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In-Situ Ion Irradiation of Nanostructured Ferritic Alloys

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Nanostructured ferritic alloys (NFAs) are a new class of steels that are of great interest for potential applications in advanced nuclear reactors, such as for fuel cladding in fission reactors and first-wall structural components in proposed fusion reactors. Instead of coarse oxides found in similarly produced oxide-dispersion-strengthened (ODS) steels, NFAs contain high concentrations ($>10^{23}$ m^{-3}) of Ti-, Y-, and O-enriched nano-clusters (NC) or nano-features (NF) that are typically <4 nm diameter. Although these small particles are not fully understood, they are clearly different from equilibrium oxide particles. Atom-probe tomography (APT) reveals NC have Ti>Y and (Ti+Y)/O>1, no second-phase diffraction from them has been obtained with electrons or X-rays, and amazingly they survive extended annealing at temperatures in excess of 1400°C, which is $>0.9T_M$. For nuclear applications NFAs are expected to be highly radiation-tolerant and this is supported by early data. The concept is that the small oxides will be the dominant sink in the microstructure, enhancing vacancy-interstitial recombination by trapping vacancies and mitigating He embrittlement by trapping transmutation-produced He in small, high-pressure cavities rather than in large bubbles at grain boundaries [1]. The stability of the nano-oxides under irradiation and their effect on the development of damage microstructures are being studied with neutron and ion irradiations. Since the NCs are inhomogeneously distributed (more so in some alloys than others) [2,3], only gross changes in the NCs following irradiation are detectable. However, by following a specific area, in-situ ion irradiation of transmission electron microscopy (TEM) specimens could potentially reveal more subtle changes due to irradiation. A complication is that NCs can be reliably imaged by TEM only with energy-filtered TEM (EFTEM) methods such as Fe-M jump-ratio imaging [4]. For elevated-temperature in-situ irradiations, the thin regions (~30 nm) required for EFTEM of NCs raise special concerns about the specimen surfaces acting as a sink for irradiation-produced point defects and about potential interstitial-impurity (e.g. O, C or N) pick-up or even oxidation.

Two exploratory approaches to in-situ irradiation of NFAs have been initiated. In the first, electropolished disk specimens of 14YWT (Fe-14.2wt.%Cr-1.95%W-0.22%Ti-0.25%Y_2O_3) were irradiated at the IVEM-Tandem facility [5] at Argonne National Laboratory (ANL) with 1-MeV Kr^{++} ions to a dose of 1 x 10^{15} ions/cm^2 [~3 displacements per atom (dpa)] at room temperature (RT) and 500°C. For the specimen irradiated at RT NCs imaged by EFTEM before irradiation were no longer visible in the same areas following irradiation. NCs were observed after irradiation at 500°C, but pre-irradiation reference images were not available to evaluate irradiation-induced changes. In the second approach, sequential in-situ irradiations and EFTEM imaging were performed at the JANNuS-Orsay facility [6], which includes EFTEM capability on the TEM that is coupled to the accelerators. Electropolished specimens were tilted 68° for in-situ irradiation with 250-keV Fe^+ ions from the ARAMIS accelerator, but EFTEM characterization was performed at 0° tilt. A specimen of 14YWT was sequentially irradiated at RT to 0.36, 1.5, 2.5 and 4.8 x 10^{15} ions/cm^2 (~0.6, 2.5. 4.2 and 8 dpa based on SRIM [7] calculations) with corresponding irradiation times of 1, 3.67, 5, and 8.5 h. Sequential EFTEM imaging was complicated by specimen buckling but it was possible to follow individual NCs in Fe-M jump-ratio images as a function of dose. Surprisingly, changes were first apparent for larger particles (≥10 nm diameter), the image-contrast of which became more diffuse before disappearing. With increasing dose smaller particles were similarly affected; eventually by 8 dpa the majority were no longer visible (Fig. 1). Following heating for 10 min at 500°C without irradiation, a disk specimen of 14YWT was sequentially irradiated at 500°C to 0.32 and 1.32 x 10^{15} ions/cm^2 (0.5 and 2.2 dpa, 1 and 3 h). Specimens were un-tilted and cooled for EFTEM characterization. The major change observed was progressive oxidation of the specimen that was apparent even after 10 min heating without irradiation. Grain boundaries were preferentially oxidized and a zone of complete oxidation that started at the specimen edge progressed to thicker

areas. In regions thick enough to avoid complete oxidation some NCs survived with no change in size. Other NCs disappeared, presumably consumed or obscured by thickening surface oxide films. A specimen of MA957 (Fe-14wt%Cr-1%Ti-0.3%Mo-0.27%Y_2O_3) subjected to a similar irradiation at 500°C (1.0 and 2.0 x 10^{15} ions/cm^2, 1.6 and 3.3 dpa, 1 and 3 h) exhibited similar overall behavior.

