gas bubble size and distribution may have changed during blister

testing in irradiated U-7Mo particles that were not attacked by Si. This information is needed to determine when fission gas may be released as irradiated U-7Mo is heated up to higher temperatures. Evidence of interconnection of fission gas bubbles could indicate that release of some fission gas had occurred from the U-7Mo particles. Figure 18 shows SE images of the location where the samples were produced on the sample, and the locations where two samples (Site 1 and 2) were produced within an irradiated U-7Mo fuel particle and where one sample (Site 3) was produced at the interface of a fuel particle and the AA4043 matrix. Additionally, higher magnification images are presented that show the microstructure of the three different samples. Only fission gas bubbles on cell/grain boundaries were observed, and a few isolated fission gas bubbles were found in the FMI layer. One high magnification image from the site 1 sample is presented that shows the solid fission product phases present in a cell/grain boundary fission gas bubble. There is no obvious indication that the fission gas bubbles have become interconnected, but this is hard to demonstrate in 2-dimensional images.

(INSERT FIGURE 18)

3.2.3.2 Transmission Electron Microscopy

3.2.3.2.1 Site 1 Sample

An SEM micrograph of the final Site 1 TEM sample is presented in Figure 19. Fission gas bubbles along cell/grain boundaries are visible. Figure 20 shows actual TEM micrographs of the sample. The TEM micrographs in (a)-(c) show the bubbles contained in the intragranular regions of the U-Mo microstructure at different magnifications. (d) is a selected area diffraction pattern at zone [113] that shows the U-7Mo is a bcc γ -(U,Mo) phase.

(INSERT FIGURE 19)

(INSERT FIGURE 20)

3.2.3.2.2 Site 2 Sample

An SEM micrograph of the Site 2 TEM sample is presented in Figure 21. The fission gas bubbles along cell/grain boundaries are visible, as was the case for the Site 1 sample. TEM micrographs for the Site 2 sample are presented in Figure 22. (a) is a TEM micrograph that shows the GBS at low magnification and (b) shows the GBS at a higher magnification at [011]. (c) is a selected area diffraction (SAD) pattern at [011] showing γ -(U,Mo) phase. (d) is a TEM micrograph showing a mixture of the fine superlattice bubbles (~ 3nm) and randomly distributed medium size bubbles (~ 20 nm). (e) shows where only medium sized bubbles are present. During blister testing, it appears the GBS within some of the intragranular region of the sample changed to a microstructure comprised of larger fission gas bubbles. The GBS is still present in about 50% of the sample area.

(INSERT FIGURE 21)

(INSERT FIGURE 22)

EDS composition analysis was performed on the Site 2 sample using the TEM, and the results are shown in Figure 23. A TEM micrograph showing specific locations in the Site 2 sample where TEM/EDS measurements were performed, and a table showing the results for U, Mo, Zr, Xe, and Si (at.%) are presented. Xe was detected in some local areas of the sample, along with low and high Mo content regions. Some of the EDS measurements reveal the presence of Si in the U-7Mo fuel particle. With respect to the Mo content in the fuel particle, low Mo concentrations were observed at the cell/grain boundaries (points 13-18). In the medium size bubble region, the results for points 9 and 10 also indicate low Mo concentration. Within the regions with observable fission gas bubbles, points 2 and 4 indicate a low Mo concentration of 16 at.%, and points 1, 3, 6, 8, 17, and 19 indicate higher Mo concentrations of 20 at.%.

(INSERT FIGURE 23)

To reflect the change in U, Mo, Zr, and Xe content across a region of the Site 2 sample, a compositional linescan was produced (see Figure 24). A TEM micrograph indicating where the TEM/EDS linescan analysis was located in the sample is presented in (a). (b) shows the results in at.% for U, Mo, Zr, and Xe, and (c) shows the results in wt.% for these elements. The concentration for U, Mo, Zr, and Xe all varied across the sample.

