High Energy X-ray Diffraction Study of the Relationship Between the Macroscopic Mechanical Properties and Microstructure of Irradiated HT-9 Steel

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Abstract

Samples harvested from an HT-9 fuel test assembly (ACO-3) irradiated for six years in the Fast Flux Test Facility (FFTF) reaching 2-147 dpa at 382-504°C were deformed in-situ while collecting high-energy X-ray diffraction data to monitor microstructure evolution. With the initiation of plastic deformation, all samples exhibited a clear load transfer from the ferrite matrix to carbide particulate. This behavior was confirmed by modeling of the control material. The evolution of dislocation density in the material as a result of deformation was characterized through full pattern line profile analysis. The dislocation densities increased substantially after deformation, the level of dislocation evolution observed was highly dependent upon the irradiation temperature of the sample. Differences in both the yield and hardening behavior between samples irradiated at higher and lower temperatures suggest the existence of a transition in tensile behavior at an irradiation temperature near 420°C dividing regions of distinct damage effects.

Keywords: Ferritic martensitic steel, HT-9, In-situ tensile test, High-energy X-ray diffraction, Lattice strain, Irradiation embrittlement

1. Introduction

Sodium-cooled fast reactors have a target fuel burnup >40% for the US DOE-NE Fuel Cycle R&D program. One of the limiting factors in achieving this high burnup is the ability of the clad materials to withstand high irradiation doses. Ferritic/martensitic (F/M) steels have been presented as one of the leading candidates for structural materials in fast, fusion, and Gen IV reactors for their superior resistance to radiation-induced swelling and creep and reasonable high temperature strength as compared with austenitic steels [1–4]. The initial HT-9 alloy design depends on a very fine carbide structure to provide good yield strength without a compromise in fracture toughness. Thus, it is important to understand how the ferrite matrix and carbide particulates interact during deformation, in particular as a function of irradiation conditions.

In addition to the carbide population, other irradiation-induced phase changes have been reported, particularly the development of α' precipitates and the G-phase, both of which have a distinct temperature and irradiation dependencies [5–8]. An understanding of the phase changes and the stability of the dislocation and carbide structures as a function of irradiation dose and temperature is important for the evaluation of this material for extended use in high irradiation environments at planned exposure temperatures between about 400°C and 500°C. In particular, the relative ability of the ferritic-martensitic matrix versus the indigenous carbide structure to resist deformation is an important part of this assessment since these are the controllable microstructural features in the material design and thermo-mechanical processing.

The deformation of as-processed HT-9 has previously been studied using in-situ neutron diffraction coupled with self-consistent polycrystalline plasticity models. In particular, a two-phase elastic-plastic self consistent (EPSC) model was used to simulate the response of the ferrite matrix of HT-9 to compare to in-situ neutron diffraction measurements during deformation [9]. However, the neutron diffraction lacked the ability to directly monitor the response of the small volume fraction of carbides during deformation and behavior of the minor carbide phase had to be inferred. Here we present a furthering of that research using high energy synchrotron X-ray diffraction (HEXRD), which can track the carbide as well as ferrite behavior, to probe both as-processed and irradiated HT-9 during tensile deformation.

2. Experiment

2.1 Material

The material in this study was recovered from a hexagonal HT-9 duct (labeled ACO-3) which experienced 6 years of irradiation exposure in the Fast Flux Test Facility (FFTF) during the Core Demonstration Experiment, a partial core loading of the FFTF to demonstrate fuel system capabilities. The hexagonal duct measured 367 cm in length, 60 mm across each hexagonal side, and 3 mm in wall thickness [10]. The nominal composition of the HT-9 steel removed in the ACO-3 duct is Fe–11.8Cr–0.2C–0.2Si–0.5Mn–0.5W–0.3V–1.0Mo–0.5Ni in weight percent [11]. Prior to irradiation, the duct experienced a heat treatment of a 30 minute hold at 1065°C followed by an air cool, then a 60 minute hold at 750°C followed by a final air cool. The ACO-3 duct was irradiated at elevated temperature (367°C–507°C).

2.2 Sample Preparation

Samples were harvested from different positions along the duct after its irradiation. Depending on their position along the duct, these samples received doses between 2 dpa and 147 dpa at temperatures between 382°C and 504°C. Note that, because of the duct position in the FFTF core, the irradiation doses were lowest at the bottom and top of the duct, while the exposure temperature increased along the length of the duct from the bottom to the top [11]. Thus the combination of irradiation dose and irradiation temperature was unique for each position along the duct. Details concerning the irradiation history of the duct and sampling technique can be found in [10–12]. Table 1 shows the irradiation conditions (both dose and average irradiation temperature) of the five test samples, historically labeled as 2E1, 5E1, 6E1, 6E5, and 6E9. Control specimens were removed from a sister duct which did not undergo irradiation. It should be noted that this control material is distinct (due to different thermo-mechanical processing) from the material used in the previous neutron diffraction tests of this alloy [9].

Tensile samples in the “S1” geometry, shown in Figure 1a, were electro-discharge machined (EDM’ed) from the control and irradiated duct material with a nominal gage length of 5 mm, width of 1.2 mm, and thickness of 0.75 mm.

