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Thermodynamic mixing energy and heterogeneous diffusion uncover the mechanisms of radiation damage reduction in single-phase Ni-Fe alloys

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ABSTRACT

Understanding and predicting radiation damage is of central importance to develop radiation-tolerant structural materials for current and next-generation nuclear systems. Single-phase solid solution alloys constitute attractive choices due to their promising mechanical properties and radiation tolerance. Here, by examining radiation-induced defect production and evolution in single-phase Ni-Fe alloys, we show that radiation damage resistance directly correlates with thermodynamic mixing energy and heterogeneity of defect diffusion. We found that radiation damage in materials decreases linearly with lowering mixing energy, and the relationship holds true for all studied Ni-Fe compositions. The damage reduction with varying composition is further ascribed to the increasing heterogeneity of point defect migration across a complex potential energy landscape that enhances defect recombination. This new insight into the dynamical evolution of radiation defects points to a thermodynamic criterion for designing radiation-tolerant materials.

1. Introduction

The main technical impediment to the development of advanced nuclear power systems lies in the creation of radiation-resistant structural materials. Various approaches are under active research, ranging from oxide dispersion strengthened (ODS) steels [1], to nanograined alloys [2], to severely plastically deformed (SPD) materials [3]. Recently, a new class of materials, called single-phase concentrated solid solution alloys (SP-CSAs) [4], including high entropy alloys (HEAs) [5], has received great attention due to their potential radiation resistance [6]. SP-CSAs are typically composed of two or more principle elements in a simple, single-phase solid solution, with nearly equiatomic concentrations. The random arrangement of atoms results in a complex local atomic environment [7], with implications ranging from high strength [8] while retaining ductility [9], to high fracture toughness [10], to excellent corrosion resistance [11]. Moreover, recent experiments and simulations indicate that these single-phase alloys have significantly better radiation resistance than their corresponding elementary metals [6,12].

Varying alloy elemental concentrations and types changes the local atomic environment, which can significantly influence the migration energy barriers and pathways of radiation defects. This has been demonstrated by tuning the composition of alloys to increase their radiation damage tolerance. Wang et al. found that radiation-induced defect sizes resulting from ion irradiation of Ni-Cu alloys decrease when increasing the Cu concentration from 10% to 50% [13]. Jin et al. and Zhang et al. observed enhanced radiation tolerance in Ni-Fe alloys by increasing the Fe concentration up to 60% [14,15], and suggested chemical disorder to be a key factor for controlling radiation performance [14]. Molecular dynamics (MD) simulations of radiation damage accumulation have been studied intensively from elementary metals to many SP-CSAs [6,12,16], revealing that more alloying (both in species and concentrations) demonstrates higher radiation resistance. However, these investigations focus on the reduction of damage in a few selected alloys in comparison to elemental Ni. A comprehensive understanding of the effects of alloy composition, defect dynamics, damage accumulation, and defect recombination are necessary to gain a more fundamental basis for designing radiation tolerant alloys.

In this work, we study ion radiation-induced defect evolution and damage accumulation in elemental Ni and $Ni_{1-x}Fe_x$ alloys with x ranging from 10 to 90%. The face-centered cubic (FCC) structure is

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considered so as to study the pure effect of the single variable of Fe concentration x on radiation resistance, without introducing any additional complexity of crystal structure. However, it should be noted that when x is larger than $\sim$ 70%, the FCC system is in a thermodynamically metastable state. We employ a hybrid Monte Carlo and Molecular Dynamics (MC+MD) approach (see the Methods section) to prepare well-mixed systems. MD is then utilized to impose consecutive radiation collision cascades to achieve radiation doses of about 0.5 DPA (displacements per atom) (See Methods). By analyzing defect production and evolution, we find that defect accumulation clearly shows a dependence on alloy composition, and the rate of damage accumulation is controlled by the speed of large defect cluster growth. The largest reduction in damage is observed near the minimum of mixing energy (see Fig. 2(c)), which we suggest as a parameter in governing radiation resistance. To obtain a more fundamental understanding of the results, we examined defect migration energy barriers and pathways of point defects in these alloys using the nudged elastic band (NEB) method [17]. The variance of these barriers, which reflects the complexity of the local potential energy landscape (PEL), indicates increasingly heterogeneous point defect migration in the more highly radiation-tolerant compositions. Our results provide atomistic details of damage evolution and reduction, suggesting a quantitative basis for developing radiation-tolerant materials.