(INSERT FIGURE 24)

3.2.3.2.3 Site 3 Sample

An SEM micrograph of the Site 3 TEM sample is presented in Figure 25. The fission gas bubbles along cell/grain boundaries are visible, as was the case for the Site 1 and 2 samples, and the FMI layer was primarily devoid of fission gas bubbles. TEM micrographs for the Site 3 sample are presented in Figure 26. The upper left image is A SAD pattern for the Al, the upper middle image is a TEM micrograph showing the Site 3 sample microstructure at the fuel-matrix interface, and the upper right image is a TEM micrograph showing the observed larger fission gas bubbles and a small area with intact GBS (dashed white box). At the lower left is a SAD pattern for the FMI layer shown in the upper middle image. The lower middle image is a SAD pattern for a region near the U-7Mo/FMI layer interface where relatively high Si is present. At the lower right is a TEM micrograph of a region in the U-7Mo where only larger fission gas bubbles are observed.

(INSERT FIGURE 25)

(INSERT FIGURE 26)

Point-to-point compositional analysis was performed on the Site 3 sample with a focus on areas around the FMI layer. Figure 27 shows the locations where the point analysis was performed along with the results of the analysis. The Mo content (at.%) measured at points 5, 6 and 7, which is located in the U-7Mo fuel particle but near the U-7Mo/FMI layer interface, is 10.9, 9.4

and 10.1, respectively. In this region, high Si concentrations (14-16 at%) were also observed. The FMI layer contains around 18-22 at% Si and is approximately 2 μm thick. More large bubbles are observed in the Si-rich region of the U-Mo compared to the actual FMI layer, possibly indicating a different irradiation performance for two different phases.

(INSERT FIGURE 27)

Point-to-point compositional analysis was also performed on the Site 3 sample with a focus on the U-7Mo fuel. Figure 28 shows the locations where the point analysis was performed along with the results of the analysis. The highest Mo content is observed at point 4 (25.0 at.%; 12.6 wt.%), and the lowest Mo content was observed at point 8 (15.6 at.%; 6.9 wt.%).

(INSERT FIGURE 28)

Figure 29 shows the results for U, Mo, Al, and Si concentration linescan analyses that were performed across the FMI layer at two locations in the sample. It was observed that the FMI layer contained U, Mo, Si, and appreciable Al. Si was also enriched just adjacent to the FMI layer and into the U-Mo fuel.

(INSERT FIGURE 29)

4. DISCUSSION

In terms of describing how fission gas bubbles form in irradiated nuclear fuel and then change in size and morphology during post-irradiation annealing, Olander discusses these behaviors for uranium-oxide fuel. [19] He highlights the fact that fission gas behavior during irradiation is affected by parameters like temperature, matrix stress, fission rate, irradiation time or burnup, fuel properties, vapor pressure, surface tension, surface and bulk diffusion, creep strength, fission gas properties, nuclear yields, dislocation density, grain size, restructuring, crack pattern, and fuel microstructure. Due to the large number of variables that affect fission gas bubble

development, it is extremely difficult to produce computer models that can describe this behavior. Olander highlights the fact that the growth of gas bubbles during irradiation is impacted by the atomic migration of fission gas atoms to existing gas bubbles. The availability of vacancies will also enable a bubble to expand as gas begins to accumulate. Surface tension and the stress state of the fuel matrix will determine the size of the bubble. Resolution of gas atoms in a bubble and the migration of bubbles also affect bubble growth. Temperature and stress gradients will impact bubble growth, as will dislocations or grain boundaries since they can provide a restraining force. On the other hand if there is grain growth, grain boundaries can sweep fission gas bubbles, and dislocations can sweep bubbles in response to a mechanical stress. Crystal defects can pin a bubble if they are immobile, or put bubbles in motion if they move. Fission gas can release from a nuclear fuel either to external surfaces such as cracks in the fuel or to gaps at the fuel/cladding interface or to internal surfaces such as grain boundaries. When grain boundary fission gas bubbles are large or numerous, they can link up and release gas to external surfaces. Release of fission gas by direct flight of energetic fission fragments out of an external surface is only significant at low temperatures.

