Technical Report

Understanding the Phase Transformation of Thermal Aged and Neutron Irradiated Duplex Stainless Steels Used in LWRs

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To understand the thermal-aging and neutron-irradiation effects on duplex stainless steels used in LWRs, *in-situ* testing with wide-angle X-ray scattering (WAXS) was used to quantify the mechanical response of individual phases under a tensile load. The X-ray experiments were complemented by microstructural characterizations using transmission electron microscopy (TEM) and atom probe tomography (APT). The aging effects were quantified in terms of (1) macroscopic tensile property, (2) lattice-strain evolutions of both the ferrite and austenite phases, and (3) dislocation-density evolution during plastic deformation. It was determined that thermal aging had a minimal effect on the austenite phase; however, thermal aging dramatically increased lattice strain and its yield strength in the ferrite phase. The increase of strength saturated at or before 1000 hours of thermal aging at 475°C. Pores that developed at the phase boundary were only observed for the 3000 hours aged specimen, with a significantly decreased ductility. The results also indicate that there is a load partition shift between ferrite and austenite phases upon thermal aging, as the ferrite phase becomes hardened. TEM and APT characterizations show that neutron irradiation introduced a similar microstructural change in the ferrite phase, including spinodal decomposition and G-phase precipitates.

Introduction

Extending the service lifetime of light-water reactors (LWRs) beyond 60 years requires a good understanding of the degradation mechanisms of materials and components in reactors. The lifetime of reactor components made of duplex stainless steels can be limited by embrittlement from thermal aging, neutron irradiation, or a synergistic effect of both. Previous studies showed that the spinodal decomposition in the delta ferrite phase is a primary embrittlement mechanism of the duplex structure stainless steels, while G-phase precipitates were also identified [1–2]. Most past studies focused on characterizations of fine-scale precipitates and phase decomposition using TEM and APT [3–5]. The fundamental mechanism and kinetics of elemental segregations occurring in the ferrite has not been fully understood. The exact

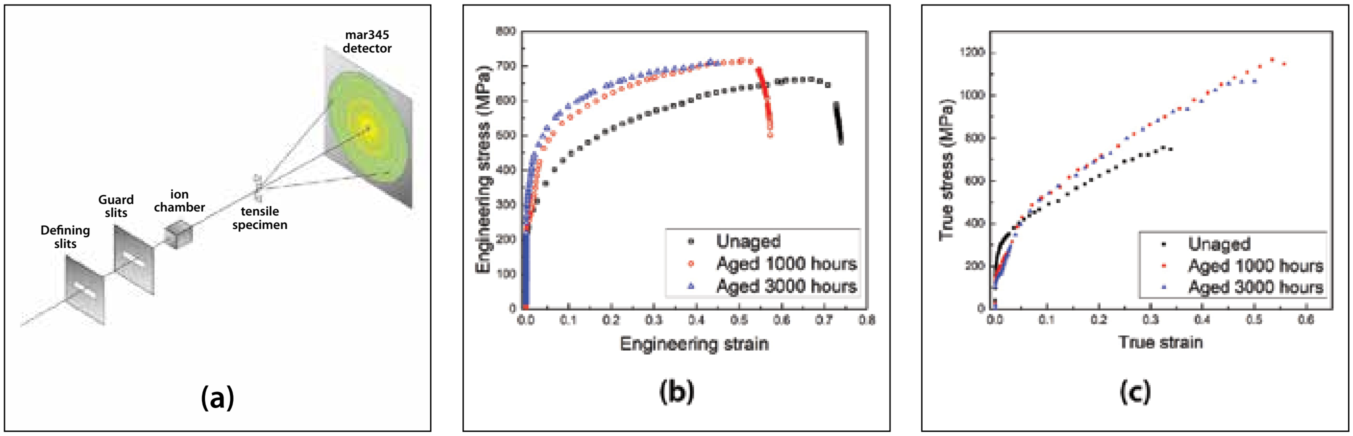

Figure 1. (a) In-situ tensile testing with WAXS, (b) engineering stress vs. stain curves, and (c) true stress vs. true strain curves.

concurrent evolution mechanism of the solute clustering and spinodal decomposition are not clear. This knowledge gap has hindered the development of thermodynamic and kinetic modeling of microstructural evolution. It has also been speculated that cracks initiate in hardened ferrites and then propagate along the phase boundaries between ferrite and austenite. However, the fundamental mechanism of how the microstructural changes decrease the materials' fracture toughness has yet to be determined. This determination is needed to construct a physical model to predict the mechanical response to justify reactor lifetime extension.

