In situ High-Energy X-ray Diffraction Study of Tensile Deformation of Neutron-irradiated Polycrystalline Fe-9%Cr Alloy

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Abstract

The effect of neutron irradiation on tensile deformation of a Fe-9wt.%Cr alloy was investigated using *in situ* high-energy synchrotron X-ray diffraction during room-temperature uniaxial tensile tests. New insights into the deformation mechanisms were obtained through the measurements of lattice strain evolution and the analysis of diffraction peak broadening using the modified Williamson-Hall method. Two neutron-irradiated specimens, one irradiated at 300°C to 0.01 dpa and the other at 450°C to 0.01 dpa, were tested along with an unirradiated specimen. The macroscopic stress–strain curves of the irradiated specimens showed increased strength, reduced ductility and work-hardening exponent compared to the unirradiated specimen. The evolutions of the lattice strain, the dislocation density and the coherent scattering domain size in the deformation process revealed different roles of the submicroscopic defects in the 300°C/0.01 dpa specimen and the nanometer-sized dislocation loops in the 450°C/0.01 dpa specimen; the dislocation loops were more effective in dislocation pinning. While the work hardening rate of stage II was unaffected by irradiation, significant dynamic recovery in stage III in the irradiated specimens led to the early onset of necking without stage IV as observed in the unirradiated specimen.

Keywords

Neutron irradiation, tensile deformation, work hardening stages, *in situ* X-ray diffraction

1. Introduction

High-Cr (9-12 wt.%) ferritic-martensitic steels are important candidate materials for in-core and out-of-core applications in advanced nuclear energy systems. This class of alloys has an excellent resistance to irradiation-induced swelling, high thermal conductivity, low thermal expansion, and low cost relative to austenitic stainless steels [1]. One of the major concerns for their application in nuclear reactors is irradiation-induced hardening and embrittlement at low temperatures [2]. Extensive studies have been conducted to understand the irradiation embrittlement mechanisms and to improve their performance under irradiation. However, a fundamental understanding of radiation-induced microstructural changes and consequent performance degradation in high Cr steels is still largely lacking [3]. Due to the complex microstructures of these engineering alloys, studies of simple Fe-Cr model alloys can elucidate the roles of various microstructural features and develop a better understanding of their collective effects in these alloys.

The effect of irradiation on the mechanical properties of ferritic Fe-Cr alloys has been investigated extensively through experiments [4-12] and modelling [13-19]. It is found that the irradiation-induced hardening and the reduction of ductility are significant even at low doses. In materials irradiated to high doses, plastic instability and dislocation channeling was observed [4, 7, 20, 21]. While the increase in yield strength in irradiated body-centered-cubic (bcc) metals and alloys can be understood by source hardening and dispersed barrier hardening models [22], the work hardening mechanisms in irradiated materials is not as clear. Classical theories characterize

the work hardening process into well-defined stages dominated by different mechanisms [23]: stage I only occurs in single crystals during single slip, commonly referred to as easy glide; stage II (also known as athermal hardening) is characterized by a constant hardening rate which is largely temperature- and rate- independent; stage III is characterized by a hardening rate that decreases monotonically with deformation and which is temperature- and rate- sensitive, resulting from a dynamic recovery process at large strains; stage IV is a continued work hardening at large strains, and has been found in a variety of polycrystals under many deformation modes [24]. Several different models exist to explain these deformation processes, and have successfully correlated the mechanical properties to microstructural features in terms of dislocation density (and sometimes including other features such as grain size) [23-25]. However, to the authors' knowledge, such efforts have largely excluded irradiated materials, at least partially due to their relative complexity in terms of microstructure and deformation modes.

High-energy, high-brilliance synchrotron X-rays allow *in situ* microstructural characterization under complex sample environment because of their ability to penetrate deep into materials non-destructively. X-ray diffraction (XRD) in particular can reveal the internal stress distributions in each crystalline phase as well as dislocation arrangements. A number of nuclear structural materials have been investigated *in situ* using high-energy synchrotron XRD to measure millimeter-sized specimens subject to thermo-mechanical loading to understand their deformation mechanisms [26-31]. This technique has largely been limited to unirradiated specimens due to the lack of capability for handling radioactive specimens. Over the past few years, Argonne National Laboratory (ANL) has been developing an *in situ* Radiated Materials (*iRadMat*) apparatus capable of conducting *in situ* thermo-mechanical tests under controlled environment on mm-sized activated specimens at a public beamline at the Advanced Photon

Source (APS) [32]. Several high-energy synchrotron X-ray techniques such as wide-angle X-ray scattering (WAXS), small-angle X-ray scattering (SAXS), far-field high-energy diffraction microscopy (ff-HEDM), and tomography can be employed to characterize activated specimens subject to thermo-mechanical loads [32].

