Observation of grain growth in U-10Mo alloy

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Abstract. Both as-cast and homogenized wedge specimens of U-10 wt%Mo (U-10Mo) alloy were thermomechanically processed in both cold and hot rolled conditions and annealed at 700 °C for periods lasting up to 2 h. The hot rolled samples had a net strain of 70% whereas the cold rolled ones were subjected to 30% strain. Annealed microstructures were examined using electron backscatter diffraction in order to estimate the completeness of recrystallization and grain size, and these were mapped as a function of strain. In as-cast samples, the stored energy with strain increases but in the homogenized samples the stored energy remains constant with strain. This is due to a difference in the initial microstructure (before deformation) of the homogenized samples.

1. Introduction

For several years, research to develop alternate fuels for high-performance research reactors (HPRR) has been under way to replace high enriched uranium fuel with low enriched uranium (LEU) fuel. To achieve similar U-235 content with LEU, given the reactor space constraints, a higher bulk density of fuel is needed, which necessitates the use of monolithic metal alloy fuel. To achieve this density, an alloy that offers a good combination of irradiation performance, oxidation resistance, strength, and ductility is required for HPRR applications. Various uranium alloys have been evaluated for their favorable mechanical properties and susceptibility to oxidation and corrosion [1] [2]. One of the broad challenges faced is the swelling kinetics of this metallic fuel during the irradiation process. The fuel plates start to swell due to emission of inert gases and effects the integrity and shape of the sample which further deteriorates the irradiation performance.

Uranium alloyed with 10 wt% molybdenum (U-10Mo) shows improvement in these characteristics compared to similar alloys. Another key factor to improve the performance is to have stable phase of U-10Mo during irradiation. The nominal fabrication process for U-Mo alloys is to cast and quench the material to stabilize the metastable gamma (γ) phase. Casting in an inert gas is recommended to prevent the formation of oxides in the alloy. Rapid cooling is necessary to keep the alloy in the γ phase. A common technique used to accomplish this includes an homogenization step in which the alloy is brought to an elevated temperature between the eutectoid temperature (560°C) and the melting temperature for the alloy (1135°C to 1200°C). The alloy is held at temperature for an extended period to ensure that the metal has completely transformed to the γ phase and to enable an even distribution of the molybdenum throughout the sample. Uranium α and β phase have orthorhombic and tetragonal lattice structure respectively whereas Uranium Γ phase has BCC crystalline structure. BCC crystalline structure possesses less anisotropic properties which somehow improves the irradiation performance of this metallic fuel [2] [3]. However, several efforts are now under way to optimize the U-10Mo
processing parameters and Pacific Northwest National Laboratory (PNNL) has supported these activities.

Research is being performed on the microstructure of this material with modeling and experimental simulation to reduce the defects caused during and after the irradiation process [1-3]. With a motive to that, microstructure evolution is studied with processing of this alloy. The first set of samples includes study of recrystallization texture in U10Mo alloy with hot rolling and annealing process. Rolling and annealing texture is studied with quantified microstructure analysis. The second set of samples includes study of recrystallization behavior in wedge shaped annealed samples. Microstructure evolution is studied with respect to strain.

2. Experimental Procedure

All samples were prepared using standard metallographic procedures and final polishing was performed on a vibro-polisher using 0.05 μm colloidal silica suspension before EBSD/SEM measurements. Recrystallization behavior was studied in U10Mo wedge-shaped samples with post annealing process after deformation. Two sets of cast samples were prepared. One set is homogenized at 900°C for 48 hours and another set is left as cast only. Each set includes two samples out of which one is deformed with cold rolling and another one with hot rolling (at 650°C). Finally, each sample is post-annealed at 700°C for two hours. After deformation/rolling process, the wedge-shaped samples were flattened with accumulation of specific uniform strains i.e. 0.1 to 0.7. All strains were calculated by using engineering strain formula along the length of the sample. On average, the cold roll deformed samples were 22 mm long, whereas, the hot roll deformed samples were 30 mm long. Strain up to 0.3 and 0.7 were calculated in cold rolled and hot rolled samples, respectively. At higher strains chances of cracking increases during cold rolling. Therefore, the total length of the cold rolled samples was kept shorter than the hot rolled samples.

