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Stability of nanosized oxides in ferrite under extremely high dose self ion irradiations

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ABSTRACT

A nanostructured ferritic alloy (NFA), 14YWT, was produced in the form of thin walled tubing. The stability of the nano-oxides (NOs) was determined under 3.5 MeV Fe^{+2} irradiations up to a dose of ~585 dpa at 450 °C. Transmission electron microscopy (TEM) and atom probe tomography (APT) show that severe ion irradiation results in a ~25% reduction in size between the unirradiated and irradiated case at 270 dpa while no further reduction within the experimental error was seen at higher doses. Conversely, number density increased by ~30% after irradiation. This 'inverse coarsening' can be rationalized by the competition between radiation driven ballistic dissolution and diffusional NO reformation. No significant changes in the composition of the matrix or NOs were observed after irradiation. Modeling the experimental results also indicated a dissolution of the particles.

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1. Introduction

Advanced nuclear systems can provide enhanced energy generation efficiency, safety and reliability, but some reactor designs require improved radiation tolerance of more than 200 displacements per atom (dpa) at high operation temperatures up to 1000 °C [1]. Nanostructured ferritic alloys (NFAs) are attractive materials for Generation IV reactors because of their excellent high temperature strength, thermal stability, creep resistance and radiation tolerance, provided by a high density of Y-Ti-O (<5 nm) nano-oxides (NOs) [2–4]. Atom probe tomography (APT) studies have been interpreted to suggest that NOs are nonstoichiometric phases with low O/(Y + Ti) ratio, while transmission electron microscopy (TEM) and X-ray diffraction (XRD) studies have shown that most NOs are pyrochlore $Y_2Ti_2O_7$ structure having Y/Ti ≈ 1 [5–7].

NOs can pin grain boundaries and dislocations, leading to high strength and superior structural stability [2–4]. Moreover, they may act as recombination centers for both point defects created from neutron damage and trapping sites for helium atoms created by transmutation reactions [2–4]. Overall, NFAs have remarkable radiation tolerance, but the stability of the enabling NOs under extreme irradiation conditions needs to be systematically tested.

The stability of NOs has been studied extensively under both neutron and heavy ion irradiations, but most of these previous studies were limited to moderate damage levels (< 200 dpa). In a brief summary, Yamashita et al. reported that small oxides dissolve under neutron irradiation in 11Cr and 13Cr alloys irradiated in the experimental fast reactor JOYO up to ~20 dpa at 450–561 °C [8]. Yamashita et al. also found that NO density decreases in MA957 after neutron irradiation at 500 °C up to 100 dpa [9]. However, Mathon et al. reported that NOs are stable at 325 °C up to 5.5 dpa in neutron irradiated MA957 [10]. Similarly, Gelles found no significant changes in the NO distribution in MA957 irradiated in the FFTF/MOTA at 420 °C to 200 dpa [11]. Ribis et al. showed that the diameter of the NOs in MA957 are almost the same after neutron irradiation up to 50 and 75 dpa at 412 and 430 °C in the same alloy

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[12–14]. For the same alloy, Bailey et al. reported that, while the diameter and number density of NOs remain almost the same at 550 and 670 °C, the diameter of the NOs decreased just slightly and density increased considerably at 412 °C after ~110 dpa irradiations in FFTF-MOTA [15]. The NOs in 9Cr and 12Cr ODS alloys were also reported to be stable under neutron irradiations at 330–500 °C up to 15 dpa [16]. Similar to the neutron irradiations, NOs were found to be stable after ion irradiation up to 10 dpa at 300 and 500 °C in 12YWT and 19Cr-ODS steels [17,18]. The same behavior was observed in various ferritic ODS alloys for 20 dpa ion irradiations at 200, 500 and 700 °C, as well as for 60 dpa at 650 °C, and for 150 dpa at 670 °C [19–21]. In contrast, Allen et al. concluded that the diameter of the NOs decreases while their density increases at the temperatures 500–700 °C under Ni ion irradiations up to 150 dpa in 9Cr ferritic/martensitic alloys [22]. Parish et al. [23] and Certain et al. [24] reported that at low temperatures (below 300 °C) the NOs dissolve under heavy ion irradiations as a result of ballistic mixing, while at and above 300 °C they are either stable or slightly increased in diameter in 14YWT and 9Cr ODS alloys. On the other hand, He et al. [25] reported that while the size and density of the NOs decrease at 300 °C, NOs are stable at higher temperatures in 14YWT, Ni ion irradiated to 100 dpa.

A significant difference between various studies is that neutrons in test reactors are typically at a damage rate of 10^{-6}-10^{-8} dpa/s, while ion irradiation studies are usually at 10^{-2}-10^{-4} dpa/s. In addition to the dpa rate, temperature, and the size and composition of the NOs are the factors affecting their stability under radiation [26,27]. While there may be a number of heats of MA957, the literature shows that they closely resemble one another as well as the 14YWT alloys with respect to Y-Ti-O NOs with some variability expected for slightly different compositions and processing routes [28]. In this particular case, TEM and APT studies show that the NOs in the 14YWT tubing are very similar to those in MA957.