This paper reports a set of *in situ* experiments on neutron-irradiated Fe-9wt.%Cr specimens using the *iRadMat* apparatus. Two low-dose neutron-irradiated Fe-9wt.%Cr specimens were examined together with their unirradiated counterpart. Room-temperature tensile tests were conducted *in situ* with X-ray diffraction measurements. The distinct effects of neutron irradiation on the deformation mechanisms and roles of different types of irradiation-induced defects were discussed.

2. Experimental Procedure and Data Analysis Methods

The material investigated was a ferritic Fe-9wt.%Cr model alloy with the chemical composition of Fe-9.04wt.%Cr-0.005wt.%C. It has an average grain size of 180 μm (determined from electron backscatter diffraction (EBSD) measurement) and no significant preferred orientation [33]. The geometry of the tensile specimen is shown in Fig. 1. The nominal gauge dimensions of the tensile specimen are $5 \times 1.2 \times 0.5$ mm and the total length is 16 mm.

Neutron irradiation was performed at the Advanced Test Reactor (ATR) at the Idaho National Laboratory. Two irradiated specimens were examined in this study, one irradiated to 0.01 dpa at 300°C and the other irradiated to 0.01 dpa at 450°C, both under a dose rate of 1×10^{-7} dpa/s, where dpa is the displacement per atom, a measure of the irradiation dose. The activity of each sample was less than 5 mR/h at a distance of 30 cm. Before being transported to the APS, each irradiated specimen was individually mounted at an ANL radiological facility into a

gripping device and then enclosed in a double-layered Kapton containment for radiation safety purposes, as shown in Figure 1. Irradiated sample assemblies were transferred in lead-shield containments to the APS 1-ID-E beamline. The unirradiated Fe-9wt.%Cr specimen was mounted in the same type of gripping device without the Kapton enclosure. At the beamline, the gripping device was installed into the *iRadMat* apparatus. The *iRadMat* apparatus has 5-mm thick tungsten radiation shielding plates surrounding the sample and tungsten shielding shutters for two X-ray windows and two viewing ports. Figure 2 is a schematic of the *in situ* experimental setup for an activated specimen at the APS 1-ID-E beamline. Tensile tests were performed at room temperature at a nominal strain rate of $2 \times 10^{-5} \text{ s}^{-1}$. The WAXS and SAXS signals were recorded simultaneously and continuously during the tensile test. Four GE-RT41 amorphous silicon based integrating area detectors, designated GE1, GE2, GE3, and GE4, form an array to record the WAXS signals, while the SAXS signals were captured by a Pixirad CdTe counter based area detector.

The X-ray energy was 122.5 keV (wavelength 0.01011 nm). The WAXS detector calibration was performed with a CeO₂ NIST standard (SRM 674b). The sample to detector distance was nominally 2635 mm, which led to a 2θ coverage of 15° on the WAXS detectors, and 9 Debye-Scherrer rings, denoted hereon as {hkl}s, were recorded. The size of the incident beam was 0.1 mm $\times$ 0.1 mm. Because of the coarse-grained structure and small size of the irradiated tensile specimen, the illuminated volume contained only a few grains, which makes the diffraction data analysis with traditional powder-averaged techniques difficult. To obtain higher grain statistics in the diffraction data, the specimen was translated along X_L with a step size of 0.2 mm and Y_L with a step size of 0.4 mm with respect to the beam to interrogate 30 volumes of material along the gauge length. The resulting 30 diffraction images per GE detector were summed to produce a

more powder-like data from approximately 100 grains. Due to the slow strain rate of the tensile test, the accumulated strain during these 30 exposures is about 1% strain, yielding a sufficient data acquisition rate. During tensile deformation, the constituent grains develop intragranular misorientation and form subgrains, thereby improving the diffraction statistics with increasing deformation.

