Journal of Nuclear Materials 501 (2018) 312–318

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Self-ion emulation of high dose neutron irradiated microstructure in stainless steels

Z. Jiao [a,*], J. Michalicka [b], G.S. Was [a]

[a] Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI, United States
[b] Brno University of Technology, CEITEC Nano Research Infrastructure, Brno, Czech Republic

ARTICLE INFO

Article history:
Received 19 June 2017
Received in revised form
19 December 2017
Accepted 29 January 2018
Available online 31 January 2018

Keywords:
Microstructure
Radiation-induced segregation
Radiation effects

ABSTRACT

Solution-annealed 304L stainless steel (SS) was irradiated to 130 dpa at 380 °C, and to 15 dpa at 500 °C and 600 °C, and cold-worked 316 SS (CW 316 SS) was irradiated to 130 dpa at 380 °C using 5 MeV Fe^{++}/Ni^{++} to produce microstructures and radiation-induced segregation (RIS) for comparison with that from neutron irradiation at 320 °C to 46 dpa in the BOR60 reactor. For the 304L SS alloy, self-ion irradiation at 380 °C produced a dislocation loop microstructure that was comparable to that by neutron irradiation. No voids were observed in either the 380 °C self-ion irradiation or the neutron irradiation conditions. Irradiation at 600 °C produced the best match to radiation-induced segregation of Cr and Ni with the neutron irradiation, consistent with the prediction of a large temperature shift by Mansur's invariant relations for RIS. For the CW 316 SS alloy irradiated to 130 dpa at 380 °C, both the irradiated microstructure (dislocation loops, precipitates and voids) and RIS reasonably matched the neutron-irradiated sample. The smaller temperature shift for RIS in CW 316 SS was likely due to the high sink (dislocation) density induced by the cold work. A single self-ion irradiation condition at a dose rate ~1000× that in reactor does not match both dislocation loops and RIS in solution-annealed 304L SS. However, a single irradiation temperature produced a reasonable match with both the dislocation/precipitate microstructure and RIS in CW 316 SS, indicating that sink density is a critical factor in determining the temperature shift for self-ion irradiations.

1. Introduction

Life extension of nuclear power plants to 60 years and beyond requires that some internal components such as baffle bolts or flux thimble tubes withstand irradiation damage levels in excess of 160 dpa (displacements per atom) [1]. Studying the effect of irradiation at such high damage levels requires either impractically long irradiations in test reactors, or accelerated irradiation with ions under conditions designed to emulate the microstructure produced in reactor.

Emulation of high dose neutron irradiated microstructures using self-ions (SIs), requires benchmarking against the irradiated microstructure of alloys that have been irradiated in reactor to high dpa. Samples from a solution-annealed 304L stainless steel (304L SS) core shroud and a cold-worked 316 stainless steel (CW 316 SS) baffle bolt that were irradiated in the BOR60 fast reactor to 46 dpa at 320 °C provide an opportunity for benchmarking self-ion irradiation. As part of the Cooperative IASCC Research (CIR) Program [2–4], dislocation loops, precipitates, cavities and radiation-induced segregation (RIS) at grain boundaries were characterized for both alloys. Dislocation loops, stacking fault tetrahedral (SFTs) and radiation-induced G-phase precipitates were observed in 304L SS. Dislocation loops and γ' precipitates were observed in CW 316 SS. Radiation-induced Ni enrichment and Cr depletion at the grain boundary were found in both alloys and the segregation magnitudes and profiles were characterized. No cavities were reported in either alloy due to the relatively low reactor irradiation temperature (320 °C).

Because self-ion irradiation will be conducted at a much higher damage rate (~10^{-3} dpa/s) than neutron irradiation (~5×10^{-7} dpa/s), the irradiation temperature must be adjusted to achieve the same total flow of defects to sinks (which drives processes like radiation-induced segregation), or the net flux of one defect over another (which drives processes like cavity or loop growth). The

* Corresponding author.
E-mail address: zjiao@umich.edu (Z. Jiao).

https://doi.org/10.1016/j.jnucmat.2018.01.054
0022-3115/

magnitude of the temperature shift can be estimated using Mansur's invariant relations [5,6], and is different for these two processes. Equations (1) and (2) represent the temperature shift required to maintain the total flow of defects (RIS), and the net flux of one defect over another (dislocation loops or voids), respectively. In these two equations, k is the Boltzmann's constant; K_0 is the damage (dose) rate. E_m^v. d E_m^f. e vacancy migration energy and formation energy, respectively.

$$T_2 - T_1 = \frac{\frac{kT_1^2}{E_m^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)}{1 - \frac{kT_1}{E_m^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)} \tag{1}$$

$$T_2 - T_1 = \frac{\frac{kT_1^2}{E_m^v+2E_f^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)}{1 - \frac{kT_1}{E_m^v+2E_f^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)} \tag{2}$$

Application of a single temperature shift has worked well for proton irradiation used in emulating irradiation of LWR components [7–9]. Neutron irradiation effects at 275 °C were successfully emulated by proton irradiation at 360 °C to doses up to 5 dpa in both 304 and 316 stainless steels [7]. Besides irradiated microstructures (dislocation loops and cavities), irradiation hardening and irradiation-assisted stress corrosion cracking (IASCC) also showed reasonable agreement between proton and neutron irradiations. A more recent survey [8] of comparison of irradiated effects on microstructures, microchemistry, hardening, and IASCC showed stunning similarities between proton irradiation at 360 °C with BOR60 irradiation at 320 °C for a set of austenitic stainless steels. The small temperature shift of 40 °C is reasonable because the damage ratio of ~100 is modest enough to keep the difference between the temperatures small. With self-ions, the ion/neutron damage rate ratio is closer to 1000 and the difference in magnitude of the shifts is correspondingly greater. Thus, it has yet to be established whether this technique can be used to match the full microstructure of irradiated austenitic stainless steels with a single temperature shift as can be done with protons.