2.3 X-ray Diffraction Experiments

High-energy X-ray diffraction measurements were performed at the 1-ID beamline of the Advanced Photon Source (APS) at Argonne National Laboratory. In-situ uniaxial tensile tests of each material were completed on an MTS closed-loop servo-

hydraulic test frame. The tensile samples were held in the load frame with pins inserted through holes at each tab on the end of the specimen. The load was transferred to the specimen via these pins, resulting in elongation of the holes with deformation. Due to activation, the samples were covered with Kapton tape and contained in a polymer bellows attached to the grips during loading, shown schematically in Figure 1b. The force required to stretch the bellows was measured prior to the experiment and found to be negligible compared to the force required to deform the samples (2 orders of magnitude less).

The engineering stress, σ_e , is calculated directly from the load cell by

$$\sigma_e = F/A \quad (1)$$

where F is the force measured directly from the MTS load frame and A is the cross-sectional area of the gage portion of the sample. Unfortunately, due to the activity of the samples (up to $\sim 100\text{mR/hr}$ at contact), the cross sectional area, in particular, the sample thickness, was not determined accurately. We estimate an error of approximately $\pm 10\%$, based on error inherent in the EDM slicing process. Because the samples were cut relatively thin, a small absolute error in the final EDM'ed thickness results in a relatively large error in the cross sectional area. Also, the containment requirement prevented direct measurement of strain on the sample. The change in length of the sample was determined by a linear variable differential transducer (LVDT), but the pin holes in the sample distorted significantly during the measurement. Therefore, neither the macroscopic stress nor strain was determined as accurately as desired in the gage section of the sample. The tensile samples were held and deformed vertically at room temperature with a constant crosshead speed corresponding to a strain rate of $\sim 1 \times 10^{-4}/\text{sec}$ under displacement control to displacements of approximately 0.5 mm, resulting in plastic strains ranging between 3% and 8% depending on the sample strength. Diffraction measurements were taken during deformation with a monochromatic 86 keV ($\lambda = 0.0144\text{ nm}$) X-ray beam impinging on the gage area of the sample. The incident beam size was $100 \times 100\text{ }\mu\text{m}^2$. The Debye diffraction rings from the matrix and the carbide precipitates contained in the diffraction volume were recorded with a GE41RT area detector composed of a 2048×2048 array of $200\text{ }\mu\text{m}$ pixels.

In order to examine internal lattice strains, diffraction measurements were conducted continuously during deformation with the detector set a distance of 1.15 m from the sample and centered on the through-beam, sampling a scattering angle 2θ of $\pm 9.7^\circ$ in both vertical and horizontal directions. This configuration allowed for recording of complete Debye rings for peaks with d-spacing greater than 0.082 nm. The Debye rings were integrated in 15° segments centered at detector azimuthal angles, η , about the beam center to generate 24 diffraction line profiles (1-D patterns) using Fit2D [13,14]. Because of the high X-ray beam energy and the resulting low diffraction angle, the diffraction vectors, Q , for each of these profiles are nearly normal to the incident beam. Therefore, to good approximation, the diffraction vectors, $Q_{\parallel}$, corresponding to the profiles representing 15° integration at $\eta = 90^\circ$ and $\eta = 270^\circ$ are parallel to the loading (axial) direction, and those diffraction vectors, $Q_{\perp}$, for $\eta = 0^\circ$ and $\eta = 180^\circ$ are transverse to the loading direction.

For diffraction line profile analysis (DLPA), diffraction patterns were captured before and after deformation of each sample with the detector set at a distance of approximately 1.83 m and offset from the center of the through-beam by 184 mm. This off-center configuration sacrificed full Debye rings for all peaks, instead giving the maximum possible scattering angle (maximum $2\theta = 13^\circ$) and optimizing instrument resolution for the best possible fitting of the peak line profiles. The instrumental broadening and peak shape were determined by fitting ceria peaks from a standard ceria reference sample. Recalibration of the detector was repeated after every change in the detector position.

2.4 X-ray Diffraction Analysis

2.4.1 Phase Strain and Stress

Rietveld refinement and single peak fits were completed for each diffraction pattern and lattice parameters for the ferrite matrix and carbides found with the General Structure Analysis Software (GSAS) [15]. A pseudo-Voigt function with fixed Lorentzian and fitted Gaussian components was used to fit the peak shapes and define their positions. Automated routines (APSRunrep and APSspf [16]), which call GSAS sub-routines, were developed to enable the analysis of the thousands of recorded diffraction patterns. Quoted uncertainties are the estimated standard deviations (esd's) returned by GSAS.

The X-rays counted in a particular diffraction peak (hkl) are diffracted from a subset of grains (family) defined by a specific plane normal (hkl) parallel to the diffraction vector. The evolution of that particular peak, e.g. change in position (d-space) or peak width, represents the evolution of that particular family of grains. In contrast, the lattice parameter determined from

Rietveld refinement of the entire pattern, **a**, represents an empirical average over all grain orientations of the phase and represents the phase response.

The elastic strains are calculated from the change in lattice spacing by

$$\varepsilon = (d - d_0)/d_0 \quad (2)$$

where d refers to the interplanar spacing of a particular set of lattice planes (d_{hkl}), or the lattice parameter, **a**. Strains calculated from single peak fits will be referred to as hkl-specific, or orientation dependent, strains. Those calculated from the lattice parameter, **a**, will be referred to as phase strains. Note that d_0 is taken to be the linear extrapolation of the lattice spacing in the elastic region to zero stress. This ignores the fact that there will be pre-existing thermal stresses in each phase due to the mismatch in coefficient of thermal expansion.