Shown in Figure 3 is a series of 2D diffraction patterns acquired by one of the WAXS detectors (GE1) at different deformation levels of the unirradiated specimen, each of which is a sum of the diffraction images from 30 volumes. As can be seen, prior to in-situ deformation even 30 volumes cannot produce a powder-like pattern, because of the small number of interrogated grains. With increasing strain, more powder-like data can be obtained due to broadening of diffraction spots, particularly in the azimuthal direction. Later in the discussion of plasticity, we chose the 4% strain as the reference state to minimize the data scatter from insufficient statistics at lower strain levels. The 1D profiles were obtained by integrating over $\pm 15^\circ$ azimuthal angle of the WAXS patterns in GE1 (marked by the solid lines in Figure 3) in the direction nominally parallel to the tensile loading axis (marked by the dotted lines in Figure 3). The same was done for the patterns recorded by GE4 for the direction nominally perpendicular to the loading axis (transverse direction). The 1D diffraction profiles were then fitted using pseudo-Voigt functions yielding peak positions in the two directions. For peak broadening analysis, it was assumed that the broadening is homogeneous in the entire azimuthal range (i.e. no directional preference) and the entire azimuths covered by GE1 and GE4 were used. Since the diffraction rings deviate from circles due to deviatoric strains, the patterns were not integrated over the entire azimuth initially; instead, they were first caked into 20° azimuthal slices, and then integrated into individual 1D profiles. Those profiles were fit with pseudo-Voigt functions to obtain the integrated intensity,

position and widths (represented by full-width at half maximum or FWHM) of each peak of interest. The FWHM results were summed over all measured azimuthal slices, using integrated intensity weighting. This maximized the number of grains in the FWHM analysis resulting in improved goodness of fits using the modified Williamson-Hall method, as will be detailed in section 4.3. The instrument contribution to the broadening ($B_{\text{instrument}}$) was quantified using a LaB6 NIST standard (SRM 660a). It was subtracted from the measured FWHM (B_{total}) to get the actual sample broadening using $B_{\text{sample}}^2 = B_{\text{total}}^2 - B_{\text{instrument}}^2$, as a result of deconvolution because the line profile is close to Gaussian (fraction of Gaussian in the pseudo-Voigt fitting is above 0.8 for most peaks).

3. Results

This paper presents the results from *in situ* WAXS measurements of the uniform tensile deformation up to the ultimate tensile strength (UTS). These coarsen-grained single-phase specimens did not scatter strongly in SAXS during uniform deformation. The *in situ* WAXS/SAXS data for the necking stage will be presented in future publications.

3.1 Macroscopic stress-strain curves

Figure 4 compares the stress-strain curves for the unirradiated and irradiated samples, with the solid symbols representing engineering stress-strain data and the open ones for the corresponding true stress-strain data (until the maximum load). Irradiation-induced hardening and reduction in ductility are significant even at this low dose. The 0.2%-offset yield stress, the UTS, the work hardening exponent, and the uniform and total elongations for each sample are listed in Table 1. Hardening is more pronounced in the 450°C/0.01 dpa sample than in the 300°C/0.01 dpa sample.

3.2 *In situ* WAXS during tensile tests

WAXS measurements of the undeformed samples gave a lattice parameter of 2.8695 ± 0.0006 Å for the unirradiated specimen, 2.8720 ± 0.0009 Å for the 300°C irradiated and 2.8722 ± 0.0008 Å for the 450°C irradiated specimens, respectively. These numbers were obtained from averaging the lattice parameters given by all the $\{hkl\}$ families (converted from the fitted d-spacings) in each sample. The relatively large uncertainty, compared to the instrumental resolution of 1×10^{-4} strain from fitting the CeO_2 standard, is due to the large scatter in $\{hkl\}$ -specific lattice parameters primarily from the residual strains.

Figure 5 (a) shows the lattice strains ε_l as a function of the true stress up to the UTS for six $\{hkl\}$ reflections in the loading direction and in the transverse direction in the three samples, calculated using $\varepsilon_l = (d - d_0)/d_0$, where d_0 is the $\{hkl\}$ -specific d-spacing prior to deformation and d is the instantaneous d-spacing during deformation. Figure 5 (b) shows the lattice strains in the loading direction as a function of the true strain. In both figures the open symbols are for the loading direction and the solid symbols are for the transverse direction. As anticipated, the lattice strains measured along the loading direction are tensile and the lattice strains measured along the transverse direction are compressive for all three samples as a result of Poisson's effect. The ratio between the lattice strains along the loading and the transverse directions is approximately 0.3 in the elastic regime.