In this research project, we use high-energy X-ray techniques, including X-ray diffraction (XRD), extended X-ray absorption fine-structure spectroscopy (EXAFS) and *in-situ* tensile testing with wide-angle X-ray scattering (WAXS) to probe the elemental segregations, phase precipitations and lattice-strain status under tensile load of different phases in selected cast stainless steels. The studies are complemented by advanced microstructural characterizations and conventional tensile testing. For the *in-situ* tensile experiments, only the pristine and thermally aged materials (sub-sized tensile specimens) were used due to the APS beamline dose limit, which excluded irradiated materials. For the microstructural studies, the samples included samples prepared from both aged and pristine *in-situ* tensile fractured specimens (TEM), and irradiated specimens with and without prior thermal aging (APT).

The research project is highly relevant to the DOE-NE Light Water Reactors Sustainability Program, and its outcome will significantly improve the scientific understanding of the degradation of duplex-structure stainless steels in LWRs and contribute to the construction of a physics-based model for predicting material performance for reactor license renewal and regulation.

> "NSUF offers a great platform for conducting scientific research and training the next generation of work force."
>
> - Yong Yang

Experimental or Technical Approach

As shown in Figure 1a, the *in-situ* WAXS tensile tests were conducted using the 10-ID-B beamline at the Advanced Photon Source (APS), Argonne National Laboratory (ANL). Two beamline experiments were conducted using two different control modes. For the first experiment, a monochromatic X-ray beam with an energy of about 64 keV ($\lambda = 0.1925$ Å) and a beam size about 600 µm × 600 µm was employed. The specimen was strained incrementally until fracture while operating in a displacement control mode. A displacement step of 0.001 mm in the elastic region and 0.05–0.10 mm in the plastic region was employed. X-ray-diffraction patterns were acquired after each displacement step using the mar345 image-plate X-ray detector. To maximize the signal-to-noise ratio without saturation, the exposure time was set to 30 seconds for diffraction pattern acquisition. To increase the data points recorded in the elastic deformation region, the second beamline experiment was conducted using a load-control mode. A load step of 10 N in the elastic region and 20–50 N in the plastic region were used. The monochromatic X-ray with energy about 52 keV ($\lambda = 0.2394$ Å) and beam size about 400 µm × 400 µm was used for the second experiment while the data acquisition process was the same as that used in the first experiment.

The dislocation density in the fractured tensile specimen was measured by TEM for both unaged and aged conditions. TEM lamellae were lifted from the gauge zone using a FEI HELIOS 600i focused ion beam (FIB). The dislocation density was measured using the scanning transmission electron microscopy (STEM) mode at selected zone axises. All TEM characterization was performed using a FEI Tecnai F30 S/TEM at the Microscopy and Characterization Suite (MaCS) of the Center for Advanced Energy Studies (CAES) in Idaho Falls, ID.

APT characterizations were conducted to quantify the spinodal decomposition and G-phase precipitates in the ferrite phase of neutron irradiated CF-8 cast stainless steel. Reconstructions and analyses were performed using Cameca's Integrated Visualization and Analysis software 3.8.0 (IVAS). Three-dimensional (3-D) reconstructions were conducted by following the standard procedure of Recon Wizard in the software, and SEM tip images were used to define tip profiles.