Sample processing is explained with the flow chart shown below. The sample name with its processing terminology is given below:

CH-700: Cast hold rolled post-annealed at 700°C
CC-700: Cast cold rolled post-annealed at 700°C
HH-700: Homogenized hot rolled post-annealed at 700°C
HC-700: Homogenized cold rolled post-annealed at 700°C

2.1. EBSD measurements

Sample preparation methods and EBSD parameters used were similar for the hot rolled and annealed samples. The total area scanned (for orientation maps) for EBSD analysis was approximately 300x900 μm² at each specific strain for all samples. Measurements were made over a regular hexagonal grid using a step size of four microns for cold rolled samples and two microns for hot rolled samples. Step size was decided as per the average grain size of the samples. Cold rolled samples have a larger mean grain size (>25μm) compared to hot rolled samples so the step size taken was larger (up to four microns). Specific strain positions were calculated by percentage deformation or engineering strain in all wedge-shaped samples.
In figure 1, dotted lines represent the initial shape of the sample before rolling. Θ is the wedge angle, Δl is the change in height, h is the final height of the sample and b is the distance of each deformed region, from the 0% deformed region. For example: 50% deformation has an engineering strain of 0.5 = \frac{\text{change in height}}{\text{original height}} = \frac{2\Delta l}{h+2\Delta l}. Δl calculated was finally used to calculate distance ‘b’ to find the X-coordinate of 50% deformation or 0.5 strain. This was done by assuming a right-angled triangle with Δl as the perpendicular distance and b as the base of the triangle. Distance b = \frac{\Delta l}{\tan \Theta} was calculated and the sample (SEM stage) was moved manually in the X direction from 0% deformation or zero strain. The wedge angle Θ is similar for all samples between 16~18⁰ and is calculated by using SEM images in photo editing software.

3. Results and Discussion

3.1. Microstructural analysis
Figure 2 shows the orientation maps (RD-ND planes) indexed as Y U-phase at different strain (0-0.7) of CC-700, HC-700, CH-700 and HH-700 samples. Black spots are either 2nd phase particles or points with CI<0.1. It was observed that the grain size drops with strain in all wedge-shaped post annealed 700°C samples, and this drop is due to recrystallization behavior of grains. In both CC-700 and HC-700 samples, the grain size drops from 100 to 30 microns approximately at 0.3 strain. Whereas in CH-700 and HH-700 samples, the grain size drops from 100 to 20 microns approximately at 0.7 strain. The drastic drop in grain size is seen at 0.2 strain in all samples as shown in figure 3 (a). It can be stated that the critical deformation required for recrystallization to begin is at these temperatures is 0.2 strain. In sample CH-700 and CC-700, bimodal mean grain size charts were observed at 0.2 strain as shown in figure 3 (b) and (c). Bimodal mean grain size represents the presence of deformed and recrystallized grains in cast samples at 0.2 strain. This signifies that in cast samples (at 0.2 strain), the deformed grains start getting recrystallized. However, no such bi-modal grain size charts were observed in homogenized samples at 0.2 strain. Near random textures were observed at all strain levels in all samples.

During recrystallization, the fraction of high angle grain boundaries tends to increase because the low angle grain boundary fraction resulting from the dislocation structure is consumed by the growing grains. From figure 4, the fraction of HAGBs in all samples is above 0.9 signifying that a majority of the grains are recrystallized at all levels of strain.

3.2. Kernel average misorientation
Kernel average misorientation (KAM) is the average local misorientation of a single point in relation to orientations of neighboring points. Based on the assumption that local misorientation gradients are generated by dislocations, the KAM, is therefore a measure of the stored energy in terms of dislocation density in the microstructure. The measure of kernel average misorientation as a quantitative value is used to characterize the distribution of dislocation density [4] [5]. The KAM value in this work is
calculated using a misorientation definition of less than 5° and the third nearest neighbors. As expected, this shows a similar trend compared to the GND (geometrically necessary dislocation) density in all samples (figure 5).

Figure 2: Orientation maps (RD-ND plane) indexed as U-ϒ phase at different strain (0-0.7) of (a) CC-700 (b) HC-700 (c) CH-700 (d) HH-700 samples. Black spots are either 2\textsuperscript{nd} phase particles or points with CI<0.1. The micron bar is 100 µm for all images.
From figure 6(a) and (c), there is no certain change in stored energy of homogenized (HH-700 and HC-700) samples. In homogenized samples, little drop in fraction of GOS<2° at 0.2 strain in HH-700 and at 0.1 strain in HC-700 is considered an outlier. However, in KAM value there is no significant change as shown in figure 6(e). Whereas in cast samples as shown in figure 6(b) and (d), the stored energy increases with strain up to 0.4 in hot rolled and up to 0.3 in cold rolled samples. Due to shorter length of cold rolled samples, the strain accumulated is only up to 0.3 so we cannot determine beyond that strain level.