In the following, the phase strain in the matrix is calculated from the Rietveld fit of the ferrite phase and the resulting lattice parameter **a**. In the control specimen, which was not activated and thus not in containment, several single carbide peaks were resolvable and Rietveld refinement of the carbide lattice parameter was completed as it was for the matrix. In contrast, the parasitic scattering caused by the polymer bellows necessary for the activated samples obscured several of the carbide peaks from the irradiated material making Rietveld refinement impossible. The strain in the carbides was thus calculated from the single peak fit of the $M_{23}C_6$ carbide (422) peak (where we have assumed M is Cr) and the obtained interatomic spacing. This was the only carbide peak with adequate counting statistics for the irradiated samples and is assumed to be representative of the behavior of the carbide.

We assume the lattice strains represent the response of the phase and thus calculate the phase stresses in the axial direction using the generalized Hooke's law:

$$\sigma_{11} = \frac{E}{1 + \nu} \varepsilon_{11} + \frac{\nu E}{(1 + \nu)(1 - 2\nu)} (\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}) \quad (3)$$

where the subscript '11' refers to the axial direction and the subscripts '22' and '33' refer to each of two transverse directions. We assume symmetry in the transverse directions ($\varepsilon_{22} = \varepsilon_{33}$) and calculate the transverse stresses from the proper permutations of indices. Poisson's ratio for both phases is taken to be $\nu = 0.3$ and moduli of $E_{\text{ferrite}} = 208$ GPa, $E_{\text{carbide}} = 330$ GPa [9,17].

Note, in general, it is not advisable to calculate a stress from hkl-specific strains because, with the exception of very specific textures, the grain families sampled by strain measurements in orthogonal directions (axial and transverse) are distinct. Thus, a stress calculated using hkl-specific strains in multiple sample directions does not represent the stress perceived by any actual set of grains. In order to defensibly calculate a stress from hkl-specific strains, one implicitly assumes that the strains in the distinct grain sets sampled in the diffraction patterns with orthogonal diffraction patterns are equivalent, i.e. that the specific grain families are representative of the phase. Residual stress measurements using diffraction, in particular at monochromatic sources where it is difficult to record multiple diffraction peaks, often make this assumption out of necessity [18,19], but the validity is seldom discussed.

In the following, strong orientation dependence of the hkl-specific strains in the steel phase will be presented. As such, it is not justified to calculate stress from the hkl-specific strains but, rather, from the phase strain. However, less orientation dependence of the hkl-specific strains is observed in the carbide phase, in particular, of the control sample. Thus, as with many residual stress measurements, born of necessity, we will assume the strain determined from the (422) oriented grains is representative of the carbide phase and use it to calculate phase stresses. The validity of this assumption will be explicitly examined in the following.

2.4.2 Line Profile Analysis

The microstructures of the HT-9 steel samples were quantitatively characterized using whole pattern diffraction line profile analysis (DLPA). DLPA methods evaluate the measured data by forward modeling the diffraction pattern based on physical models of the microstructure and matching the theoretical pattern with the measured data [20–25]. The models describe the effect of the following microstructural features: (i) the size distribution of the coherently scattering domains, (ii) the density, arrangement and character of dislocation structures and (iii) frequency and type of planar faults [23–27] on diffraction peak breadth and shape. The calculated profile functions corresponding to the various microstructural features, together with the

experimentally determined instrumental peak broadening effects, are convolved to obtain the full theoretical diffraction pattern, which is fitted to the measured pattern in order to obtain the details of the microstructure [28].

The Extended Convolutional Multiple Whole Profile (eCMWP) line profile analysis software package was used to determine dislocation densities in the ferrite matrix both before and after deformation [25,28]. The analysis was completed by modeling six independent diffraction peaks from the ferrite: (110, 200, 211, 220, 310, 321). The microstructure was refined for average sub-grain size and dislocation density. Background scattering from the containment and diffuse scattering from the radiation damaged samples obfuscate the eCMWP fits, in particular the fitting of the tails of the peaks. In these cases, it was necessary to fix the variance of the lognormal crystallite size distribution and the Wilkens dislocation arrangement parameter, M . This arrangement parameter characterizes the configurational energy of the dislocation structure and it is closely linked to the dipole character of the structure. Since the effect of this arrangement parameter mostly manifests in the shape of the tails of the diffraction peaks, the results for M were unreliable due to the additional background from the polymer containment. Thus, the values of M were fixed to those values obtained by eCMWP fits of high resolution neutron diffraction patterns of the same set of as-irradiated materials before loading [12]. The neutron diffraction patterns, taken with higher resolution and without the polymer bellows containment, had better quality peaks through the tail region yielding more reliable results for dislocation arrangement parameter. We assume for simplicity that the arrangement parameter is unchanged following the plastic deformation of the samples. This is not an ideal assumption, but the plastic strains are relatively small and, moreover, we have no basis to claim any particular evolution in M and therefore any prescribed variation in the parameter would be similarly unfounded.