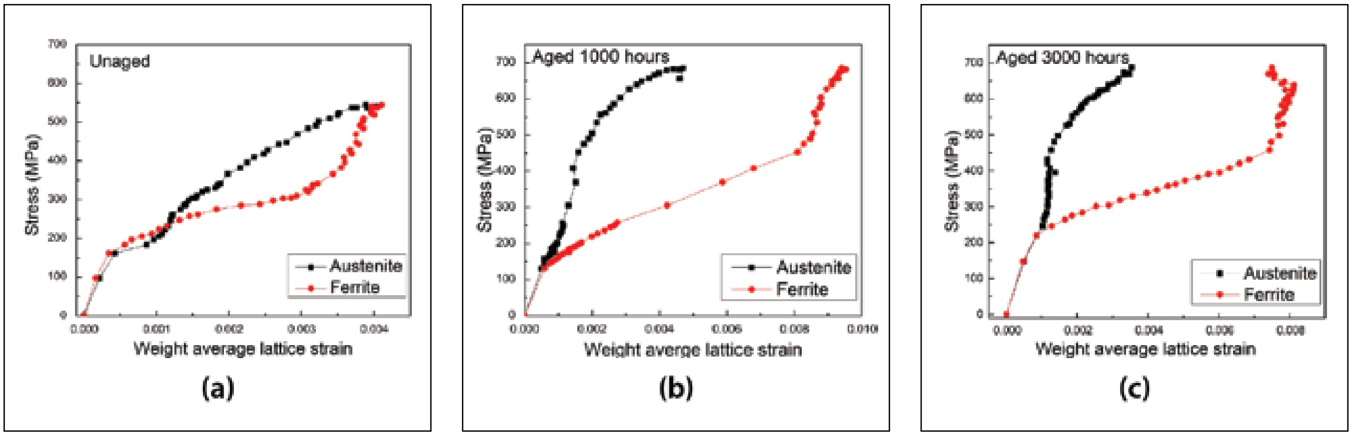

Figure 2. Stress vs. weight average lattice strain for unaged, 1000 hours aged and 3000 hours aged specimens, respectively.

Developing a mechanistic understanding of mechanical response of duplex stainless steel upon long-term reactor service is critical for component life evaluation and reactor license renewal.

Results

Figures 1b and 1c show the engineering strain vs. engineering stress and true strain vs. true stress curves, respectively, from the first beamline experiment using displacement control mode. It can be seen that thermal aging for 1000 hours greatly changes the mechanical behavior of the materials, which exhibit increased yield strength but reduced ductility. To better understand the load partition between the austenite and ferrite phase under different aging conditions, the weight average lattice strain was calculated. The average bulk lattice strain for austenite was derived from the lattice strains of the {111}, {200}, {220}, {311}, and {420} reflections while, for ferrite phase, it was derived from the lattice strains of the {110}, {200}, and {211} reflections. The weighted averaging algorithm was developed by Daymond [6]. The stress vs. weight average lattice strain for the first experiments are plotted in Figures 2a–c for the conditions of unaged, aged for 1000 hours, and aged for 3000 hours, respectively. The yield of austenite and ferrite phases were then determined based on ToMOTA's theory [7]. It clearly showed that the yield strength of the ferrite significantly increases from 315 up to 437 MPa upon thermal aging for 3000 hours, while the yield strength of austenite remains nearly unchanged at around 210 MPa.

