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Radiation-induced segregation on defect clusters in single-phase concentrated solid-solution alloys

Chenyang Lu [a], Taini Yang [a], Ke Jin [b], Ning Gao [c], Pengyuan Xiu [a], Yanwen Zhang [b, d], Fei Gao [a], Hongbin Bei [b], William J. Weber [d, b], Kai Sun [e], Yan Dong [e], Lumin Wang [a, e, *]

[a] *Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI, 48109, USA*
[b] *Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA*
[c] *Institute of Modern Physics, Chinese Academy of Sciences, Lan Zhou, Gansu, 73000, China*
[d] *Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN, 37996, USA*
[e] *Department of Materials Science and Engineering, University of Michigan, Ann Arbor, MI, 48109, USA*

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ABSTRACT

A group of single-phase concentrated solid-solution alloys (SP-CSAs), including NiFe, NiCoFe, NiCoFeCr, as well as a high entropy alloy NiCoFeCrMn, was irradiated with 3 MeV Ni^{2+} ions at 773 K to a fluence of 5×10^{16} ions/cm^2 for the study of radiation response with increasing compositional complexity. Advanced transmission electron microscopy (TEM) with electron energy loss spectroscopy (EELS) was used to characterize the dislocation loop distribution and radiation-induced segregation (RIS) on defect clusters in the SP-CSAs. The results show that a higher fraction of faulted loops exists in the more compositionally complex alloys, which indicate that increasing compositional complexity can extend the incubation period and delay loop growth. The RIS behaviors of each element in the SP-CSAs were observed as follows: Ni and Co tend to enrich, but Cr, Fe and Mn prefer to deplete near the defect clusters. RIS level can be significantly suppressed by increasing compositional complexity due to the sluggish atom diffusion. According to molecular static (MS) simulations, “disk” like segregations may form near the faulted dislocation loops in the SP-CSAs. Segregated elements tend to distribute around the whole faulted loop as a disk rather than only around the edge of the loop.

1. Introduction

Recently, a novel class of materials called single-phase concentrated solid-solution alloys (SP-CSAs), including high entropy alloys (HEAs), has drawn much attention. Unlike traditional alloys, SP-CSAs contain two or more principle elements in equal or near-equal molar ratios. These elements form random solid solutions in simple face-centered cubic (fcc) or body-centered cubic (bcc) lattice structures, which grant SP-CSAs extraordinary properties, such as high yield strength, high thermal stability and hardness, high-temperature strength, great wear and fatigue resistance, and excellent corrosion resistance [1–4]. Notably, SP-CSAs also exhibit promising radiation resistance as demonstrated by theoretical and experimental studies [5–9]. Because of these superior properties, SP-CSAs are considered as potential candidates for fission and fusion reactor applications.

High-level lattice distortion and compositional complexity in SP-CSAs could change the process of energy dissipation and facilitate the recovery of radiation damage in the very early stages of irradiation [5,6,8,10]. Zhang et al. found that chemical disorder effectively reduced the electron mean free path, electrical and thermal conductivities, which significantly delayed defect evolution during ion irradiation at room temperature [5]. Jin et al. found that equiatomic NiFe presented significant delay in damage accumulation and evolution compared to pure nickel and NiCo at room temperature [11]. Using cross-sectional transmission electron microscopy (TEM), Lu et al. demonstrated that defect clusters migrated slower in NiFe than in pure nickel and NiCo at room temperature [6]. This finding was confirmed by the molecular dynamics (MD) simulations from Aidhy et al. [10]. Similar performance persists even at elevated temperatures and at higher irradiation doses. Kumar et al. found that non-single phase FeNiMnCr exhibited good microstructural stability and mechanical

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[*] Corresponding author. Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI, 48109, USA.
E-mail address: lmwang@umich.edu (L. Wang).

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behavior under high temperature irradiation up to 10 displacement per atom (dpa) [9]. Jin et al. reported that HEA alloy NiCoFeCrMn showed much higher swelling tolerance than pure nickel when irradiated by 3 MeV Ni ions to a peak dose of 60 dpa at 773 K [7]. Lu et al. performed detailed studies on a group of SP-CSAs (from binary to quinary alloys) irradiated to high doses by cross-sectional TEM characterization. They observed a unique void and dislocation loop separation, successfully linking the distribution with the defect cluster migration behavior, and explained the intrinsic mechanisms of void resistance in SP-CSAs [12]. In spite of these efforts, further study is required to examine the evolution of dislocation loops and local chemical segregation, particularly as a function of the chemical composition.