In order to examine the dislocation evolution during deformation, we performed Rietveld refinement of the ferrite peaks using GSAS and extracted the microstrain from the variance of the Gaussian component of the line shape. The Thomson-Cox-Hastings pseudo-Voigt function has a variance that depends on 2θ as follows:

$$\sigma^2 = U(\tan \theta)^2 + V \tan \theta + W + P/(\cos \theta)^2 \quad (4)$$

where the parameters U , V , and W are Cagliotti coefficients [29]. To get a qualitative measure of microstrain evolution during deformation, Rietveld fits of the ferrite peaks using this pseudo-Voigt function were completed by refining only one Gaussian fitting parameter, U . V was set to zero and W was fixed. Examining the variance equation above and considering that strain broadening goes as

$$\Delta 2\theta = \Delta d / (d \tan \theta) \quad (5)$$

we can observe the strain broadening contribution to the variance in the parameter U . Consider that the variance is

$$\sigma^2 = \langle \varepsilon^2 \rangle - \langle \varepsilon \rangle^2 \quad (6)$$

If we assume an appropriate selection of d_0 , $\langle \varepsilon \rangle$ becomes zero and the variance is equal to the root mean square (rms) strain,

$\varepsilon_{rms} = \langle \varepsilon^2 \rangle^{1/2}$. If we further assume that the development of microstrain in the ferrite is coming primarily from an increase in dislocation density, then we can monitor the evolution of dislocation density in the ferrite during deformation via the evolution of the ferrite rms strain values. This method is used merely as a qualitative guide to dislocation evolution, recognizing it as complimentary to the more quantitative analysis of the dislocation density before and after deformation done with eCMWP. Thus, we use quantitative line profile analysis to determine the dislocation density before and after deformation, and qualitative analysis to monitor the evolution during deformation.

3. Modeling

The Elastic-Plastic Self-Consistent (EPSC) polycrystalline deformation model [30] has proven to be an excellent companion for diffraction measurements of internal strains [31]. The combination of EPSC modeling and diffraction measurements provides additional information about the tested material, such as yield strength and hardening behavior of the individual phases within a composite [32]. Detailed reviews of the EPSC model can be found in the literature [30,33], and only a brief description will be presented here. The EPSC model is based upon the single crystal behavior of the material. There are no direct grain-to-grain interactions defined within the model, however, each grain interacts with a homogeneous equivalent medium (HEM) with properties determined as the appropriate texture weighted average of all the grains. The interaction between grain and HEM is found using the Eshelby inclusion formalism [34].

A two phase EPSC model [35] assuming spherical carbide particulates was used to calculate the response of the ACO-3 control HT-9 material to enforced deformation. A grain set of approximately 23,000 grains representing the measured texture was used in the calculations. Single crystal elastic constants are not known for either the Fe alloy matrix or carbide particulate of HT-9. The two major alloying components of HT-9 are 12% Cr and 1% Mo, by weight. Elastic constants for various $\text{Fe}_{1-x}\text{Cr}_x$ alloys have been calculated by Zhang et al. [36]. Following their strategy, approximate single crystal elastic constants have been determined for a Fe90Cr10 alloy, see Table 2. While the exact composition of the carbide inclusions in the HT-9 material is not known, we make the assumption that it is mainly Cr_{23}C_6 as Cr is the major alloying component. The single crystal elastic constants for the Cr_{23}C_6 have also not been determined experimentally, but a few directional stiffnesses were calculated by Young et al. [17]. Their values for E_{100} , E_{110} and E_{111} are all approximately (330 ± 5) GPa, and thus we have assumed single crystal elastic constants based upon the assumption that the carbide is isotropic and that its Poisson's ratio is 0.3, see Table 2.

The carbide was assumed to remain elastic throughout the simulation. All of the bcc “pencil-glide” slip systems were assumed active with identical parameters for the ferrite matrix. The parameters describing plastic flow of the matrix, the critical resolved shear strength, initial and final hardening rates and how quickly the hardening decays from the former to the latter, were determined by fitting the output of the model to the observed macroscopic flow strength and lattice strains of the ferrite, see Table 2. Of note particularly is the highly anisotropic elastic response of the ferrite compared to the isotropic response of the carbide. Because the model fitting depends critically on the macroscopic stress and strain determinations which, as noted, were not as accurate as required due to the mandatory sample containment, model parameters were not determined for the irradiated specimen.

4. Results

4.1 Macroscopic Response

Figure 2 shows the observed macroscopic stress-strain curves of the control and irradiated HT-9 materials determined in-situ. The stress-strain response of the control material determined by the EPSC simulation is also shown; the agreement is excellent. With the caveat of increased uncertainty in the macroscopic stress and strain determined for the irradiated samples, the in-situ results are consistent with prior ex-situ mechanical tests [12,37]. Material irradiated at temperatures below 420°C shows a significant increase in flow strength compared with the control material, while that irradiated above 420°C shows little increased strength relative to the control material [12]. A decrease in the hardening rate is observed with increased yield strength. Unloading data are not shown for clarity.

4.2 Lattice Strains

Figures 3a and c show several hkl-specific strains, ϵ_{hkl} , and the phase strain, ϵ_a , for the ferrite and carbide phases, respectively, in the control material as a function of the applied stress. For clarity only every 20th data point is indicated by a symbol, but the representative scatter in the experimental data can be gleaned from the noise in the lines, which go through every experimental point. The uncertainties in the strains in the ferrite are roughly $25 \mu\epsilon$, and are smaller than the symbols. The uncertainties in the carbide strains are considerably larger, roughly $60 \mu\epsilon$, comparable to the size of the symbols. Recall that the uncertainties given here consider only the uncertainty in the position of fitted diffraction peaks output by GSAS (one esd). While there are other factors affecting the accuracy and precision of our measurements, the resulting error is not easily quantified; the true uncertainty in the strain measurements may be substantially higher than quoted herein. Figures 3b and d show corresponding results from the EPSC simulations of the deformation of the control material.

