Acta Materialia 125 (2017) 231–237

Contents lists available at ScienceDirect

Acta Materialia

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

Full length article

Suppression of vacancy cluster growth in concentrated solid solution alloys

Shijun Zhao [a, *], Gihan Velisa [a], Haizhou Xue [b], Hongbin Bei [a], William J. Weber [b, a], Yanwen Zhang [a, b, **]

[a] Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA
[b] Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN 37996, USA

ARTICLE INFO

Article history:
Received 9 August 2016
Received in revised form
26 October 2016
Accepted 20 November 2016

Keywords:
Molecular dynamics simulations
Ion irradiation
Concentrated solid-solution alloys
Ni-based alloys
Vacancy cluster
Stacking fault tetrahedron

ABSTRACT

Large vacancy clusters, such as stacking-fault tetrahedra, are detrimental vacancy-type defects in ion-irradiated structural alloys. Suppression of vacancy cluster formation and growth is highly desirable to improve the irradiation tolerance of these materials. In this work, we demonstrate that vacancy cluster growth can be inhibited in concentrated solid solution alloys by modifying cluster migration pathways and diffusion kinetics. The alloying effects of Fe and Cr on the migration of vacancy clusters in Ni concentrated alloys are investigated by molecular dynamics simulations and ion irradiation experiment. While the diffusion coefficients of small vacancy clusters in Ni-based binary and ternary solid solution alloys are higher than in pure Ni, they become lower for large clusters. This observation suggests that large clusters can easily migrate and grow to very large sizes in pure Ni. In contrast, cluster growth is suppressed in solid solution alloys owing to the limited mobility of large vacancy clusters. The differences in cluster sizes and mobilities in Ni and in solid solution alloys are consistent with the results from ion irradiation experiments.

1. Introduction

Structural materials suffer from damage production under irradiation. In particular, the formation of large vacancy clusters (VCs) greatly degrades the performance of materials, as VCs may lead to hardening and void swelling, as well as creep and plastic instabilities [1–3]. Therefore, effective modulation of VC migration and suppression of large VC growth are two of the main design criteria for the development of advanced structural materials that are of great importance for future applications in nuclear energy and high-energy accelerators.

Several mechanisms of VC formation and growth have been proposed. As stacking-fault tetrahedra (SFT) are the dominant VC types in various irradiated face-centered cubic (*fcc*) materials, especially those with low stacking fault energies [2,4,5], most studies have focused on these specific VCs. It has been argued that SFT can be created by the gliding of dislocation loops that are formed as a result of vacancy accumulation [6,7]. SFT can also be formed directly in collision cascades [8], indirectly from the collapse of VCs [9] or by the aggregation of vacancies [10]. Perfect SFT are considered to be immobile once they are created because their glide planes are not the close-packed [111] planes in *fcc* structures. As a result, the removal of SFT is very difficult because of their high stabilities. Possible methods of suppressing SFT formation include annealing at very high temperature [11] and the incorporation of interstitials [12] or mobile dislocations [13,14] that are induced by ion irradiation. However, these methods could also cause other undesirable modifications.

In the pursuit of irradiation-tolerant structural materials, Ni-based single-phase concentrated solid solution alloys (CSAs) are considered to be promising candidates owing to their extraordinary mechanical properties and irradiation resistance [15,16]. In contrast to traditional dilute alloys, CSAs consist of multiple principal elements that significantly influence the energy dissipation pathway and defect dynamics during radiation [17]. Experimentally, larger SFT are observed in pure Ni, whereas smaller SFT are found in CSAs, such as $Ni_{0.5}Fe_{0.5}$ [15]. Although the growth of SFT is greatly suppressed in CSAs, the mechanism for the suppression is not clear. Recently, molecular dynamics (MD) simulations were used to gain

* Corresponding author.
** Corresponding author. Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA.
E-mail addresses: zhaos@ornl.gov (S. Zhao), zhangy1@ornl.gov (Y. Zhang).

http://dx.doi.org/10.1016/j.actamat.2016.11.050
1359-6454/

understanding of SFT diffusion in Cu. The results revealed that defective SFT, which are not composed of a magic number of vacancies (3, 6, 10, 15, 21, 28 …), have large mobilities several orders of magnitude higher than those of perfect SFT with a magic number of vacancies [18]. The successful MD work in Cu suggests that it may be feasible to study VC diffusion and growth in CSAs using MD within an affordable simulation time.

In this work, an integrated effort based on MD, ion irradiation, and ion beam analysis is carried out to understand the diffusion of VCs in some model CSAs and to demonstrate concentrated solid solutions as an effective way to suppress the growth of VCs. In pure Ni and its solid solutions with Fe and Cr, the diffusion coefficient of VCs is calculated, and the irradiation-induced damage depth is measured. It is found that the calculated diffusion properties are consistent with experimental observations. The simulation results are further validated by comparing them with previously reported cluster sizes in irradiated CSAs [19]. Our results show that the diffusion and growth of VCs in Ni can be modulated by forming solid solutions of Ni with Fe and Cr. In particular, our MD results predict that the growth of SFT can be suppressed significantly in $Ni_{0.4}Fe_{0.4}Cr_{0.2}$, suggesting that $Ni_{0.4}Fe_{0.4}Cr_{0.2}$ exhibits the most efficient alloying environment for restricting SFT growth.