Although NCs in a Japanese 9Cr NFA were observed after in-situ irradiation with 300-keV Fe ions to 12 dpa at 25°C [3], dissolution of NCs was observed in both RT irradiations of the present work. Such dissolution is attributed to cascade mixing with limited possibility of back diffusion to reform NCs. This behavior may be of limited technological importance since extended irradiation at low temperatures is not envisioned for potential applications. However, the temperature range over which the effect occurs has not been determined. The pronounced oxidation during 500°C irradiations at the JANNuS-Orsay facility is likely related to vacuum quality and to time-at-temperature rather than dose. Such oxidation presents a major barrier to elevated temperature in-situ ion-irradiation studies of NFAs. For the 500°C irradiations at the IVEM-Tandem facility, where gross oxidation was absent, the potential absorption of interstitial impurities is still a concern [8].

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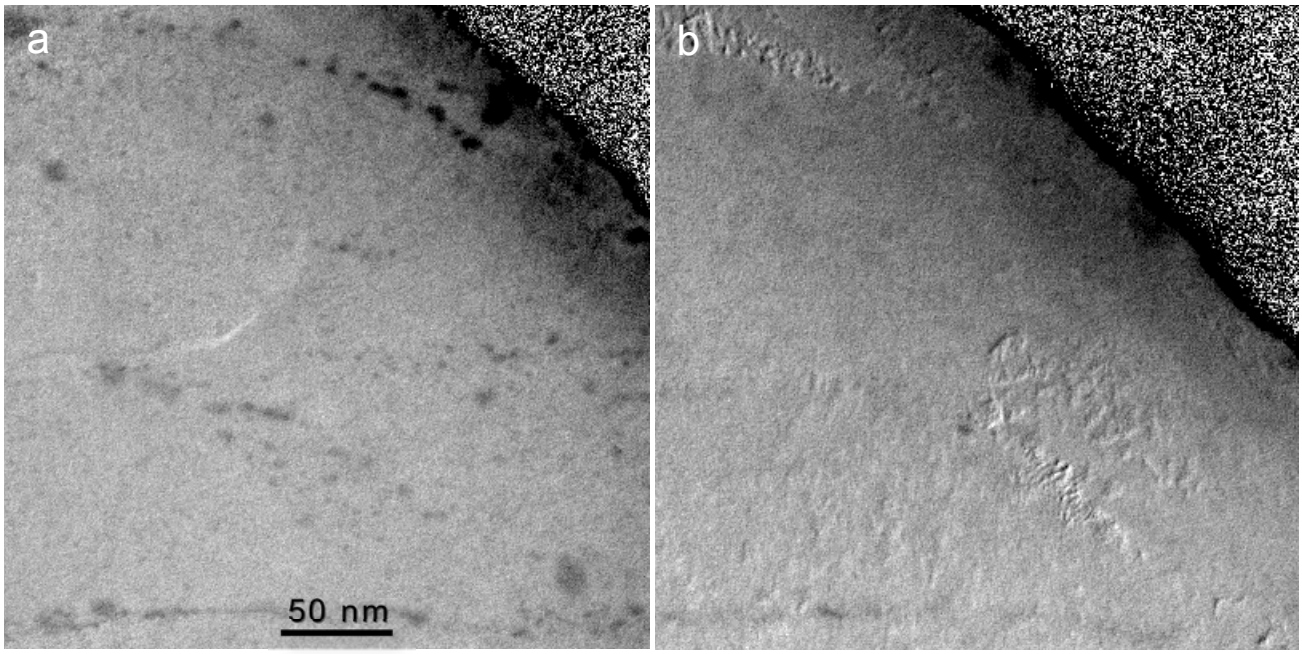

Fig.1. EFTEM Fe-M jump-ratio images of NCs in 14YWT. (a) Pre-irradiation. (b) The same region following in-situ irradiation at RT with 250-keV Fe^+ ions to a dose of 4.8 x 10^{15} ions/cm^2 (~8 dpa).

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