Journal of Nuclear Materials 477 (2016) 273–279

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Short communication

Methodology for determining void swelling at very high damage under ion irradiation

E. Getto [a, *], K. Sun [b], S. Taller [a], A.M. Monterrosa [a], Z. Jiao [a], G.S. Was [a, b]

[a] *Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI 48109, United States*
[b] *Department of Materials Science Engineering, University of Michigan, Ann Arbor, MI 48109, United States*

ARTICLE INFO

Article history:
Received 4 April 2016
Received in revised form
11 May 2016
Accepted 14 May 2016
Available online 18 May 2016

Keywords:
Void swelling
Ferritic-martensitic alloys
Radiation effects
Ion irradiation

ABSTRACT

At very high damage levels in ion irradiated samples, the decrease in effective density of the irradiated material due to void swelling can lead to errors in quantifying swelling. HT9 was pre-implanted with 10 appm He and subjected to a raster-scanned beam with a damage rate of ~1 × 10^{-3} dpa/s at 460°C. Voids were characterized from 0 to 1300 nm. Fixed damage rate and fixed depth methods were developed to account for damage-dependent porosity increase and resulting dependence on depth. The fixed depth method was more appropriate as it limits undue effects from the injected interstitial while maintaining a usable void distribution. By keeping the depth fixed and accounting for the change in damage rate due to reduced density, the steady state swelling rate was 10% higher than calculation of swelling from raw data. This method is easily translatable to other materials, ion types and energies and limits the impact of the injected interstitial.

1. Introduction

Prediction of material behavior and response to high irradiation damage levels remains a key challenge facing the next generation (Generation IV) fast reactor community. By virtue of their accelerated damage rates, self-ion irradiations [1–6] have been used to interrogate materials at high displacements per atom (dpa). This means that the high damage regime can be accessed both consistently and systematically, enabling rapid assessment of high damage material response. Void swelling in candidate structural steels, such as ferritic-martensitic steel alloys, is a high damage phenomenon that can lead to dimensional instability. Neutron irradiations of HT9 [7,8] have yet to reach swelling values above 2% despite exposure in reactor for durations of 7 years or more. Conversely, self-ion irradiations has been used to reach swelling of several percent in a matter of days, enabling the timely study of the void swelling response in the steady state swelling regime [2,3,6,9].

Ion irradiations are limited in penetration depth and have a strongly peaked damage distribution, requiring careful quantification of void swelling. The concern is that at high swelling the effective density of the target material decreases, altering the ion damage depth profile and resulting in an extension of the ion range, relative to that of an as-received sample. This is not a consideration when the swelling is low [2,3,10–12] or for neutron irradiated materials. The influence of voids on the ion damage curve is important in the steady state swelling regime when volume increases begin to exceed a few percent. At 200 dpa and beyond, accurate characterization of the microstructure is essential to understanding the irradiation induced or radiation enhanced features that cause the materials degradation at such high damage levels, thus ensuring safety and reliability in next generation reactors. In particular, accurate experimental data is needed for the development of predictive models of void swelling to levels that may limit component lifetime.

Several attempts have been made to account for the effect of voids on the ion damage distribution. Odette et al. [13] characterized the effect of voided samples on particle range and energy deposition in pure iron. The initial swelling profile was approximate, not based upon an actual experimental profile. Energy deposition was updated by an approximate swelling profile. With increased overall void swelling, the energy deposition curve as a function of depth into the sample was found to have a “flattened” profile and the ions were found to have an extended range into the sample.

Using the framework developed by Odette et al. [13], Johnston et al. [14] experimentally determined the swelling profile of 5 MeV

---

* Corresponding author.
E-mail address: embecket@umich.edu (E. Getto).

http://dx.doi.org/10.1016/j.jnucmat.2016.05.026
0022-3115/

Ni-irradiated stainless steel using systematic electropolishing to cross section the irradiated area to various depths within the sample. A swelling vs. depth profile was determined, which was then compared to the calculations of Odette et al. They found agreement within experimental error of the corrected damage curve from Odette, but the method was deemed too time consuming to repeat for most irradiation cases, as this was prior to the development of both SRIM [15] and focused ion beam technology for easier cross sectional sample preparation.

Swelling profiles in 46.5 MeV Ni-irradiated Ni, and separately, in 20 MeV C-irradiated pure Ni were measured to verify displacement curves calculated by Nelson et al. [16] The swelling profile was then applied as an additional method to verify that the damage distribution calculation was performed correctly since this was prior to the development of SRIM, which had the capability to handle discrete layers of materials with differing densities. This method was then applied to 316 stainless steel irradiated at 525 °C by Mazey et al. [17] as a method to approximate the total damage. However, saturation of swelling was observed which casted doubt on the validity of the correction method. Additionally, injected interstitial effects were not taken into account near the end of range of the particle. The injected interstitial was later shown to suppress void swelling by Kumar and Garner [18].

The results of Odette et al. [13] and Johnston et al. [14] are especially promising, considering recent improvements in both damage distribution calculation, ion beam irradiation reliability and sample preparation technology. Thus, a systematic characterization of the swelling profile at very high damage/swelling and the effect on damage distribution is feasible. The objective of this paper is to develop a methodology for accurately accounting for the effect of void swelling on ion damage depth profiles.