The elastic anisotropy observed in the ferrite below the yield point (~ 680 MPa) is expected based on the single crystal stiffness matrix [36] (the crystal is stiffest along the (111) plane normal, and most compliant along the (100)). The phase response closely matches that of (110) and (211) oriented grains. In contrast, the observed elastic response of the carbide particulate is relatively elastically isotropic. The hkl-specific and phase responses are well captured by the EPSC simulation utilizing the directional stiffnesses of Young et al. [17].

Above 680MPa, the experimentally observed ferrite lattice strains (hkl-specific and phase) saturate, indicating that the imposed strain is being accommodated by mechanisms other than elasticity, i.e. by plastic deformation in the matrix. Concomitantly, the carbide lattice strains increase at an elevated rate above the matrix yield point as subsequent increases in the applied load are shed from the ferrite matrix to the carbide particulate. Beyond 830MPa, the ferrite grains again accumulate elastic strain, indicating that they have hardened. At this point, the first hint of strain anisotropy is observed in the carbide. The (422) oriented carbide grains continue to accumulate strain at an enhanced rate while the lattice strains in the

(420) and (440) oriented grains, as well as the phase strain, accumulate at a decreased rate above 830MPa. The relative isotropy of the grain response of the carbide phase throughout most of the deformation lends credence to the use of a single peak strain in the calculation of the phase stress, described above.

The EPSC model continues to accurately reproduce the lattice strains in the ferrite phase throughout the plastic region, including the initial yield of the matrix at 680MPa and subsequent hardening observed at 830MPa. The plastic anisotropy is due to a combination of both the orientation dependent distribution of stress due to the elastic anisotropy and the orientation dependent resolved shear stress based on the assumed active slip system.

The initial response of the carbide phase to yielding in the matrix at 680MPa is also well captured by the model, as is the up-turn in the (420), (440), and phase strains evident beyond 830MPa. However, the anisotropy that is apparent in the carbide strains above 830MPa is not captured. As the elastic stiffness tensor assumed for the carbide phase was forcibly isotropic and the material was assumed to deform only elastically, the model has no way to reproduce the observed distinct behavior manifested by the (422) oriented grains. Indeed, all of the modeled carbide hkl-specific strain and phase strains are identical throughout the simulation.

Figure 4a shows a plot of applied stress versus the experimentally observed phase strains (symbols) in the loading (solid symbols) and transverse (open symbols) directions for the matrix and carbide phases of the control material. The model results are represented by the solid lines as indicated. As plotted in Fig. 4a, the difference in the loading direction phase strains between the ferrite and carbide is immediately evident. The ratio of the elastic moduli of the two phases individually is $E_{\text{carbide}}/E_{\text{ferrite}} = 1.47$. However, the mutual geometric constraint of the phases in the composite forces the effective moduli ratio much closer to unity, experimentally observed to be 1.23 ± 0.02 and calculated by the model to be 1.22. The plastic response in Fig. 4a is as described in reference to Figure 3, but less distinct because the scale is enlarged to include the transverse response.

Figure 4b shows similar data (experimental only) collected on the 2E1 material. The uncertainty in the carbide lattice strain increases in the irradiated material to roughly $150\mu\epsilon$. Overall, the response of the individual phases is similar to the control material, with the yield strength reduced from 630MPa to roughly 590MPa. The ratio of the effective elastic moduli in the 2E1 material is 1.18 ± 0.04 , again similar to the control material. Also similar to the control material, the phase strain in the ferrite saturates above the yield point and begins to increase again, in this case, above ~ 750 MPa.

Figure 4c-f shows similar data collected on material with the remaining irradiation conditions (5E1, 6E1, 6E5, and 6E9, respectively). The relative elastic response of the constituent phases is fundamentally different in these materials from that observed in the control and 2E1 material. In the material which received higher doses of radiation at lower temperatures, the carbide and ferrite phase strain curves are collinear in the elastic regime. This is unexpected, as this behavior typically presents in the case of e.g. a fiber composite, with fibers parallel to the straining direction, wherein both phases are geometrically constrained to accumulate the same strain [38]. Since we know the carbides are not constrained by fiber geometry in this material, it is unclear what is causing the coupling of the phase strains.

While cross sample comparison of the phase moduli is ill-advised because of the uncertainty in the macroscopic stress measurement, the ratio of the carbide to ferrite stiffness is insensitive to uncertainty in the macroscopic stress and can be compared both across samples, and to the predictions of the EPSC model. In the 5E1, 6E1, 6E5, and 6E9 materials, the observed ratio $E_{\text{carbide}}/E_{\text{ferrite}}$ is 0.98, 1.06, 1.12, and 1.09. We note that the EPSC model contains no physics which could predict this shift to collinear deformation behavior of the matrix and particulate inclusions other than changing the input elastic stiffness tensors.

The behavior of the ferrite in the 5E1 material following the activation of plasticity, i.e. saturation of the lattice strains at 710MPa followed by subsequent accumulation of strain above 900MPa, is similar to the control and 2E1 material with the expected increase in the yield strength based on the macroscopic flow curve. However, the evolution of the phase strain in the ferrite beyond the initiation of plasticity is distinct in the 6E1, 6E5 and 6E9 materials. In each of these materials, the ferrite strain saturates at yield of the matrix, but does not later increase as observed in the control, 2E1 and 5E1 materials.