From figure 7 (e) in sample CH-700 (cast hot rolled), the stored energy starts to drop from 0.4 and gets constant after 0.5 strain level. This study shows us that the critical deformation/ strain required for cast hot rolled samples is in between 0.4 and 0.5 for grains to get fully recrystallized with post-annealing process (at 700°C). After 0.5 strain, the deformation gets constant, which denotes that concurrent recovery at such high strains and temperature probably did not increase the stored energy (with deformation) [6]. In sample CC-700 (cast cold rolled), the stored energy at 0.3 strain is enough for grains to get fully recrystallized. Study of KAM maps concluded that the stored energy is higher in cast samples as compared to homogenized samples. Due to high stored energy, the grain refinement is more at higher strains in cast samples as the mean grain size is reduced to 16 microns (at 0.7 strain). Whereas, in homogenized samples the stored energy is little (compared to cast samples) so the mean grain size drops to 20 microns at a maximum strain of 0.7.
3.3. Grain orientation spread

A suitable value of GOS is required to differentiate the deformed from the recrystallized regions in the sample. The GOS value taken was less than 3° and grain size greater than 9μm² [7-10]. In figure 8(a), the decrease in grain size with strain is due to recrystallization. Fraction of GOS (<2°) in sample HH-700 is up to 0.8 at all strain levels (except at 0.2 which is an outlier) whereas in sample CH-700, the fraction of GOS (<2°) reaches to 0.8 after 0.5 strain. The critical deformation required for recrystallization phenomenon to occur was studied. The minimum strain required is 0.5 for grains to get fully recrystallized in CH-700 (cast hot rolled) sample. Whereas in CC-700 (cast cold rolled) sample, the minimum strain required is 0.3 for grains to get fully recrystallized. It can also be concluded that the recrystallization behavior depends on the initial microstructure of the material. Before deformation, the homogenized samples had fully strain-free (recovered) microstructure but in cast samples the microstructure was not strain-free. So, in homogenized samples with post-annealing process (at 700°C), grains are fully recrystallized at all strains (except few strains which are outliers as explained earlier). In cast hot rolled sample, grains start to get fully recrystallized after 0.5 strain. This is probably because of concurrent recovery occurred during hot rolling at and beyond 0.5 strain [6]. Whereas in cast cold rolled sample the stored energy is too high that even at 0.3 strain the grains get fully recrystallized.

![Figure 4: Fraction of HAGB>15° vs Strain of all samples](image)

![Figure 5: KAM vs Strain of all samples. The threshold value taken for misorientation is maximum of 5° with nearest 3rd neighbor](image)
Figure 6: KAM maps of (a) HH-700 (b) CH-700 (c) HC-700 (d) CC-700 samples at all strains (0-0.7)

Figure 7: (a) GOS (<2°) maps of all samples at different strains
4. Conclusions

In CH-700 (cast and hot rolled) sample, the recovery and recrystallization phenomenon overlapped up to 0.4 strain wherein partial recrystallization occurred. It is anticipated that concurrent recovery, at and above 0.5 strain, diminished the increase in stored energy with deformation in sample CH-700. The fraction of GOS (<2°) increases to 0.8 after 0.5 strain and remains constant with further deformation. Values of KAM and the fraction of GOS (<2°) remain constant after 0.5 strain and this denotes that there was no further stored energy in the structure after 0.5 strain. Whereas in cast cold rolled samples, the fraction of GOS (<2°) is greater than 0.8 at 0.3 strain. The stored energy in the cast cold rolled sample at 0.3 strain is about double compared to the cast hot rolled sample. In the homogenized hot rolled and cold rolled samples, the recrystallization fraction is greater than 0.8 at all strain levels, except one, which is considered as an outlier. There is no strong evidence for the small drop in the fraction of GOS (<2°). However, as explained it could be anticipated that due to shear deformation during hot rolling (at that strain), the critical deformation (required for recrystallization phenomenon) did not occur.

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