Although the macroscopic stress-strain curves for the three samples show similar deformation behavior, the X-ray measurements of the lattice strain show different characteristics, either when comparing different reflections within one sample or comparing between samples for a given reflection. For a given sample, the load redistribution between grains is manifested by

the elastic anisotropy, i.e. the difference in stiffnesses of the reflections [34]. Figure 5 shows that the {200} lattice strains are the largest while the {222} lattice strains are the smallest in the loading direction before the macroscopic yield stress. The lattice strains for other crystallographic planes mostly lie between the {200} lattice strains and the {222} lattice strains. These trends are consistent with the commonly recognized stiffest and the most compliant directions in bcc metals. In the transverse direction, this relationship largely holds true in the 450°C irradiated sample, but is not so clear in the unirradiated and 300°C irradiated samples, which could be attributed to the uncertainties in determining d_0^{hkl} of the coarse-grained samples. It is also noted that lattice strains in the loading and the transverse directions increase monotonically up to the macroscopic yield stress where linearity breaks down and inflections of the lattice strain vs. the macroscopic stress are evident. This is an indication that the crystal stresses are redistributing or reorienting [34, 35].

When comparing between the samples, it is observed that the lattice strain evolution in the plastic deformation regime shows very different characteristics in the 450°C irradiated sample from the unirradiated sample, which is more evident in Figure 5 (b), where the deformation state is presented in terms of macro strain rather than stress for the loading direction. For instance, in the 450°C irradiated sample, after ~9% macroscopic strain the lattice strains all decrease, which is also observed in the 300°C irradiated sample after ~12.4% strain, but not in the unirradiated sample. The phenomena can also be observed in Figure 5 (a) by close examination. These responses are discussed in terms of the interactions between irradiation- and deformation-induced defects in the discussion section.

Figure 6 shows the full-width half-maximum (FWHM) values of the diffraction peaks as a function of true strain (starting from ~4%) for the same six reflections plotted in Figure 5. In

general, the FWHM values increase with macroscopic strain, indicating a high level of nano-scale activities due to plastic deformation. The modified Williamson-Hall (W-H) analysis was performed to understand the FWHM evolution. According to the diffraction theory, the size broadening is independent of the diffraction vector length g_{hkl} ($g_{hkl} = \frac{1}{d_{hkl}}$, d_{hkl} is the d-spacing of the $\{hkl\}$ plane) while strain broadening increases with g_{hkl} [36]. The modified W-H theory takes into account the anisotropy in strain broadening due to geometric factors controlling dislocation movement on different $\{hkl\}$ planes and relates the strain broadening *linearly* with g_{hkl} [37] (under the condition that the diffraction angle θ is small so that $\cos \theta \approx 1$, which is the case here with high-energy X-rays). The slope of this linear relationship is proportional to the square root of the mobile dislocation density ρ_l (in particular, the dislocations that do not belong to sharp boundaries [38]), and the intercept is inversely proportional to the coherent scattering domain size D by the Scherrer equation $\beta = 0.9\lambda/D$, where β is the intercept and λ is the wavelength. The anisotropy due to geometry factors is accounted for by including $\{hkl\}$ -dependent dislocation contrast factors C_{hkl} using $g_{hkl} \cdot C_{hkl}^{0.5}$. Values for C_{hkl} for edge type $\{110\}\langle 111 \rangle$ and screw type $\langle 111 \rangle$ dislocations in BCC-Fe can be calculated using the software ANIZC [39] with input parameters specified in [26]. The effective C_{hkl} used in the analysis is a linear superposition of values from these two types of dislocations weighted by their density fraction. To find the optimum C_{hkl} , the modified W-H fitting using the C_{hkl} with the screw/edge ratio ranging from 0 to 1 was performed with 0.01 step size, and the C_{hkl} that gave the highest R-square value was selected, giving the optimum edge/screw dislocation fraction. In this way, the evolution of the edge/screw dislocation fraction can be tracked with deformation.

The slopes, the coherent scattering domain sizes and the screw dislocation fractions obtained from the modified W-H analysis are plotted as a function of true strain for the three samples in

Figure 7. The R-square values from all the fittings are above 0.92 and most of them are above 0.95. Domain size values at low deformation levels and greater than 1.5 μm are removed from the plot, because they may not be statistically accurate.