2. Materials and Methods

Archival material from ferritic martensitic alloy HT9 (Heat 84425) removed prior to use in the ACO-3 duct in FFTF [8], was irradiated in this study. The composition of this alloy is Fe-11.8Cr-1.03Mo-0.51Ni-0.5Mn-0.33V-0.24W-0.21C-0.21S with trace amounts of N, P and S. HT9 was heat treated at 1065 °C/30 min followed by air cooling to room temperature then was tempered at 750 °C/60 min followed by air cooling to room temperature, which resulted in a tempered martensitic structure [8,19]. This alloy was previously examined under both neutron [8] and ion irradiation [1–3,9] for void swelling and other microstructure characterization. Additional results from this irradiation series are presented in detail in Ref. [9] along with a full description of the irradiation procedure, and a brief summary is provided here. After mechanical polishing up to 0.25 μm, specimens were electropolished in a 90% methanol and 10% perchloric acid solution. Samples were then preimplanted with 10 appm helium at room temperature using a 400 keV National Electrostatics Corporation ion implanter. Helium preimplantation was performed at 80, 140, 220, 310 and 420 keV to yield an approximately flat (±10%) implant profile over the range 300–1000 nm from the irradiated surface. After helium implantation, irradiations were performed with a 3 MV Pelletron accelerator with 5 MeV Fe^{++} ions in raster-scanning mode with a frequency of 255 Hz in the x axis and 1055 Hz in the y axis. Nominal damage was calculated at a depth of 600 nm using the Quick Kinchin Pease Mode in SRIM with displacement damage of 40 eV. All damage levels reported refer to dpa at a depth of 600 nm [15]. Damage rates varied in the range $0.6–1.2 \times 10^{-3}$ dpa/s and were irradiated from 75 to 650 dpa at a temperature of 460 °C.

Samples for transmission electron microscopy were made using the Focused Ion Beam (FIB) lift-out method, which were then thinned to thicknesses below 150 nm. At least two lift-outs per irradiation condition were analyzed. Samples were examined in either a JEOL 2010F or 2100F analytical electron microscope (AEM). Bright field (BF) and high angle annular dark field (HAADF) images for void analysis were taken in scanning transmission electron microscopy (STEM) mode at a magnification of 50,000×. Thickness was measured using the electron energy loss spectroscopy (EELS) low loss method. Voids were tallied in 100 nm wide bins to a depth of 1300 nm. To avoid surface effects and the effect of the injected interstitial [2], the region of 500–700 nm was used for all analysis when no corrections were applied. To minimize the effect of the spatial heterogeneity of voids, an effort was made to count at least 50 voids per bin for a total of 500–4000 voids per irradiation condition in nominally 2 μm^2 of 120 nm thickness volume. Area examined is reported in Table 1. SRIM was used to calculate the damage in HT9 specimens. At least five SRIM runs of 100,000 ions each were calculated then averaged to remove fluctuations from SRIM at bin interfaces for the resulting figure as suggested in the SRIM manual [20]. Fig. 1 shows the damage vs. depth profile and the implanted ion distribution for 5 MeV Fe^{++} in HT9 at 0 dpa. After voids were profiled in depth, the resulting swelling profile was used to recalculate the damage profile at each of the 6 damage levels at which voids were observed. Using the swelling profiles, the SRIM calculation was adapted for the change in material density as a result of void swelling at each nominal damage, which assumes that effect of voids is homogenous throughout each layer. The updated densities were input into SRIM as discrete layers of 100 nm each. The effective density was determined as follows:

$$\rho_{effective} = \left(1 - \frac{\Delta V}{V}\right) * \rho_{HT9}, \qquad (1)$$

where $\rho_{effective}$ is the effective density, $\Delta V/V$ is the swelling and ρ_{HT9} is the density of HT9. The result of this calculation, i.e. the effective damage rate under ion irradiation with voids was used to determine an appropriate method for correction of the damage curve.

Table 1
Swelling as a function of damage of HT9 irradiated with 5 MeV Fe^{++} at 460 °C. Nominal swelling and nominal damage are compared to fixed damage correction.

No correction		Fixed depth correction		Area examined (μm^2)	Number of voids (500–700 nm)
Nominal damage (dpa)	Nominal swelling (%)	Worst case (dpa)	Average (dpa)		
0	0	0	0	N/A	N/A
75	0	75	75	2.3	Not observed
130	0.38 ± 0.05	130.3	130.2	3.6	829
250	3.35 ± 0.40	246.8	248.7	5.3	733
350	6.00 ± 0.72	341.0	344.4	3.9	437
450	8.81 ± 1.05	432.2	437.1	5.5	742
550	13.3 ± 1.6	517.7	525.4	4.1	396
650	16.0 ± 2.0	600.1	609.4	5.3	502
Steady state swelling rate (250–650 dpa)	0.033 ± 0.004%/dpa	0.036 ± 0.004%/dpa	0.036 ± 0.004%/dpa		