Kulcinski et al. [20] discussed the behavior of irradiated U when it was heat-treated. They observed that for temperatures above 500°C the U begins to expand due to growth of fission gas bubbles. The amount of bubble swelling that will be observed is a function of annealing temperature, time, burnup, external restraint, chemical composition, and mechanical history. They declared that the effect of external constraint is not well understood. Based on their work, hydrostatic pressure is very effective in reducing swelling and directly impacts the size and number of bubbles formed in the U. Two common sources of constraint are the material strength and the surface tension of the bubble. Surface tension is very important for controlling fission gas swelling at low stresses. As temperature is increased, the vacancy to gas atom ratio for a bubble increases (i.e., the number of vacancies captured per gas atom increases as the bubble grows). The amount of swelling in uranium increases as the average bubble size increases. During annealing, the larger bubbles can grow by impingement as well as diffusion by smaller bubbles. Kulcinski et al. [21] has also discussed the growth of fission gas bubbles during heat treatment of irradiated U wafers at 600°C, and they observed bubble coalescence at the boundaries of newly recrystallized grains, and the size of the bubbles increased as the time of the

anneal was extended. For bubble growth during a heating test, Olander [20] points out that people have modeled bubble growth allowing for diffusion of the single gas atoms still remaining in the matrix to the bubbles or by coalescence where bubbles are mobile in the solid and move either in a random fashion or under the influence of a driving force. Coalescence is efficient at making large bubbles from small ones, and can have a profound effect on the swelling of a nuclear fuel.

Kashibe et al. also investigated growth of fission gas bubbles during the heating of irradiated UO_2 [22]. Initially, they characterized irradiated UO_2 fuel pellets using TEM and SEM fractography and then performed heating experiments at 1600 and 1800°C. The starting fuel had a high density of small bubbles about 2 nm in diameter uniformly distributed in the matrix. Higher burnup fuel samples had 20 nm sized bubbles in addition to the small bubbles. When heating samples to high temperatures the bubble growth was saturated in a few minutes. Enhanced coarsening of bubbles was observed near grain boundaries in medium burnup samples and throughout the grains for high burnup samples. The observed bubble growth was associated with a decrease in the bubble density. The coarsening of bubbles was attributed to coalescence via bubble migration and Ostwald ripening occurred due to a sufficient supply of vacancies from external sources such as free vacancies, grain boundaries or irradiation induced sub-grain boundaries for the very-large bubbles that were observed that exceeded 100 nm in diameter. It is possible that the irradiated U-Mo fuel samples exhibit behaviors that are similar to what has been seen for irradiated UO_2 samples. The R3R030 sample discussed in this paper is considered a medium-burnup sample. A higher burnup sample may have large bubbles completely through the grains. Further work is needed to understand the potential coarsening of fission gas bubbles during heating and the Ostwald ripening that may occur.

The growth and eventual interconnection of fission gas bubbles can eventually lead to the development of blisters on a fuel plate. For irradiated UO_2 fuels, the fission gases accumulate in the grain boundaries of the material during base irradiation and begin to release in a burst within a short duration of about 1-2 min on a temperature ramp above the critical temperature. [23]. For UO_2 , the rapid release of accumulated fission gases transpires through the rapid growth and interlinkage of grain boundary bubbles and formation of grain edge tunnels. Une and Kashibe