The general observation in Figure 7 is that the deformation in each sample progresses in distinct stages. For example, in the unirradiated sample, between 7.5% and 11.5% strain, the dislocation density (reflected by the slope values in Figure 7 (a)) increases slowly, the screw dislocation fraction is almost unchanged, but the domain size decreases quickly. Beyond 11.5%, the dislocation density increases quickly as well as the screw dislocation fraction, but the domain size reaches a plateau. In the 450°C irradiated sample, before 9% strain the dislocation density increases quickly as well as the screw dislocation fraction, while the domain size remains relatively stable. After 9% strain, the dislocation density decreases slowly as well as the screw fraction, while the domain size decreases quickly. In the discussion section, these W-H analysis results are revisited together with the lattice strain data, and the implications on the plastic deformation mechanisms and the effect of irradiation will be discussed.

The coherent scattering domain size at the end of the uniform deformation in the unirradiated sample given by the modified W-H analysis was verified by post-mortem TEM examination of the unirradiated specimen. A TEM sample was prepared using the Focused Ion Beam lift-out method. It was extracted from the uniformly deformed gauge section of the fractured unirradiated sample in cross section, with the long edge parallel to the gauge width. Figure 8 shows a bright-field image of cells/subgrains formed during tensile deformation. The mean size of dislocation cells measured by TEM is about 0.5 μm , which is consistent with the coherent scattering domain size, 0.5 μm given in Figure 7 (b).

4. Discussion

1). Barrier strength of irradiation-induced defects

Neutron irradiation significantly increases the yield stress and the ultimate tensile strength of the Fe-9wt.%Cr alloy as shown in Figure 4, which can be understood by the pinning of the dislocation motion by the irradiation-induced defects. TEM observations of microstructures of as-irradiated samples [33, 40] showed no visible irradiation-induced defects after neutron irradiation to 0.01 dpa at 300°C, while small dislocation loops with the average size of 3.8 nm and a density of $2.9 \times 10^{21}/\text{m}^3$ in the 450°C/0.01 dpa specimen. The majority of the dislocation loops have the Burgers vector of $a\langle 100 \rangle$. Atom probe tomography showed no phase separation in either of the two irradiated specimens [33]. WAXS measurements of the samples before tensile deformation showed that irradiation resulted in an increase in the lattice constant of 0.0025 Å in the 300°C irradiated sample and 0.0027 Å in the 450°C irradiated sample, implying that the dislocation loops are predominantly interstitial-type and the irradiation-induced vacancies may form sub-microscopic voids which do not affect the lattice constant [41].

It is widely recognized that dislocation loops can pin the dislocation motion by either contact or elastic interaction, which leads to an increase in the yield stress and a reduction in ductility ([42]). The barrier strength of dislocation loops, α can be estimated using the dispersed barrier model [43]:

$$\Delta\sigma = M\alpha\mu b\sqrt{Nd} \quad (1)$$

where $\Delta\sigma$ is the yield stress increase after irradiation, M is the Taylor factor (3.06 for BCC), μ is the shear modulus (8.3×10^4 MPa at room temperature), b is the magnitude of the loop Burgers vector, and N and d are irradiation-induced loop density and diameter, respectively. For the 450°C irradiated sample, the barrier strength α of the TEM-visible dislocation loops was

estimated to be ~ 0.68 for the 165 MPa increase in the 0.2%-offset yield stress determined from the macroscopic stress-strain curves. In this study, the application of *in situ* WAXS technique provides a method in estimating the micro-yield stress using the lattice strain – macroscopic strain curve. More specifically, for both the unirradiated sample and the 450°C irradiated sample, the micro-yield stress was determined from the first deviation from linearity in the lattice strain-true strain curves in Figure 5 (b). The micro-yield stress was estimated to be 101 MPa for the unirradiated sample and 197 MPa for the 450°C irradiated sample. Therefore, the irradiation-induced micro-yield stress increase was 96 MPa, giving an estimated α of ~ 0.40 , which is significantly lower than the value calculated from the 0.2%-offset yield stress. Zinkle and Matsukawa [22] attributed the unusually high barrier strengths obtained from macroscopic stress-strain curves for visible defect clusters to submicroscopic nanovoids in bcc metals. Our work shows that the α value is also sensitive to the measurement of the yield stress. In the 300°C/0.01dpa specimen, almost no loops were visible under TEM, and therefore a lower yield stress increase compared with the 450°C irradiated sample. Its increase in yield strength may be explained by the source hardening from the locking of grown-in dislocations by the submicroscopic defects [44].