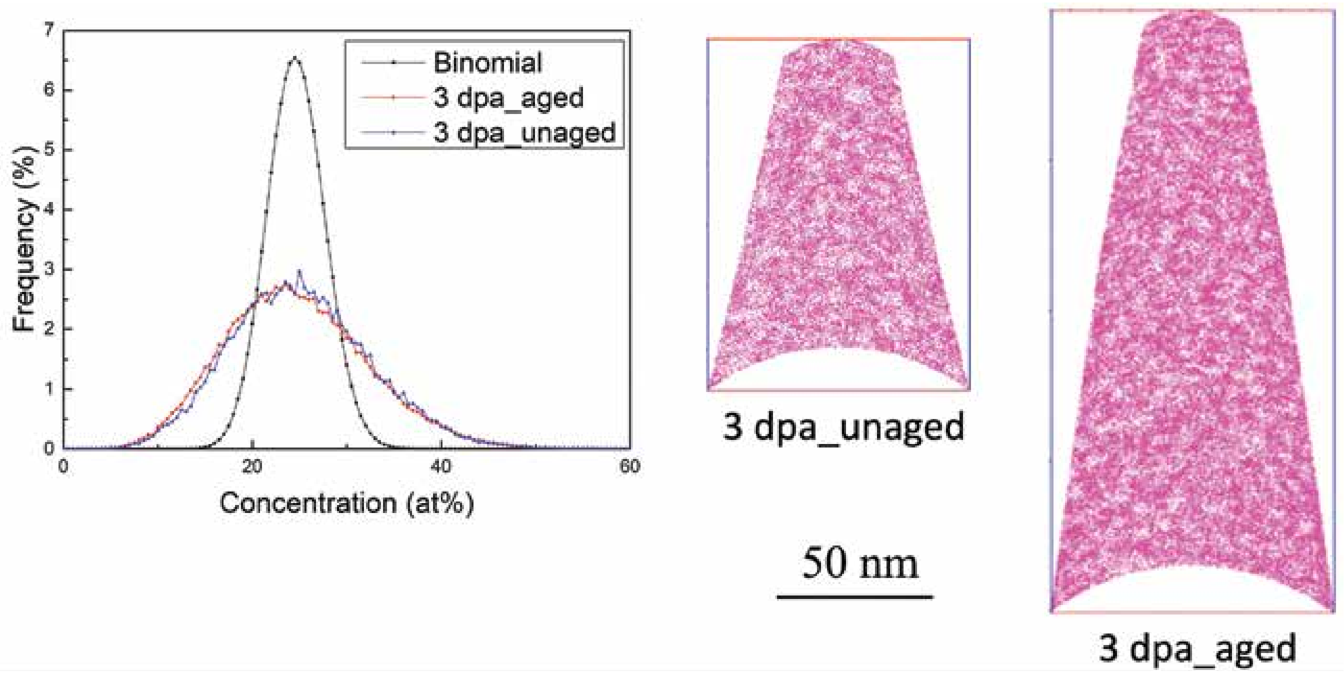

Figure 3. Atom probe tomography results (a) Cr elemental frequency distribution in ferrite phases and (b) spinodal decomposition of Cr in irradiated ferrite phases with or without prior thermal aging.

APT results show that neutron irradiation to 3 dpa induces significant spinodal decomposition and G-phase precipitation, as shown in Figures 3 and 4 for the materials without and with prior thermal aging. By using the Cr-Cr radial distribution function, the spinodal decomposition wavelength and amplitude were quantified as 13.2 nm/16.72 at.%, and 12.4 nm/16.99 at.% for the 3 dpa irradiated-unaged and 3 dpa irradiated-aged specimens, respectively. Thermal aging prior to neutron irradiation has nearly no impact on the level of spinodal decomposition in the ferrite beams completely overshadowed by neutron irradiation. This fact confirmed using the Cr elemental frequency distribution analysis, shown in Figure. 3a. The G-phase precipitates (Mn-Ni-Si clusters) were quantified using a widely accepted maximum-separation method (MSM) [8–9]. The measured number density and mean size are $(1.48 \pm 0.17) \times 10^{24}\ m^{-3}/1.06 \pm 0.05$ nm and $(1.44 \pm 0.06) \times 10^{24} m^{-3}/1.06 \pm 0.03$ nm for the 3 dpa irradiated unaged and 3 dpa irradiated aged conditions, respectively. The G-phase precipitates have a nearly identical volumetric fraction of around 0.75% in the ferrite phase in those two specimens.