investigated the behavior of irradiated UO_2 -2 wt% GdO fuel and compared their results with irradiated UO_2 fuel. They concluded that the critical temperature for the onset of burst release for an irradiated UO_2 -2 wt% GdO fuel became lower with increasing burnup, as was the case for UO_2 fuels. It was observed to be constant between 1400 and 1500°C at higher burnups above 20 GWd/t for both fuels. Below 20 GWd/t, the burst release temperature for the irradiated UO_2 -2 wt% GdO fuel was about 100°C lower than that for the irradiated UO_2 fuels. The fractional burst releases of Kr for UO_2 -2 wt% GdO, increased with increasing burnup, and were about 2-3 times larger than the values of UO_2 , at a given burnup. The remarkable difference was linked to a grain size effect. Bubble swelling was investigated for both fuels by looking at the difference in porosity before and after annealing. The bubble swelling for both fuels increased with burnup. The UO_2 -2 wt% GdO, fuels had a stronger burnup dependence than the UO_2 fuels, and at high burnups above 20 GWd/t, bubble swelling for the former was about 1.5 times larger than that for the latter. This difference in bubble swelling was due to the difference in grain size of both fuels. It was concluded that at a given bubble size, bubble swelling becomes smaller with increasing grain size, because of the smaller grain boundary area or smaller area of precipitation sites per unit fuel volume for larger grain size fuels.

Even though no other microstructural characterization data is available in the literature for blister-tested, irradiated U-Mo dispersion fuel, there is data for other types of heating tests. [24] Golosov et al. used diffraction to characterize irradiated U-Mo/Al dispersion fuel that was annealed within the range 150 to 550°C. They observed a decrease of the lattice parameter of both γ -(U,Mo) and Al. The decrease of the γ -(U,Mo) lattice parameter depended on the burn-up value. The Al lattice parameter decreased with increasing annealing temperature. At the lower annealing temperatures, the lattice parameters decreased for both γ -(U,Mo) and Al, and this was attributed to the annealing of the radiation defects in U-9Mo, the Al matrix, and the Al cladding. Also, the nucleation of gas pores in the Al matrix was suggested to have an effect. Further decrease of the lattice parameter at higher temperatures for both γ -(U,Mo) and Al was suggested to be due to the precipitation of the gaseous fission products from the γ -(U,Mo), along with further development of gas porosity in the Al.

Additionally, results of fission gas release studies have been reported for irradiated U-Mo

dispersion fuel samples with AA6061 cladding, but no microstructural characterization was reported for the samples that were tested to 1,000°C. [25] The samples were tested at a constant heating rate and the temperatures where fission gas was released was determined. Burkes et al. tested samples from U-7Mo/Al-2wt%Si AFIP-1 fuel plates that were irradiated in the Advanced Test Reactor with a beginning-of-life (BOL) surface heat flux of between 116 and 316 W/cm² for 158.2 effective full power days. [26]. They performed fission gas release measurements by heating square irradiated AFIP-1 segments in a thermogravimetric/differential thermal analyzer instrument that was combined with a mass spectrometer. The AFIP-1 samples were heated from room temperature to approximately 1000°C at 10°C/minute, and samples that had been irradiated to either a fuel particle fission density of 5x10²¹ fissions/cm³ or 6.5x10²¹ fissions/cm³ were tested. Each of the samples exhibited three distinct fission gas release events between 400 and 700°C. The authors associated the first release at between 450 and 480°C to be release of fission gas that had recoiled into the surrounding matrix or fuel matrix interaction layers. At between 530 and 550°C, it was suggested that fission gas was being released from intergranular fission gas bubbles in the U-7Mo fuel particles. They hypothesized that the amount of fuel particle grain refinement during irradiation and the amount of FMI layer formation could play a significant role in the amount of fission gas that is released. At around 640°C (near the Al matrix melting point), it was determined that an exothermic reaction occurred between the U-7Mo fuel particles and Al matrix. Dramatic cracking of the U-7Mo fuel particles was observed near this temperature. The remainder of the fission gas was released from the sample via events that occurred near 700 and 900°C.

