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Anisotropic radiation-induced segregation in 316L austenitic stainless steel with grain boundary character

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Abstract

Radiation-induced segregation (RIS) and subsequent depletion of chromium along grain boundaries has been shown to be an important factor in irradiation-assisted stress corrosion cracking in austenitic face-centered cubic (fcc)-based alloys used for nuclear energy systems. A full understanding of RIS requires examination of the effect of the grain boundary character on the segregation process. Understanding how specific grain boundary structures respond under irradiation would assist in developing or designing alloys that are more efficient at removing point defects, or reducing the overall rate of deleterious Cr segregation. This study shows that solute segregation is dependent not only on grain boundary misorientation, but also on the grain boundary plane, as highlighted by markedly different segregation behavior for the Σ3 incoherent and coherent grain boundaries. The link between RIS and atomistic modeling is also explored through molecular dynamic simulations of the interaction of vacancies at different grain boundary structures through defect energetics in a simple model system. A key insight from the coupled experimental RIS measurements and corresponding defect–grain boundary modeling is that grain boundary–vacancy formation energy may have a critical threshold value related to the major alloying elements' solute segregation.

Keywords: Radiation-induced segregation; Grain boundaries; Ion irradiation; Atomistic modeling; Austenitic steel

1. Introduction

To improve existing and develop new structural alloys for nuclear energy systems, it is critical to tailor alloy microstructures for extended material lifetime, thus providing sufficient resistance to more demanding irradiation, higher temperature and corrosive environments. One approach to microstructural design is to produce alloys by increasing the density of specific types of grain boundaries (GBs). GBs are critical interfaces in materials and play a key role in a wide range of properties such as mechanical strength [1], radiation tolerance [2,3] and corrosion [4]. By controlling the distribution of GBs, a sophisticated microstructure that promotes the recombination or annihilation of irradiation-induced point defects at GBs, and also limits deleterious segregation of elements critical to chemical and mechanical alloy stability, can be created. Traditional materials used in these extreme environments, such as austenitic stainless steels, undergo non-equilibrium segregation and clustering of vacancies and interstitials created during high-energy collision cascades, which ultimately lead to changes in local microstructure and microchemistry [5,6]. A key result of this phenomenon is radiation-induced segregation (RIS), which is the preferential interaction between the flux of vacancy and interstitial

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point defects and the flux of specific alloying and solute elements to point defect sinks such as GBs [7–11].

It is well accepted that in 304L and 316L austenitic stainless steels, RIS at GBs is characterized, in terms of the major alloying elements, by the depletion of Cr and the enrichment of Ni [6]. The depletion of Cr may cause GBs to be susceptible to corrosion and induce irradiation-assisted stress corrosion cracking (IASCC) [5]. Limited studies exist that relate how the structure of the GB alters segregation profiles in these steels. In order to develop materials that are able to withstand more extreme environments, it is crucial to understand what type of GB character promotes point defect recombination and effectively minimizes Cr segregation. Although previous research has examined trends in segregation with respect to misorientation, little information is provided with respect to the GB plane. An early high-voltage electron irradiation study by Watanabe and Takamatsu [12] studied the segregation effect at low sigma (Σ) coincidence site lattice (CSL) boundaries, where the CSL Σ notation describes the geometric relationship between the lattice sites in adjacent grains and describes the degree of coincidence of lattice sites at a GB [13]. These studies found that $\Sigma3$, $\Sigma9$ and low-tilt-angle GBs experienced a reduced amount of Cr depletion while more random, high-angle tilt GBs did not suppress RIS. In additional studies, researchers [14,15] found a local segregation minima at the $\Sigma3$ GB while noting a general trend of lower Cr segregation with decreasing CSL Σ value. Limited experimental results indicate or predict why particular types of GB structures are able to suppress or limit Cr segregation. A more recent study by Sakaguchi et al. [16], however, provides insight into the role of the full GB character by examining GBs with the same misorientation, $\Sigma3$, but different GB planes. To adequately describe GB character, five macroscopic degrees of freedom must be taken into account: three for the boundary misorientation between adjacent grains and two for the boundary plane [17]. GB character dependence of phenomena other than RIS has shown that trends are present based on the GB plane in addition to misorientation. Examination of the denuded zone of voids in copper under helium irradiations by Han et al. [18] found significant correlations between the width of the denuded zone and the full GB character and GB sink efficiency [18]. Atomistic modeling studies [19–22] have recently indicated the importance of the atomic GB structure on the interaction of interstitial and vacancy point defects which varies with the GB plane. These findings indicate very specific differences in interstitial and vacancy GB segregation energies and interaction strength between the various types of GB structures for model body-centered cubic and face-centered cubic (fcc) crystalline systems. The results reveal a difference between tilt and twist GBs in addition to boundary plane symmetry dependency. Unfortunately, limited research has been executed to compare or explain these atomistic simulations with experimental GB-dependent phenomena such as RIS or denuded zone width.

In this study, an expanded insight into how the GB character interacts with irradiation-induced point defects and subsequent solute segregation (RIS) in 316L austenitic stainless steel under heavy ion irradiation is presented. Well-developed thermomechanical processing techniques for austenitic stainless steel [23], or “GB engineering” [24], are used to obtain microstructures with specific variations in the GB character distribution which includes variations in misorientation and boundary plane. This permits the examination of the full GB character (i.e., five macroscopic degrees of freedom) dependence of Cr and Ni segregation after irradiation. The experimental results of RIS dependency on GB character are compared with simulated vacancy–GB atomistic metrics such as vacancy formation energy. This provides a method for a more detailed understanding, combining modeling and experiments, of the interaction of point defects in RIS with variations in the atomic GB character or structure.

2. Experimental design and simulation method

2.1. Experimental design

AISI 316L austenitic stainless steel was obtained in the mill-annealed state from Carpenter Technology (Reading, PA) with a chemical composition given in Table 1. Specimens from the as-received 316L sample were cut to a nominal thickness of 10 mm and homogenized for 30 min at 1050 °C in an air furnace, followed by a water quench. A homogenization heat treatment was then used to remove any unforeseeable solute segregation or precipitation during alloy processing, or delta ferrite banding that would otherwise alter the GB RIS profile [25]. After the homogenization, the 316L was thermomechanically processed by a 5% rolling reduction and subsequent 1000 °C anneal, followed by a water quench. The thermomechanical processing was used to induce a high number fraction of twin GBs, both coherent and incoherent, and twin related GBs. Jet-thinned disks of the processed 316L alloy were prepared by standard metallographic techniques and electropolishing with a TenuPol 5 polishing unit using 5 wt.% perchloric acid–95 wt.% methanol solution at -35 to -45 °C with a voltage of $\sim$20 V and varying current.

Heavy ion irradiations were carried out at the Ion Beam Laboratory at Sandia National Laboratories using a Tandem linear accelerator. The 316L specimens were irradiated using broad beam irradiation (3 mm × 3 mm spot size) conditions with 3 MeV Cu^{2+} ions at 500 ± 10 °C to a fluence of $\sim 2.3 \times 10^{16}$ ions cm^{-2}. The estimated displacement per atom (dpa) for the irradiation was $\sim$11 dpa at a dose rate of $\sim 1.0 \times 10^{-3}$ dpa s^{-1}. The dpa was estimated from stopping and range of ions in matter (SRIM) recoil/damage calculations made with Kinchin–Pease estimates [26] for a thickness of 110 nm. This thickness corresponds to the electron transparent region of interest in the transmission electron microscopy (TEM) foils. At all regions of interest