2) Effect of irradiation on lattice strain evolution during plastic deformation

The lattice strain evolution in the plastic deformation regime shows significant differences between the unirradiated and the irradiated samples, as shown in Figure 5. To better reveal the lattice strain evolution in the plastic regime, Figure 5 (b) was re-plotted in Figure 9 by taking the d-spacings at $\sim 4\%$ strain in each reflection as the reference d-spacings to calculate the $\{hkl\}$ -specific lattice strains. This eliminates the elastic anisotropic effect and reduces the data scatter from the diffraction patterns not being “powder-like” at lower deformation levels. We denote

these lattice strains as *pseudo* lattice strains and they are plotted as a function of plastic strain in Figure 9 in solid symbols.

The equivalent elastic strains were calculated using the method A reported by Daymond [45], which weighs the strain contribution from individual reflections by α_{hkl} , defined as:

$$\alpha_{hkl} = T_{hkl} m_{hkl} E_{hkl} / \bar{E}, \quad (2)$$

where T_{hkl} is the texture index, which is unity for the untextured alloy in this study, m_{hkl} is the multiplicity of the {hkl} reflection, E_{hkl} is the diffraction elastic constant for the {hkl} reflection, which is measured as the slope in the elastic deformation regime in the lattice strain- stress curve in Figure 5 (a), and $\bar{E}$ is the bulk modulus. The equivalent elastic strain $\bar{\varepsilon}$ is then calculated as:

$$\bar{\varepsilon} = \frac{\sum_{hkl} \alpha_{hkl} \varepsilon_{hkl}}{\sum_{hkl} \alpha_{hkl}}, \quad (3)$$

where ε_{hkl} is the instantaneous lattice strain of the {hkl} reflection. The equivalent elastic strain is plotted as a function of plastic strain in Figure 9 in open symbols. For the unirradiated sample it shows a steady increase with plastic strain, despite the fact that the {hkl}-specific lattice strains are largely anisotropic. In contrast, for the 300°C irradiated sample, both the {hkl}-specific lattice strains and the equivalent elastic strain show a decrease starting at 12.3%. For the 450°C sample this synchronized decrease start at 8.5%. It is also noted that the equivalent elastic strain and {321} lattice strain show a similar trend with respect to the plastic strain in all three samples, implying that the 321 reflection is suitable for characterizing the bulk behavior of this alloy.

3) Work hardening mechanisms

In situ WAXS data provides new insights into the work hardening mechanisms in the Fe-9wt.%Cr alloy, as revealed by the lattice strain data and the results from the modified W-H analysis. To better understand the WAXS data, Figure 10 is plotted by reorganizing the content in Figure 7 and Figure 9, with an emphasis on changes with plastic strain in individual samples. Note that the slope from the W-H fitting in Figure 7 is replaced by $A\rho^{0.5}$ in Figure 10, where A is a constant and ρ the dislocation density. Based on the modified W-H theory [46], $A = \lambda \left(\frac{\pi m^2 b^2}{2} \right)^{\frac{1}{2}}$, where λ is the X-ray wavelength, m is a constant depending on the effective outer cutoff radius of dislocations, and b is the magnitude of the Burgers vector. The value of m is not being pursued in this paper and A is assumed to be the same for the three samples. Dotted vertical lines in Figure 10 define deformation regimes: for the unirradiated sample, two lines are proposed at 7.3% and 11.2% plastic strain; for the 300°C irradiated sample at 12.3%, and for the 450°C irradiated sample at 8.5%, respectively.

The first deformation regime in all three samples has strikingly similar characteristics: 1) the $\{hkl\}$ -specific lattice strains first show heterogeneous behaviors followed by a synchronized increase with the transition at 6.1% for unirradiated, 10% for 300°C irradiated and 7.2% for 450°C irradiated samples, respectively; 2) dislocation density shows a rapid increase with plastic strain at the same rate, i.e. $\frac{dA\rho^{0.5}}{d\varepsilon_p} = 2.0 \times 10^{-5}$ in all three samples, as marked by the green dotted lines; 3) coherent scattering domain size is relatively unchanged in the two irradiated samples with some data scatter. The lattice strain increase can arise from the dislocation pile-up at grain boundaries, and the decrease may be attributed to the relaxation of intergranular stresses from slip transfer across grain boundaries [47]. The constant slope in the

dislocation density – plastic strain curve indicates a constant work hardening rate ($\frac{d\sigma}{d\varepsilon} \propto \frac{d\sqrt{\rho}}{d\varepsilon}$), i.e. “stage II” athermal work hardening [23, 25], and the work hardening rate was unaffected by irradiation. The minimal change in the coherent scattering domain size implies insignificant dislocation patterning and dislocation substructure formation.