Zweifel et al. [27] have reported results for fission gas release studies using samples from irradiated U-7Mo/Al-2wt%Si dispersion fuel plates with AG3NE and AlFeNi cladding. The fuel particles in these fuel plates were oxidized during manufacturing so that UO₂ was present in a layer that surrounded the fuel particles that was around 1 micron thick. The fuel plates had been irradiated in the IRIS-4 experiment in the OSIRIS reactor. The samples came from a fuel plate that had been irradiated to a fission density of around 3.4x10²¹ fissions/cm³ (mean value average across the whole fuel plate). The peak beginning-of-life heat flux for the fuel plate was 258 W/cm², and the outer cladding temperature reached around 100°C. Zweifel et al. observed that below 400°C around 2.4% of the fission gas was released from the samples; at between 400 and

500°C around 13.8% was released; and between 500 and 670°C another 57.0 % of the fission gas was released. At 670°C, the fission gas release appeared to be associated with a strong matrix interaction with the U-7Mo fuel particles close to the cladding. Zweifel et al. also heat-treated an irradiated fuel sample to 500°C (at 6°C/minute) at which time the furnace was turned off. The heat-treated sample was then microstructurally characterized. Using EPMA, cracks were observed in the FMI layers. Additionally, complete Si diffusion into the FMI layers was observed (i.e., no more Si was observed in the fuel meat matrix). Within the U-7Mo fuel particles, small fission gas bubbles were observed and cracking, and compositional analysis showed that Xe was still observed in the U-7Mo (as relatively large precipitates) and in the FMI layers. Based on results of x-ray diffraction the presence of α -U in the U-7Mo fuel particles was identified, the γ -(U,Mo) lattice parameter had decreased, and UAl_3 was identified due to crystallization of the FMI layer.

In addition, results have been reported for a heating test of irradiated U-10Mo fuel by itself using a TEM in-situ heating stage [28]. Gan et al. used a FIB-generated sample U-10Mo sample (10 microns by 15 microns in size) from an irradiated monolithic fuel plate (L1F040), where the local fission density and irradiation temperature of the sample were estimated to be 3.5×10^{21} fissions/cm³ and 140°C, respectively. The heating steps consisted of heating to 700°C at 10°C per minute where it was held for 34 minutes and then further increased to 850°C at a rate of approximately 5°C per minute. It was observed that no significant changes occurred in the fission gas superlattice that was present in the sample before heating. The γ -(U,Mo) phase was observed to be stable to high temperatures.

By comparing the results for controlled heating tests using irradiated U-Mo dispersion fuels with what has been found for a blister-tested R3R030 sample, some differences and agreement can be identified. The Gan et al. results suggest that the FGS will remain stable in irradiated U-Mo fuel up to high temperatures if a constant heating rate is used (10°C per minute) followed by isothermally holding the sample at a high temperature. Whereas, the FGS is not stable for a sample blister-tested to 500°C, where the sample is cyclically heated and cooled from a high temperature to near room temperature. It is not clear how testing a FIB sample may impact heating test results. It is speculated that the stress state may be quite different between the thin

TEM foil from in-situ heating and the fuel particles in bulk sample from blistering test. As stated earlier, Kulcinski et al. [20] pointed out that the amount of bubble swelling during heating will be a function of annealing temperature, time at temperature, burnup of the sample, external restraint on the sample, sample chemical composition, and mechanical history of the sample, and some of these parameters could be different for a U-10Mo FIB sample compared to a bulk U-7Mo sample. Also, the behavior of vacancies, which can contribute to bubble growth during heating, may behave differently in a FIB sample, where they may annihilate more at surfaces and not contribute as much to bubble growth compared to a bulk sample. For the blister test sample, the time above 350°C is around three hours. For the sample heated by Gan et al., the time above 350°C is about 1.5 hours. The blister-tested sample has also gone through cyclic heating, which results in severe thermal stresses, and it can impact potentially the constraint that the irradiated U-Mo provides on the fission gas bubbles, along with other properties of the material. Furthermore, the U-Mo particles are in contact with a dissimilar matrix material that results in interdiffusion of fuel and matrix constituents. The effects of Si attack along grain boundaries, of growth of the FMI layer, and of crystallization of the FMI layer on the stability of the FGS are not well understood. All these phenomena could also affect the properties of the U-Mo, which may affect the growth of the fission gas bubbles. None of the R3R030 TEM samples displayed the presence of α -U. However, Zweifel heat-treated a U-Mo dispersion fuel sample at 6°C per minute to 500°C (total heating time above 350°C was 25 minutes with added time cooling in the furnace) and they observed α -U using x-ray diffraction analysis. Therefore, it is possible that α -U was present in the R3R030 sample after blister testing, but the TEM analysis of the three 15 x 15 micron samples did not capture the development of this phase. On the other hand, the samples in the Zweifel et al. study had UO₂ coating around the fuel particles, which may have contributed to the identification of α -U in their heat-treated sample. The R3R030 sample had no coating. Analysis of the R3R030 TEM samples did show that even in samples with no evidence of α -U the FGS breaks down during heating for a material comprised of the γ -(U,Mo) phase, and the fission gas bubbles grow in size, become randomly distributed and are no longer assembled as a superlattice. The randomly distributed fission gas bubbles have increased from about 2-3 nm in size to around 20 nm in size. This suggests that it was not a phase change in the material that caused the observed changes in the distribution of the fission gas bubbles.