It is difficult to discern interesting behavior in the transverse strains, which are relatively small. The one exception is the 6E5 material where the transverse strain is nearly 0 throughout the deformation test. This is unphysical and likely an artifact of the data analysis. We note that the carbide strain in the irradiated material, in particular in the transverse direction, was often not well determined by the Rietveld analysis. In the case of the 5E1 material, the carbide phase in the diffraction pattern

transverse to the straining direction could not be fit accurately at all. In the case of the 6E1, 6E5, and 6E9 materials, the fits to the carbide in the transverse direction became unstable prior to the completion of the deformation test.

4.3 Phase Stresses

Figure 5a shows the evolution of phase stresses in the ferrite during deformation of the control and irradiated materials. Phase stresses were calculated using equation (3). The lack of reliable transverse strains from the carbides in the irradiated samples throughout the entire duration of the deformation tests makes it impossible to calculate the carbide phase stresses in these samples. Thus, for comparison with Fig. 5a, Figure 5b shows the longitudinal phase strain in the carbides in the deforming control and irradiated materials, which is well determined and, in a tensor multiplication sense, is proportional to the phase stress. Reliable transverse strain data were available for the control material, enabling determination of the carbide phase stress in this case. Thus the right axis of Fig. 5b shows approximate stress levels associated with the lattice strains in the carbide of the control material. These provide the reader with a rough idea of the stress carried by the carbides in all of the samples.

The evolution of the stress in the ferrite varies greatly depending on whether the material was irradiated above or below 420°C. Specifically, in the control material, the ferrite matrix yields when the phase stress is roughly 590MPa, corresponding to a macroscopic applied stress of 700MPa (Fig. 2). The ferrite matrix hardens to 740MPa at the end of the test, 0.07 strain. Beyond the yield point, the subsequent increases in the macroscopic stress are being predominantly carried by the small volume fraction of carbide reinforcement phase as the stress on that phase increase from roughly 800MPa at the yield point to over 3000MPa at the completion of the test.

The phase specific responses of the 2E1 and 5E1 materials (irradiated above 420°C) are similar to the control material, with the yield stress on the ferrite slightly lower. It is interesting to note that the macroscopic flow stress of the control material and 5E1 material are very similar. Yet, the ferrite yields at a lower phase stress in the 5E1 material, while the carbide carries a larger portion of the load. This suggests a change in the mechanism of stress transfer between the materials.

In contrast, the yield strength of the ferrite in the material irradiated below 420°C has increased to approach 1GPa, and softens with subsequent plastic strain. The load carried by the carbide is correspondingly higher in these materials. We note that in an absolute sense, the ferrite phase stress observed in the 6E5 material is inconsistent with the observed macroscopic flow strength, i.e. the phase stress is larger than the macroscopic stress. This is due to the observed transverse strain which was noted to be unphysical above.

4.4 Peak Broadening

Figure 6 shows the (1 1 0) ferrite diffraction peak before and after deformation for two irradiation conditions (2E1 and 6E5), with the intensity and peak position normalized to highlight the broadening; the broadening post-deformation is readily apparent. The full width at half maximum (FWHM) of the peaks evolve from 0.050nm⁻¹ to 0.063nm⁻¹ and from 0.056nm⁻¹ to 0.069nm⁻¹ in the 2E1 and 6E5 material, respectively. The lower temperature irradiated material starts with broader peaks, consistent with [12], and they remain as such following the deformation.

To give a qualitative view of the evolution of the dislocation density during deformation, Figure 7a shows the development of the root mean square (rms) strain, $\epsilon_{rms} = \langle \epsilon^2 \rangle^{1/2}$, in each ferrite phase with increasing plastic strain. The control material as well as material irradiated at temperatures above 420°C exhibit initially low ϵ_{rms} , while the material irradiated at temperatures below 420°C have higher ϵ_{rms} . ϵ_{rms} increases with plastic strain in all of the material, approaching 0.8% where it seems to saturate. It is important to recognize what this number represents: the width of the distribution of the lattice strain around the average strain, $\langle \epsilon \rangle$. A variance of 0.8% represents a large distribution of elastic strain and is not consistent with variation in stress distribution across grain sets as the distribution itself would have to be much larger than the yield stress. This indicates that the major source of microstrain in the ferrite phase are the heterogeneous strain fields associated with individual dislocations, consistent with our assumption, so we may view the evolution of the ferrite rms strain as representative of the qualitative dislocation density evolution. If changes in the Wilkens dislocation arrangement parameter (M) are neglected, ϵ_{rms} scales roughly with the square root of the dislocation density.

Figure 7b shows the ϵ_{rms} observed in the carbide. The scale is held fixed relative to Fig. 7a to emphasis the difference between the ferrite and carbide. Nearly an order of magnitude less microstrain develops in the carbide phase of each material

with plastic strain, saturating at roughly 0.1%. At this level of microstrain, it is reasonable to associate it with heterogeneous stress distribution based on variations in the local neighborhood of the individual carbide grains.