The effect of irradiation is mostly exemplified by a significantly higher density of dislocations and fraction of screw dislocations in the 450°C irradiated sample relative to the unirradiated sample in stage II. Before transitioning to next deformation stage, the dislocation density is the highest in the 450°C irradiated sample ($A\rho^{0.5} = 3.81 \times 10^{-4}$), lower in the 300°C irradiated sample ($A\rho^{0.5} = 3.57 \times 10^{-4}$), and the lowest in the unirradiated sample ($A\rho^{0.5} = 2.92 \times 10^{-4}$). The higher density of mobile dislocations in the irradiated specimens may be understood by considering the interactions of dislocations with irradiation-induced defects. As revealed by TEM during *in situ* ion irradiation and straining of an stainless steel specimen [48], dislocations moved relatively slowly and in a wave-like motion, in contrast to fast and smooth dislocation motion observed in a comparable unirradiated specimen. This difference was attributed to pinning effects from the irradiation induced defects. The jerky flow in the irradiated specimen increased the overall length of dislocation segments. The higher fraction of screw dislocations observed in the 450°C irradiated specimen in Figure 7 (c) can also be explained by dislocation – irradiation-induced loop interactions. When the loop pin a mobile dislocation, the line bows out forming screw segments, resulting an increase in length (i.e. an increase in line density) and the screw dislocation fraction. This dislocation behavior has been demonstrated in simulations of various situations of dislocation-loop interactions [17, 18, 42, 49]. The submicroscopic radiation defects in the 300°C specimen seem to have minimal effect on the

screw dislocation fraction, as seen in Figure 7 (c), probably due to the defect annihilation by dislocation lines [42].

While the first regimes in all samples indicate active dislocation multiplication with little effect on the coherent scattering domain size, the second regimes is manifested by significant reductions in coherent scattering domain size in all three samples. The lattice strains in the two irradiated samples show synchronized decrease but are heterogeneous in the unirradiated sample. The dislocation density increases slightly in the unirradiated, while decreases rapidly in the 450°C irradiated sample and relatively unchanged in the 300°C irradiated samples. Those phenomena suggest active dislocation cell formation and dynamic recovery, characteristic of working hardening “stage III”

To better understand the effect of neutron irradiation on dislocation density evolution, dislocation cell formation and their consequent work hardening behavior, the following empirical relationship is considered [50]:

$$\sigma = \sigma_0 + \alpha\mu b\rho^{0.5} + K\mu b/D \quad (4a)$$

where σ_0 is the friction stress, α and K are the constants, μ is the shear modulus, b is the magnitude of the Burgers vector, and D is the cell size. The work hardening rate is deduced as:

$$d\sigma/d\varepsilon = \alpha\mu b(d\rho^{0.5}/d\varepsilon) - K\mu b/D^2(dD/d\varepsilon) \quad (4b)$$

Stage II is dominated by the first term in Eq. (4b), which is the constant in all three specimens. In stage III, while cell formation occurred in both the unirradiated and the 450°C irradiated specimens at a similar rate, the dislocation density continues to increase (though at a much lower rate than in stage II) in the unirradiated specimen, as oppose to a decrease in the 450°C irradiated specimen, which leads to the overall reduced work hardening rate and an early

onset of necking in the 450°C irradiated specimen. This greater dynamic recovery in the 450°C irradiated specimen may be attributed to its higher screw dislocation fraction and the resultant extensive cross-slip of screw dislocations. In the 300°C irradiated specimen, the “stage II” linear work hardening was extended to a larger strain, ~12%. Due to the lack of data points in stage III, the change in work hardening rate in the 300°C irradiated sample is speculated to be similar to the 450°C irradiated specimen, as they both have the synchronized reduction in lattice strains and the cell size.

After the second deformation regime (i.e. stage III), the two irradiated samples entered the necking stage. In contrast, the unirradiated sample exhibited further work hardening, showing a third regime, characterized by the rise in dislocation density and the plateau in domain size. The rate of dislocation density increase is smaller compared to that of the first regime. This may indicate the sample entered the work hardening stage IV at large strains. In the study of fcc metals, Rollett et.al. [23] explained the mechanism for stage IV being the dislocation debris accumulating inside the dislocation tangles or loose cell walls resulted from stage III. Their model predicted a constant strain hardening rate (i.e. a constant $\frac{d\sqrt{\rho}}{d\varepsilon}$), and its magnitude is smaller compared to that of the stage II. The constant strain hardening rate was also reported in X-ray line profile analysis by Ungar and co-workers [51, 52]. Estrin et.al. [53] explained stage IV as a net result from the decreasing hardening from the cell walls as they became thinner and the increasing hardening from the accumulation of dislocations from the cell interiors, and predicted a decreasing hardening rate in stage IV. Our results show that the microstructural domain size remains constant in this stage while the hardening rate is also a constant, the value of which is about half of the rate in stage II, significantly higher than the prediction by Rollett et.al. [23]. The fact that the two irradiated samples exhibited localized deformation without the

stage IV implies that the interaction between the dislocations and the irradiation-induced defects exhausts work hardening capability, resulting in the onset of instability at large strains.