With respect to the chemical interactions that occur between the U-7Mo fuel particles and the matrix during the cyclic heating of a blister test and how this may impact release of fission gas from fuel particles, Si penetrated along the U-Mo cell/grain boundaries resulting in the formation of Si-rich phases, which may hold fission gas differently than U-7Mo. There is little evidence of fission gas bubbles in these regions of the fuel particles suggesting that fission gas has migrated from these regions to different areas of the fuel meat. During fabrication, some Si has penetrated along some cell/grain boundaries from the matrix into the U-Mo particles, and it appears that penetration of Si has continued during blister testing, along with the almost complete consumption of the Si-rich precipitates still present in the as-irradiated matrix. These Si-rich "pathways" can extend from the center of the fuel particles all the way to the fuel/matrix interface. It is possible that Si attack along grain boundaries provides pathways for mobilization of fission gases. However, Xe x-ray maps show that significant Xe is still in the fuel particles and the AA4043 matrix after blister testing up to 500°C for a region of the fuel exposed to around 3.3×10^{21} fissions/cm³. The TEM results confirm this observation by showing significant Xe in the U-7Mo fuel and also the FMI zone in a region of the fuel plate. TEM data also shows that these FMI layers are no longer amorphous, but instead are crystalline. The initial amorphous FMI phase present in irradiated fuel [13] becomes nano-crystalline during blister testing. Figure 30 shows that based on electron diffraction patterns, the pattern for the FMI layer in the blister-tested sample agrees well with an electron diffraction pattern produced from the FMI layer in as-fabricated fuel. During heating, the mobility of fission gases present in the amorphous interaction layer could change during exposure to higher temperature due to the nucleation and growth of crystallites, which could change the properties of the interaction layer with respect to the diffusion behavior of fission gases. Therefore, it appears that these changes in the microstructure due to fuel/matrix interaction that occurred during blister-testing combined with the changes in the fission gas bubble size, morphology, and distribution within the U-7Mo particles may be important contributors to the formation of the blisters that were observed at the final testing temperature of 500°C. At the locations where actual blisters developed on the R3R030 fuel plate sample, the fission density was higher and the fuel/matrix interaction may have been more aggressive and the fission gas bubbles in the U-7Mo may be larger. Future work will focus on the microstructure at actual locations on a blister-tested fuel

plate where blisters are present to determine how much FMI and growth of fission gas bubbles in the U-7Mo actually leads to blistering, along with identification of the microstructural changes in the cladding. These results will be compared to those discussed in this paper, where the fuel meat microstructure changes that occur during heating but do not lead to blistering have been highlighted. These results will be reported in a future paper.