2. Methods

MD simulations were performed using the code Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [20]. The interatomic forces were modeled using the embedded atom method (EAM) based on the Bonny 2013 interatomic potential [21]. This potential was developed for the modeling of irradiation effects in ternary Ni-Fe-Cr systems, and the predicted defect properties are in good agreement with *ab initio* calculations. To cross-check the results, another potential for pure Ni developed by Mishin et al. was also used [22]. The simulation cell was oriented along the [112], [$\bar{1}$10] and [$\bar{1}\bar{1}$1] directions with the dimensions of around 4.4, 5.1, and 5.0 nm, respectively. All simulations were carried out in the NPT ensemble at zero pressure. Temperatures ranging from 800 to 1200 K were employed. The total linear and angular momentum were both set to zero at each time step. Periodic boundary conditions were imposed in all directions, and a time step of 2 fs was used to ensure the energy conservation.

VCs were created by removing a triangular region of atoms in the [$\bar{1}\bar{1}$1] plane located at the center of the simulation cell. After a short dynamics run and following structural relaxation, stable VCs were formed. A perfect stacking fault tetrahedron was created when the vacancy number was equal to a magic number; otherwise a defective tetrahedron was produced. The software OVITO was used for visualization, and a centro-symmmetry analysis was employed to identify SFT configurations [23]. To locate the defect positions, a Wigner-Seitz defect analysis was also performed as implemented in OVITO.

The diffusion coefficient was calculated at different temperatures using the Einstein formula:

$$D = \frac{R^2}{2dt}.$$

Here, R^2 is the square displacement (SD), d is the dimensionality of the system, and t is simulation time. The mean square displacement (MSD) of all atoms was used to estimate the SD and to calculate the self-diffusion coefficient D^* [24,25]. For comparison, the diffusion coefficient was normalized with respect to the vacancy number.

Smaller VCs in some CSAs, as compared with those in pure Ni, were previously reported from both experiment [26,27] and MD simulation [26]. To validate the limited migration of VCs in CSAs predicted from MD simulations, Ni, $Ni_{0.8}Fe_{0.2}$ and $Ni_{0.8}Cr_{0.2}$ were irradiated at room temperature with 1.5 MeV Mn ions at 7° off the <001> direction to avoid channeling effects. Cascade damage accumulation at irradiation fluences of up to 2×10^{14} cm^{-2} and 1×10^{16} cm^{-2} was studied; and the resulting disorder profiles, mainly interstitial-type loops and vacancy-type SFT, were determined from Rutherford backscattering spectrometry (RBS) along the <001> direction [28]. The interstitial loops and SFT cause local strains and lattice distortions that lead to an increase in the dechanneling yield, in addition to the direct backscattering yield from imperfections visible to the channeled probing ions. The ion ranges of Mn and corresponding damage profiles induced by direct cascade damage in these materials are comparable, owing to the similar densities of Ni and its alloys [29]. Therefore, differences in measured disorder profiles under the same irradiation condition should be attributed to irradiation-induced defect migration and can reflect the MD-predicted modulation of defect migration resulting from the alloying effects of Fe and Cr in Ni.

3. Results and discussion

3.1. Diffusion of VCs

The evolution of MSD calculated from VCs with different

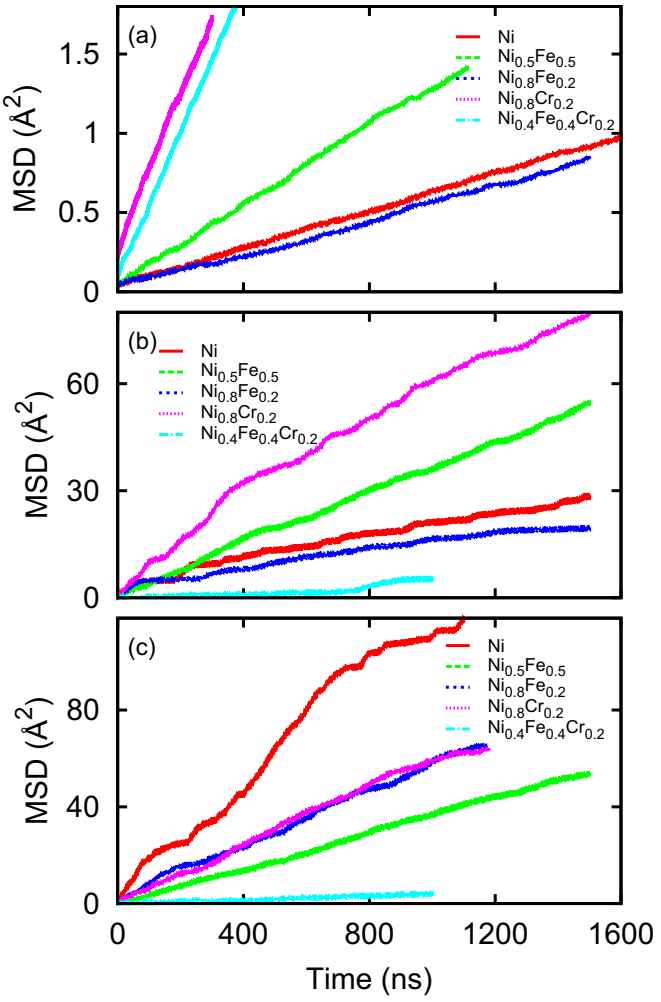

Fig. 1. MSD for vacancy clusters with different size N (1 (a), 9 (b) and 13 (c)) in Ni, $Ni_{0.5}Fe_{0.5}$, $Ni_{0.8}Fe_{0.2}$, $Ni_{0.8}Cr_{0.2}$ and $Ni_{0.4}Fe_{0.4}Cr_{0.2}$ at 1000 K.