While the macroscopic stress-strain curves in Figure 4 are important for identifying the overall effect of the complex deformation processes, *in situ* X-ray diffraction is a sensitive technique which enables us to probe the mechanical deformation processes within a polycrystalline aggregate, revealing the microscopic mechanisms that are masked in the bulk response. In the effort to understand the effect of neutron irradiation to mechanical properties, this technique is demonstrated to be particularly valuable.

Conclusion

Characterization of room temperature tensile deformation processes of an unirradiated and two low-dose neutron-irradiated Fe-9wt.%Cr polycrystalline samples were performed using *in situ* high-energy synchrotron X-ray diffraction techniques. In addition to the observations from the macroscopic stress-strain curves of irradiation-induced hardening, reduction of ductility and work hardening exponent, the microscopic deformation mechanisms were probed by performing X-ray line profile analysis to obtain the lattice strains, the dislocation densities and the domain sizes as a function of macroscopic strain. The calculation of the barrier strength of the irradiation-induced dislocation loops was found to be sensitive to the determination of the micro-yield point. Work hardening stages were identified for the deformation processes: in the unirradiated sample, stage II athermal hardening, stage III dynamic recovery and stage IV at large strains were present; in the two irradiated samples, only stage II and stage III were present. Irradiation-induced nm loops pinned the dislocation motion, resulting in a higher dislocation density in stage II and a higher screw dislocation fraction, which could contribute to the fast

recovery rate in stage III. This study demonstrates that the *in situ* diffraction measurements using high energy synchrotron X-rays can provide great insights into the deformation mechanisms in neutron-irradiated materials.

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Table 1. Tensile properties obtained from the macroscopic stress-strain curves

Sample	0.2% offset Yield stress (MPa)	Ultimate tensile strength (UTS) (MPa)	Work-hardening exponent	Uniform elongation (%)	Total elongation (%)
Unirradiated	115	222	0.380	20.8	28.7
300°C/0.01dpa	200	282	0.254	16.6	18.4
450°C/0.01dpa	280	338	0.261	16.1	22.7

Figure 1. The sample geometry and the sample-grip assembly with a double-layered Kapton containment.

Figure 2. *In situ* WAXS/SAXS setup with the *iRadMat* apparatus interfaced with the MTS load frame at beamline 1-ID-E at the APS.

Figure 3. 2D diffraction patterns from the unirradiated sample on the GE1 detector at different deformation levels. Each pattern represents the summation of 30 diffraction images. The solid lines in each pattern mark the azimuthal range for integration, and the dotted line shows the projection of the loading direction onto the detector plane.

Figure 4. Stress-strain curves obtained from *in situ* room-temperature tensile tests of the unirradiated and the two irradiated Fe-9wt.%Cr specimens. Solid symbols denote the engineering stress-strain data. Open symbols denote the true stress-strain data.

Figure 5. (a) Lattice strain evolution as a function of true stress from six {hkl} reflections in the three samples, showing for both the loading direction (open symbols) and the transverse direction (solid symbols). (b) Lattice strain evolution as a function of true strain for the loading direction.

Figure 6. Evolution of FWHM as a function of true strain for six {hkl} reflections in the three samples.

Figure 7. Results from modified Williamson-Hall analysis of the three samples as a function of true strain: (a) the slope ($\propto \sqrt{\rho_l}$); (b) the coherent scattering domain size; (c) the screw dislocation fraction.

Figure 8. Bright field TEM image showing the dislocation cells formed during tensile deformation.

Figure 9. *Pseudo* lattice strains and the equivalent strains in the loading direction as a function of true strain for the three samples, taking the d-spacing of each {hkl} reflection at ~4% strain to be the reference point.

Figure 10. Illustration of deformation stages in the three samples. Dotted vertical lines mark the transition points between stages. Dotted inclined lines mark the slope of the dislocation evolution curves in the first stage of each sample.

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