(INSERT FIGURE 30)

5. CONCLUSIONS

Based on the results of the microstructural characterization of an irradiated U-7Mo dispersion fuel that was blister-tested at temperatures up to 500°C, the following conclusions can be drawn:

1. The FGS in U-7Mo fuel particles that remain as γ -(U,Mo) phase becomes unstable during the blister test to high temperatures resulting in an order of magnitude change in the size of the fission gas bubbles, and the larger bubbles become randomly, but fairly uniformly, distributed in the U-7Mo.
2. FMI occurs during high temperature blister tests resulting in Si attack along the cell/grain boundaries of the fuel particles, almost complete consumption of the Si-rich precipitates in the matrix, and some change in thickness of the FMI layers. The areas of Si attack exhibit a reduction in the amount of observable fission gas bubbles compared to cell/grain boundaries not attacked by Si.
3. The FMI layers that are amorphous in irradiated U-Mo dispersion fuel plates crystallize during blister testing to high temperatures. However, a Si-rich phase that is adjacent to the U-7Mo remains amorphous.

4. Xe is observed in the U-7Mo fuel particles and the FMI layer, suggesting that for a sample irradiated at intermediate irradiation conditions that is later blister-tested to 500°C significant fission gas does not migrate from the original locations of the as-irradiated fuel microstructure.

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Figure 1. Schematic diagram showing pieces that were sectioned from fuel plate R3R030 and used for SEM and OM analysis, along with burnup analysis using chemical analysis techniques. The punching locations are approximately 0.64 cm from the plate edge. The 2.5 cm-long, cross-hatched section of the plate was later used for blister testing.

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Figures

(Figure 1)

(Figure 2)

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(Figure 4)

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(Figure 7)

(a)

(b)

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(Figure 9)

(a)

(Figure 10)

(Figure 11)

(Figure 12)

(Figure 13)

(e)

(Figure 14)

(a)

(Figure 15)

(Figure 16)

(b)

(c)

(Figure 17)

(Figure 18)

(Figure 19)

(Figure 20)

(Figure 21)

(Figure 22)

(Figure 23)

ID	Si (at%)	Zr	Mo	Xe	U
1	4.7	3.4	20.2	0	71.8
2	3.4	3.6	16.9	10.1	66.0
3	3.2	3.9	20.2	0	72.7
4	2.4	4.0	15.1	8.7	69.8
5	3.0	3.0	18.9	2.8	72.3
6	4.2	4.3	20.4	0	71.1
7	4.1	4.6	18.1	0	73.2
8	2.0	3.4	20.6	0	74.1
9	0	0	5.8	14.8	79.5
10	0	0	12.5	9.0	78.6
11	2.4	4.6	18.3	0	74.7
12	2.4	3.4	18.5	0	75.8
13	0	0	10.6	2.4	87.0
14	4.0	4.7	15.6	6.1	69.6
15	0.3	1.8	14.5	13.0	70.4
16	0	0	14.7	0	85.3
17	0.6	6.4	21.7	0	71.3
18	0	2.7	14.1	0	83.2
19	1.5	2.2	23.8	0	72.5
20	3.0	4.9	17.3	7.3	67.4

(b)

(Figure 24)

(c)

(Figure 25)

(Figure 26)

(Figure 27)

ID	Al (at%)	Si	Mo	Xe	U
1	100.0	0	0	0	0
2	65.5	20.3	3.3	0	10.9
3	59.5	22.1	3.6	1.7	12.9
4	60.4	17.6	3.9	4.8	13.4
5	5.7	15.1	19.1	0	60.2
6	6.2	14.0	16.6	2.3	60.9
7	5.6	15.7	17.7	0	61.0

(b)

(Figure 28)

(a)

ID	Zr (at%)	Mo	Xe	U
1	4.1	20.5	4.4	71.0
2	4.4	17.9	15.4	62.4
3	3.8	18.6	7.5	70.1
4	5.7	25.0	4.0	65.3
5	4.1	22.6	1.6	71.7
6	4.8	21.0	0	74.1
7	3.3	21.7	0	75.1
8	0	15.6	0	84.4
9	5.1	18.0	8.9	68.1
10	4.3	22.0	2.5	71.2
11	3.6	21.0	7.5	68.0

(Figure 29)

(b)

(c)

(d)

(Figure 30)