The increase in peak breadth of the ferrite peaks following deformation is unequivocal. As is often the case, the interpretation of same is more problematic. Line profile analysis of high resolution neutron and X-ray diffraction data completed specifically to determine the dislocation density and coherent crystal size was reported in a previous publication [12]. The current samples were analyzed analogously in the as-received state, but the data were less robust due to parasitic scattering from the polymer bellows containment. Figure 8 shows the dislocation density as a function of irradiation temperature (pre-deformation), determined using eCMWP for line profile analysis, in the current work and the previous work. The agreement is very good, lending credibility to the results of the current work, even with data obscured by scatter from the containment. Figure 8 also shows the dislocation density in each material at the completion of the deformation test. In each case the dislocation density has increased considerably following deformation.

5. Discussion

The parameters obtained from the fits to the diffraction data can be related directly to the evolution of the microstructure, phase stress, dislocation density, etc., during deformation of the different materials. From this, we try to understand the evolution of the macroscopic mechanical properties following irradiation, in particular, the repeated theme of strong changes in mechanical properties for samples irradiated above and below 420°C.

5.1 Elastic Deformation

The elastic response of the control and 2E1 materials are well predicted by the EPSC model using fixed elastic parameters derived from the literature [17,36]. This is to be expected, as the 504°C/2 dpa and control samples have similar microstructures and have been shown not to contain the same irradiation-induced phase structures, namely α' and G phase, present in the other samples [5].

In contrast, the remaining irradiated materials exhibit the same phase specific modulus of the ferrite and carbide in the elastic region; this is unexpected in a particulate composite and is not reproduced by the two phase EPSC model. It is unlikely that the irradiation-induced dislocation density alters the elastic interaction between the ferrite and carbide. Also, the 5E1 material, which exhibits the elastic collinear deformation, has the same dislocation density as the 2E1 material, which has distinct moduli for the ferrite and carbide phases. The material irradiated to higher dose and a lower temperature has been shown to contain irradiation-induced minor phases, including the α' and G phases, as well as voids [5,12]. The precipitation of these phases could affect the local (if not global) chemistry of the matrix and/or locally strengthen the matrix. We can only speculate that the effects of these microstructural features alter the elastic interaction between the ferrite and carbide phases.

5.2 Plastic Deformation

In the previous work [12], it was shown that the initial yield strength of the irradiated material was roughly proportional to the square root of the dislocation density, consistent with the Taylor law [39] $\sigma = \sigma_0 + M\alpha Gb\sqrt{\rho}$, where σ is stress, the Taylor factor, M_T , is 3 [39,40], G is the shear stress (86.95 GPa for HT-9 steel), b is the length of the Burgers vector, .2466nm for the $\langle 111 \rangle \{110\}$ slip system, and ρ is the dislocation density. α is a factor describing the strength of the obstacles that a moving dislocation has to overcome during plastic deformation; for metals, the value of α is usually roughly 0.3 [41].

It is perhaps more appropriate to remake the Taylor plot using the yield stress of the ferrite determined herein, rather than the macroscopic yield strength. This is shown in Figure 9. Note, we have omitted the 6E5 data as the yield strength is skewed by the unphysical transverse strains (Fig. 4e). The plot of the square root of the dislocation density with the phase yield strength is roughly linear.

The evolution of the dislocation density during deformation is not consistent with the Taylor Law. In particular, the dislocation density of materials irradiated below 420°C, i.e. 6E1, 6E5, and 6E9, all increase, while the flow strength of the ferrite peaks at very low strains and thereafter remains constant or decreases. Transmission electron microscopy examination of the as-irradiated and deformed microstructures has been performed on the condition at 20 dpa at the irradiation temperature of 380°C. This condition was selected for further examination since it shows the largest increase in yield strength of any of the irradiated conditions and the as-irradiated microstructure has been examined previously [5,6]. The as-deformed microstructure shows high dislocation densities and precipitate densities. This is consistent with the earlier TEM results on

this material condition in the undeformed state [5,6]. Relatively large void structures in the deformed material are seen on the lath boundaries in Figure 10a. The high precipitate and dislocation densities and their interactions are evident in Figure 10b. More quantitative information regarding the precipitate structures is available from the previous studies [5,6]. These earlier studies show the presence of a high number density of alpha prime precipitates at 382°C with sizes between 4 and 11 nm in the as-irradiated material. The number density reduces with increasing irradiation temperature and the precipitates were not found at the highest two irradiation temperatures. Despite the high number densities at moderate temperatures, Cr content in this material is on the edge of the compositions where alpha prime is found. The highest experimentally derived alpha prime content in any of these irradiation conditions is less than 4% and may not have a major impact on the observed mechanical properties. Nevertheless, the highest observed concentrations of alpha prime are found in the materials condition with the highest yield strength: 382°C at 20 dpa [5,6].

In the current study, there is no evidence of dislocation channeling or flow localization processes which have been observed in other post-irradiation deformation studies of similar materials [42,43]. The tensile results here, Figure 2, indicate sustained plastic hardening following yield in all of the tests. This behavior indicates some level of uniform plastic flow as opposed to sharp load drops following yield and more localized flow typical of flow localization behavior. In the two lower temperature samples, 382°C and 399°C, there is identifiable work hardening with a large increase in dislocation density. This behavior has been seen before in steels [44] and is an indication of uniform plastic flow over the range of plastic deformation investigated in these studies. This supports a process where mobile dislocation structures in the material can annihilate the irradiation-induced dislocation loops initially present in the as-irradiated material as they move during plastic deformation, reducing the work hardening of the material. The TEM results, Figure 10, show a widely dispersed dislocation structure within the laths as well as the higher density dislocation structures in the lath boundaries. The radiation-induced precipitation structure, particularly at lower irradiation temperature [5,6], accounts for the elevated yield strength. The relatively uniform distribution of dislocation structures within lath boundaries accounts for the observed strain hardening behavior. This more uniform plastic deformation behavior and the ability to generate copious dislocations during plastic deformation, even at the lower irradiation temperatures, is an important aspect of the post-irradiation mechanical performance of this alloy.