2. Methods

2.1. Hybrid MC+MD simulations and atom swapping

One possible concern in atomistically simulating SP-CSAs involves the creation of the initial atomic structure. The approach used in previous MD studies [12,18,19] was to randomly replace one atom type with the other, which may not recreate truly mixed SP-CSA structures. This random procedure can produce local atomic structures with high potential energy, which are thermodynamically unstable. The as-prepared metastable system becomes a particular concern when simulating radiation damage, as the dissipation of radiation damage energy and structural relaxation of the initial high-energy atomic configuration would become convolved. Recently, an effort to construct SP-CSAs via a quasirandom structure was made in Ref. [20], where the Warren-Cowley short range order [21] was optimized to produce a truly random atomic structure. In this work, we use a hybrid MC+MD algorithm to prepare stable alloy systems. $Ni_{1-x}Fe_x$ alloys are modeled using the embedded atom method (EAM) potential developed by Bonny et al. [22]. This potential is an improved version of the Bonny 2011 potential [23], which was developed to model the production and evolution of collision cascade-induced defects. It has been extensively applied to successfully predict concentration-dependent radiation defects in NiFe alloys as observed in experiments [6,15]. We prepare the initial FCC alloy structures by randomly mixing Ni and Fe atoms at each specified composition. The simulation cell contains 108,000 atoms, and periodic boundary conditions are applied in all three directions in all simulations. A hybrid MC+MD algorithm [24,25] is used to anneal each generated system. In each MC trial step, a randomly selected Ni atom is swapped with another randomly selected Fe atom. The trial move is accepted with a probability of unity or $P = exp(-\Delta U/k_B T)$, whichever is smaller, depending on the value of ΔU, where ΔU is the change in system potential energy after the atom exchange and T is the chosen system temperature of 300 K. Following 100 MC trials, we perform 0.1 ps of MD simulation in the isothermal-isobaric condition (NPT ensemble). We repeat the above MC+MD procedures to anneal each alloy, and a total of 50,000 MC trials and 50 ps MD runs are performed. The equilibrated systems show much lower potential energies than those which were randomly generated (see Supplementary Fig. S1).

2.2. Molecular dynamics simulations

The MD method as implemented in LAMMPS [26] is utilized to simulate self-ion radiation damage in each annealed system. To deal with high energy radiation damage collisions, a Ziegler-Biersack-Littmark (ZBL) repulsive potential [27,28] is smoothly joined to the aforementioned EAM potentials [29], and an adaptive time step algorithm is used to limit atomic movement to 0.05 Å in each timestep. The choice of ZBL joining parameters could affect the generation of primary damage as discussed by Stoller et al. [30]. In this work, the smoothly joined potentials adequately capture the features of Fe concentration-dependent radiation defect production and evolution as seen in ion irradiation experiments. We first relax each system at 300 K for 100 ps. An atom is randomly chosen as the primary knock-on atom (PKA), and we shift the entire cell to move the PKA to the center. This avoids the damage cascade from reaching the system boundaries. The PKA is assigned a kinetic energy of 5 keV, and 50,000 adaptive timesteps of MD simulation ($\sim$45 ps) are performed. The Nosé-Hoover temperature-rescaling thermostat [31,32] is applied to the atoms at the sides of the simulation cell with a width of half the lattice constant, to absorb the collision cascade energy and cool the system to 300 K. This process is repeated with each PKA randomly chosen and shifted to the center of the cell, simulating the spatially random arrival of radiation damage cascades. Up to 1500 consecutive 5 keV cascades result in radiation damage levels of $\sim$0.5 DPA. The short simulation time (an inherent limitation of MD) between collision cascades yields a dose rate several orders of magnitude higher than that in experimental conditions. Although the difference in dose rate between MD and experiments is quite large, a recent study using similar approaches has shown good agreement with experiments [6]. We performed twelve independent 1500 consecutive 5 keV cascade simulation runs for each system to obtain more reliable results. Defects are identified by comparing the irradiated system with the initial perfect structure using Wigner-Seitz cell method. The defects analysis and visualization are performed with OVITO package [33] with an adaptive common neighbor analysis.

2.3. Defect migration energy barrier calculations

We compute vacancy and interstitial migration energy barriers with the climbing image nudged elastic band (NEB) method [17]. We start with the MC+MD equilibrated system. By removing one atom, we create a vacancy and consider all twelve (not necessarily equivalent) diffusion pathways on the FCC lattice by exchanging with the twelve first nearest neighbors. With the NEB method, we find the saddle point along each minimum energy pathway, and from that the migration energy barrier is calculated (see Supplemental Information). We consider all possible vacancy sites in the relaxed system, and the energy barriers corresponding to $\sim$10,368 pathways are evaluated. Similarly, we insert Ni or Fe atoms to create [100] dumbbell interstitials. Note that due to local lattice distortion, placing an atom at the designated position may not create the desired dumbbell structure after relaxation. Therefore we take extreme care to confirm the initial and final configurations for interstitial migration. Only valid rotations are considered in our calculations.

3. Results

3.1. Annealed alloy systems and their mixing energies

We take extra care to prepare a stable, well-equilibrated single-phase alloy using the hybrid MC+MD method. We found that