The objective of this study is to determine if the complete reactor-irradiated microstructure can be emulated with a single self-ion irradiation condition in austenitic stainless steels.

2. Experiments

2.1. *Self-ion irradiation experiments*

Virgin materials of solution-annealed 304L SS and a cold-worked 316 SS (CW 316 SS) were used for comparison of the irradiated microstructure with the same heats irradiated to 46 dpa at 320 °C in BOR60 [2–4]. The chemical compositions of the alloys are given in Table 1. Characterization results of dislocation loops, voids, precipitates and RIS from Ref. [4] for both neutron irradiated 304L SS and CW 316SS at 46 dpa are summarized in Table 2. The 304L SS, originally from a core shroud block, was homogenized, cold-worked, and followed by solution annealing at 1050 °C for 30 min, resulting in an average grain size of ~40 μm. The CW 316 SS was from a baffle bolt rod with an estimated cold work of 20%. Samples for self-ion irradiations were fabricated using electrical discharging machine (EDM) in the form of bars of dimensions of 3 mm × 1.5 mm × 20 mm. Samples were mechanically polished using SiC paper up to #4000 grit followed by electropolishing for 30–60 s in a 90% methanol, 10% perchloric acid solution at −40 °C prior to self-ion irradiation.

Neutron irradiation experiments were conducted in the BOR60 fast reactor. The BOR60 irradiation temperature and dose rate were estimated to be 320 °C and 5×10^{-7} dpa/s, respectively, and were used as the reference neutron irradiation condition in the selection of matching self-ion irradiation conditions. The nominal BOR60 irradiation temperature of 320 °C was used although the actual irradiation temperature slightly varied from irradiation cycle to cycle, typically within 10 °C. Kiritani and coworkers [10,11] have demonstrated that the temperature-flux histories, particularly the short transient irradiation at a lower temperature during the startup of the reactor, can have a profound effect on defect microstructure evolution. As temperature history effect on irradiated microstructure evolution was not investigated for the BOR60 irradiation, it was not accounted for in selection of ion irradiation conditions. Parameters for the ion irradiation experiment are temperature, dose rate and dose. Fixing two allows the third to be determined using Mansur's invariant relations [6]. As the self-ion irradiation will be conducted at a dose rate of ~10^{-3} dpa/s, the temperature to match dislocation loop growth was estimated to be ~380 °C (temperature shift of 60 °C) based on Equation (2), and to match the RIS behavior, the temperature should be ~600 °C (temperature shift of 280 °C) based on Equation (1). A vacancy migration energy of 1.2 eV and a vacancy formation energy of 1.8 eV [12,13] were used for the temperature shift estimation.

Because of the large difference in temperature to match the different irradiation features, an irradiation temperature of 500 °C was also selected as a compromise to determine if a single irradiation temperature will capture both processes adequately. For neutron irradiations, irradiated microstructure features such as dislocation loops typically saturate after a few dpa [14]. The BOR60 irradiation dose of 46 dpa should then be well past the dpa for saturation. The dpa to reach microstructure saturation can also be affected by damage rate and temperature with higher saturation dpa expected for high dpa rate and lower saturation dpa expected for high temperature. Based on this argument, irradiations to 130 dpa at 380 °C and 15 dpa at 500 °C and 600 °C were selected for self-ion irradiations to ensure that all of the self-ion irradiations were well into the steady-state microstructure.

Irradiation experiments were conducted using a 5 MeV Fe^{++} or Ni^{++} raster-scanned beam in the 1.7 MV Tandem accelerator at the Michigan Ion Beam Laboratory (MIBL). The damage depth was ~2 μm with a peak at ~1.5 μm for 5 MeV Ni^{++} and slightly shallower for 5 MeV Fe^{++} (damage depth of ~1.5 μm with the peak at ~1.2 μm), Fig. 1. As specified, irradiations were conducted at 600 °C:15 dpa (irradiation temperature: irradiation dose) to match RIS and 380 °C:130 dpa to match dislocation loops. In addition, irradiation was also conducted at 500 °C:15 dpa as a compromise for dislocations/precipitates and RIS. The dpa was calculated using SRIM 2008 [15] with the Kinchin–Pease option (recommended by Stoller et al. [16]) and a displacement energy of 40 eV [17] for major elements, at the characterization depth (typically 600 ± 200 nm from the surface). The dpa rate at the characterization depth was ~10^{-3} dpa/s.

Table 1
Chemical compositions (wt%) of 304L SS and CW 316 SS.

Alloy	Fe	C	Mn	Si	P	S	Cr	Ni	Mo	N
304L SS	Bal.	0.023	1.82	0.56	0.023	0.015	19.95	10.80	0.53	0.072
CW 316 SS	Bal.	0.056	1.13	0.73	0.022	0.022	16.84	10.54	2.25	0.021