6. Conclusions

In-situ X-ray diffraction measurements during deformation of control and irradiated HT-9 steel samples were completed toward understanding the documented changes in macroscopic mechanical properties in terms of the grain-scale response. To this end, a two phase elastic-plastic self-consistent model [9,30] was developed to aid in interpretation of the observed materials response and the associated diffraction patterns. Specifically, the model was optimized to reproduce the observed macroscopic flow strength and microscopic lattice strains observed in both phases of the control material. The model well reproduced both the elastic anisotropy observed in the ferrite matrix and the distribution of load between the matrix and carbide particulate in the elastic region. Further, the evolution of the lattice strains in both phases in the elastic plastic transition and early plasticity was also well captured by the model. Only anisotropic plastic behavior of the carbide near the completion of the test was not reproduced because the model assumed isotropic elastic properties for the carbide and did not allow for plastic deformation of the carbides.

The distribution of stress between the phases during elastic loading was observed to be distinct in the material irradiated below 420°C, i.e. material with a high irradiation-induced dislocation density [12] and irradiation-induced minor phases [5,6]. Collinear elastic deformation behavior, similar to that expected for fiber composites [38], was observed in this material. The EPSC model has no physical mechanism that can reproduce this behavior. The change in the elastic behavior occurs concurrently with the formation of irradiation-induced minor phases and we suppose, without solid evidence, that the effects are related. Moreover, the hardening of the ferrite matrix was distinct between the material irradiated below 420°C and that irradiated above. In the control material and high temperature irradiated material, the ferrite hardened significantly with deformation, while the ferrite in the material irradiated at low temperature showed more limited hardening, but with solid evidence of uniform distribution of dislocation multiplication and flow.

Qualitative indications of increases in dislocation density were observed during the deformation in the form of increased peak breadth and increased root mean square strain in the material in each irradiation condition. Quantitative measurements of the dislocation density before and after deformation indicated a clear and substantial increase during deformation. The initial dislocation density in the ferrite scales linearly with the phase yield strength as expected based on the Taylor law [39].

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Figure 1. (a) Geometry of a tensile sample and (b) deformation geometry within containment.

Figure 2. Macroscopic flow curves of control and irradiated materials determined in-situ during X-ray diffraction measurements. In the case of the control material, the solid black line represents EPSC model calculations of the flow strength.

Figure 3. Control material: (a) Measured and (b) calculated stress vs. lattice strain curves for the ferrite matrix, and similarly, measured (c) and calculated (d) curves for the carbide particulate. Here the (hkl) labels indicate the strain in the specified family of grains and the label “a” indicates the whole phase strain as measured by the change in lattice parameter “a” (via

Rietveld fit). Only the phase strain, ϵ_a , is shown (black dots) during unloading to avoid clutter in the plot. Both phases unload linearly; the ferrite to a state of residual compression, the carbide to residual tension. Note that there is considerable residual tensile strain in the carbide after unloading.

Figure 4. Stress vs. phase strains for the matrix (black circles) and carbide (red squares) in the control and irradiated materials. The closed symbols represent the loading direction and the open symbols (showing negative strain) the transverse direction, perpendicular to loading. In the plot of the control materials, (a), the solid lines represent the corresponding EPSC model calculations.

Figure 5. Evolution of (a) ferrite phase stresses and (b) carbide phase strains with tensile deformation. The right axis on (b) represents the approximate carbide phase stress in the control material to provide a rough stress scale for the carbide phases in the irradiated materials as described in the text.

Figure 6. (110) diffraction peak observed from material in the 2dpa/504°C and 42dpa/399°C condition before (black open circles) and after (red closed circles) deformation.

Figure 7. Evolution of the rms strain in the (a) ferrite and (b) carbide phases with deformation. In the ferrite, the rms strain evolution represents the dislocation density evolution during deformation.

Figure 8. Observed dislocation density, calculated by eCMWP line profile analysis, before (in this work and from [12]) and after deformation.

Figure 9. Dependence of the ferrite yield strength on the square root of the observed dislocation density.

Figure 10. Bright Field TEM images of HT9 irradiated to 20 dpa at 380°C, after deformation to ~3.5% strain: (a) most of the voids are seen along lath boundaries and (b) there is a high density of dislocations and precipitates with the potential for interaction and pinning.

Table 1
Irradiation conditions of HT-9 samples harvested from the ACO-3 duct.

Sample	Irradiation temp (°C)	Dose (dpa)
2E1	504	2
5E1	441	147
6E1	416	110
6E5	399	42
6E9	382	22

Table 2
Single crystal elastic constants and hardening parameters for the matrix and carbide phases. All values are in GPa.

	C_{11}	C_{12}	C_{44}	τ_0	τ_1	θ_0	θ_1
Fe90Cr10	237.0	127.9	120.7	0.300	0.052	0.200	0.200
Cr_{23}C_6	444.2	190.4	126.9	--	--	--	--