Radiation-induced segregation (RIS) is a typical radiation-induced phenomenon caused by the preferential interaction between the flux of vacancy and interstitial point defects and the flux of solutes to point defect sinks [13], such as grain boundaries, dislocations and voids. RIS could result in serious degradation of mechanical properties of structural materials, such as radiation-induced hardening and embrittlement. Currently, most studies focus on the RIS behavior of grain boundaries. For instance, radiation-induced depletion of Cr at grain boundaries was observed in austenitic stainless steels for decades, and it was widely known as one of the contributing factors to irradiation-assisted stress corrosion cracking (IASCC) [14–16]. Kumar et al. claimed that FeNiMnCr HEA exhibited better resistance against RIS on grain boundaries compared to conventional Fe-20Cr-24Ni alloy [9]. On the other hand, few studies on RIS to the defect clusters have been reported. The small scale of defect clusters and low magnitude of RIS around them make the characterization very difficult. Nevertheless, such studies are important because RIS around defect clusters may alter the defect cluster evolution under continued irradiation and thus affect the mechanical properties of the material. For example, the austenite structure can be destabilized due to the depletion of Ni in the matrix while Ni atoms segregate to the void/dislocation and/or loop/grain boundaries caused by RIS [17]. Jiao [14] and Dong [18] observed RIS on dislocation loops in austenitic stainless steels using atom probe tomography (APT); however, the exact nature and structure of the loops could not be identified by APT. Another interesting and open question is what are the configurations of the segregated atoms near the loop. Do they form a ring around the edge of the loop, or a “disk-like” plate within the whole loop? APT results from Jiao [7] and Dong [18] indicate that the segregation is more like first scenario, but it is questionable whether the segregation regions of both perfect and faulted dislocation loops are the same.

In this study, conventional TEM characterization was applied in two-beam condition to compare the dislocation loop distribution in Ni and a group of Ni-containing SP-CSAs after high temperature ion irradiation. The mechanisms of irradiation-induced hardening in SP-CSAs can be correlated with a previous study [7]. RIS on defect clusters (loops and voids) was observed and analyzed by ultra-fast electron energy loss spectroscopy (EELS) system equipped on a Cs-corrected scanning TEM (STEM). Molecular static (MS) simulation has also been conducted to clarify the shape of the segregation region and reveal the intrinsic segregation mechanism.

2. Experiments

2.1. Alloys

Pure Ni, Co, Fe, Cr and Mn elemental metals (>99.9% pure) were used to prepare the alloy ingots by arc-melting. Detailed information of alloy preparation was described in a previous study [7]. Four Ni-containing single-phase fcc equiatomic alloys, single crystal binary NiFe, ternary NiCoFe, and quaternary NiCoFeCr as well as a polycrystalline quinary alloy NiCoFeCrMn (with millimeter size grains) were prepared. Prior to irradiation, the specimens were ground using up to #4000 grit SiC paper, followed by chemical-mechanical polishing with up to 0.05 μm colloidal silica polishing solutions. “Mirror-like” surfaces were achieved with roughness below 3 nm.

2.2. Ion irradiation

The specimens were irradiated with 3 MeV Ni^{2+} ions to a fluence of 5×10^{16} ions/cm^2 at 773 K in the Ion Beam Materials Laboratory (IBML) at the University of Tennessee. The flux was controlled at 2.8×10^{12} /cm^{2}s. A rastered beam was employed to ensure homogeneous irradiation. Ion irradiation doses and stopping range in samples were computed by SRIM 2013 assuming a displacement threshold energy of 40 eV in Kinchin-Pease option [19]. The damage and implanted ion concentration profiles are shown in Fig. 1. The region of 500 ± 100 nm with a dose about 38 ± 5 dpa was chosen for the statistic of loop distribution and chemical characterization, in order to avoid the artificial effects associated with the surface sinks and the injected interstitial effects. The studied region is highlighted as shown in Fig. 1. To be noted, the loop images shown in Fig. 2 are from a larger region for enhancing visualization and a better comparison.

2.3. Microstructural characterization, EELS data acquisition and analysis

Cross-sectional TEM foils from irradiated samples were all prepared by focused ion beam (FIB) lift-out techniques using a FEI Helios Nanolab Dualbeam workstation. In order to remove unwanted FIB-induced damage, which would confuse the observation of small defect clusters created by Ni ions, “Flash polishing” was subsequently conducted. Significant improvement of the TEM sample quality after “flash polishing” was presented in Ref. [6]. Prior to characterization, TEM foils were cleaned using a Fischione Plasma Cleaner to remove the carbonaceous contamination. Low-magnification loop characterization was performed in a JEOL 3011 TEM operated at 300 keV. A double Cs-corrected JEOL 3100R05 STEM operated at 300 keV was employed for STEM-bright field (STEM-BF) and high angle annular dark field (HAADF) imaging.

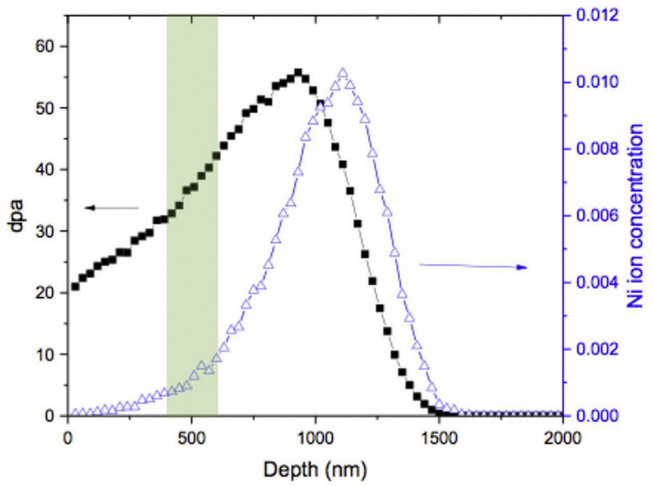

Fig. 1. Depth profiles of displacement per damage (dpa) and induced Ni ion concentration predicted by SRIM code for SP-CSAs irradiated with 3 MeV Ni^{2+} ions to a fluence of 5×10^{16} ions/cm^2.