Discussion

The *in-situ* X-ray tensile testing clearly showed the hardening of ferrite phase upon thermal aging, and it is anticipated that the ductility of

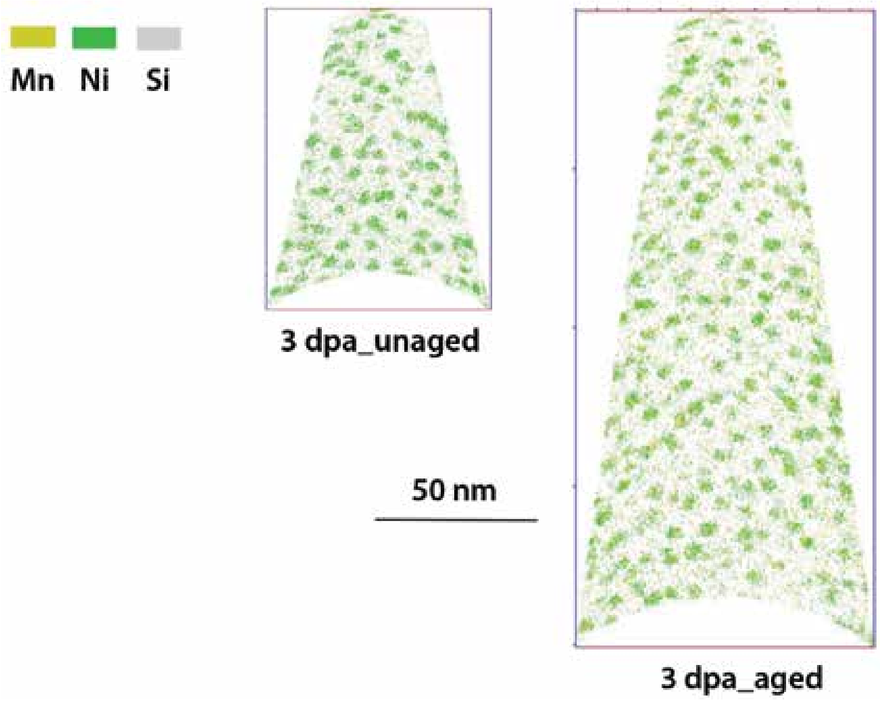

Figure 4. G-phase (Mn-Ni-Si) precipitates in irradiated ferrite phases.

ferrite would decrease subsequently. Limited study showed that the cracks initiate in hardened ferrite phase and propagate along the phase boundary, which reduce the overall fracture toughness of the material [10]. The APT microstructure characterization on the 3 dpa irradiated duplex stainless steel clearly displayed that the neutron irradiation introduced very similar phase evolutions in the ferrite phase as compared with thermal aging. With additional data collected from the samples irradiated at 5, 10, 20 and 40 dpa, it was found that neutron irradiation induced spinodal decomposition started to saturate at between 10 and 20 dpa. However, it is generally agreed that the decrease of bulk fracture toughness starts to saturate at 5 dpa. The discrepancy is attributed to the austenite phase becoming hardened and embrittled and essentially determining the bulk mechanical properties.

The correlation between the microstructural changes and mechanical responses was established through the mechanistic understanding and finite element modeling developed in the project. Prediction can be made based on the microstructural data from small samples exposed to long-term thermal aging with or without neutron irradiation conditions, that normally poses significant challenges in experimental measurements due to the limit of surveillance samples. By simply characterizing the microstructures of the specimens extracted from reactor-surveillance samples or salvaged components, the related mechanical properties can be derived, and expected service life can be assessed. The research project directly contributes to the construction of a physics-based model for predicting material performance for reactor license renewal and regulation.

Conclusion

In-situ tensile testing using WAXS and post-tensile microstructural characterizations show that thermal aging induces a significant hardening effect on the ferrite phase and reduces overall ductility of the bulk material. Thermal aging shifts the load partition onto the ferrite phase, and the enhanced strain misfit introduces stress concentrations and promotes nucleation of pores at the phase boundary.

Future Activities

Further microstructural characterizations on the neutron irradiated spcimens and tensile fractured specimens using TEM and APT will provide a better interpretation of the results from the X-ray *in-situ* tensile testing. Ultimately, this data will help us to build a model to correlate the mechanical response with microstructural evolution in cast stainless steels.

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