The references cited above were conducted at different damage rates and with slightly different NO compositions and sizes, thus the issue of NO stability has not been resolved, especially at the temperatures above 300 °C and doses greater than ~200 dpa. In contrast to previous studies, here we examine the irradiation response of a NFA 14YWT under extreme radiation damage levels up to ~585 dpa at 450 °C at a comparable damage rate with the above cited references. Notably, the NFA studied here is in the form of an extruded tube targeting an actual application as fuel cladding.

2. Experimental

The 14YWT heat called FCRD NFA-1, with a nominal composition of 14Cr-3W-0.4Ti-0.21Y-Fe wt.% was developed in an extensive collaboration between Los Alamos National Laboratory, Oak Ridge National Laboratory, University of California, Santa Barbara and Case Western Reserve University. In this work, we examine NFA-1 in the form of tubing that was fabricated by hydrostatic extrusion of a mandrel mounted mother tube cut from an extruded and cross-rolled plate. Further details can be found in Ref. [29].

Samples extracted from the tubes were irradiated at the Texas A&M University Ion Beam Laboratory by 3.5 MeV Fe^{2+} ions to reach peak displacement damage levels of 500, 700, 900 and 1100 dpa at 450 °C. The corresponding ion fluences are 4.9×10^{17}, 6.9×10^{17}, 8.8×10^{17} and 1.1×10^{18} cm^{-2} for 500, 700, 900 and 1100 peak dpa, respectively. The 6 × 6 mm ion irradiated area was achieved by defocusing rather than rastering the beam, since the latter has been shown to suppress void swelling [30–33]. The beam current was controlled at ~200 nA to minimize local heating. The sample temperature was monitored and controlled by a thermocouple mounted on the face of the hot stage that provided feedback to the heater controller. Temperature fluctuations during the irradiation were less than ±5 °C. The peak displacement rate was 1.7×10^{-3} dpa per second.

Fig. 1 shows the calculated damage and Fe implantation profiles for 1100 peak dpa irradiation, obtained from SRIM 2013 code [34]. The SRIM calculations used the Kinchin-Pease model with an Fe displacement energy of 40 eV. The projected range (R_p) of 3.5 MeV Fe^{+2} ions is ~1.2 μm, while the dpa peaks at ~1.0 μm. The injected Fe atoms exceed 10 at.% at depths beyond 700 nm for the 1100 peak dpa irradiation. Therefore, in order to minimize the effects of injected interstitials, the microstructural characterization focused on a sampling depth region from 400 to 600 nm, shown by the shading in Fig. 1. At this depth interval, the concentration of injected atoms, even at 1100 peak dpa, is less than 3 at.%. The average local doses in the sampled region for 500, 700, 900 and 1100 peak dpa were ~270, 375, 480 and 585 dpa, respectively.

The microstructures prior to and after irradiation were characterized by Transmission Electron Microscopy (TEM) and Atom Probe Tomography (APT). Samples were punched in 3 mm disks from the faces of the cladding tubes. Pre-irradiation TEM foils were prepared by mechanical polishing followed by jet electropolishing using a solution of perchloric acid (5%) and methanol at −40 °C with an applied voltage of 20 V. Those foils were thinned down to electron-transparent thickness. Both TEM and APT studies were conducted on those 3-mm electron-transparent TEM foils. On the other hand, the foils for the irradiations were jet electropolished on one side for ~30 s in order to clean the surface, while avoiding forming a deep dimple. After irradiation, standard FIB lift-out techniques, followed by low energy cleaning at 1 kV, were used to prepare the electron transparent TEM samples in an FEI Helios Nanolab 600 dual beam focused ion beam (FIB) instrument. The TEM studies were carried out on a FEI Tecnai F30 TEM operating at 300 kV; a Gatan image filter (GIF) was used for energy filtered TEM (EFTEM) foil thickness measurements. A subset of data was imaged both in EFTEM and bright field (BF) TEM modes and the two techniques agreed with each other within ~10%. However, since BF methods are convenient and have been widely used to image the NOs [35,36 as examples only] (which are a diffracting phase with a lower density than the ferrite matrix), this technique was adopted for characterization of the full test matrix. Notably BF TEM also yields results that compare well with APT data (see below).

APT specimens were prepared from the electropolished foils

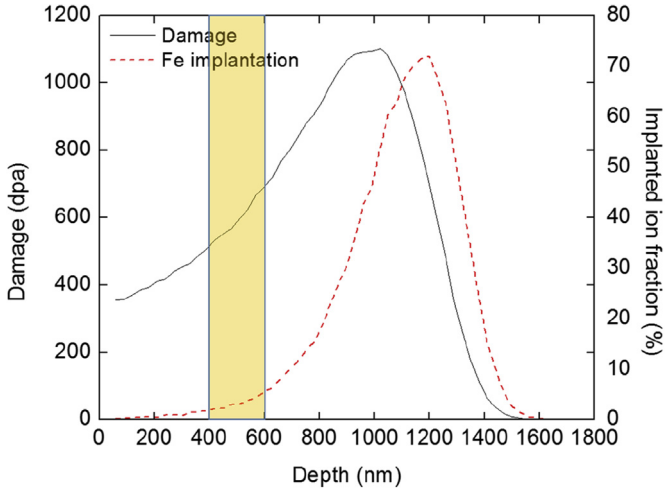

Fig. 1. SRIM-calculated depth profiles of damage and implanted Fe atoms for 1100 peak dpa irradiation in pure Fe. Shaded area shows the sampling depth region from 400 to 600 nm.