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Point defect evolution in Ni, NiFe and NiCr alloys from atomistic simulations and irradiation experiments ☆

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ABSTRACT

Using molecular dynamics simulations, we elucidate irradiation-induced point defect evolution in fcc pure Ni, $Ni_{0.5}Fe_{0.5}$, and $Ni_{0.8}Cr_{0.2}$ solid solution alloys. We find that irradiation-induced interstitials form dislocation loops that are of $1/3\langle 1\,1\,1\rangle\{1\,1\,1\}$-type, consistent with our experimental results. While the loops are formed in all the three materials, the kinetics of formation is considerably slower in NiFe and NiCr than in pure Ni, indicating that defect migration barriers and extended defect formation energies could be higher in the alloys than pure Ni. As a result, while larger size clusters are formed in pure Ni, smaller and more clusters are observed in the alloys. Vacancy diffusion occurs at relatively higher temperatures than interstitials, and their clustering leads to the formation of stacking fault tetrahedra, consistent with our experiments. The results also show that the surviving Frenkel pairs are composition dependent and are largely Ni dominated.

1. Introduction

In contrast to conventional alloys that contain one principal element with minor alloying elements, single-phase concentrated solid-solution alloys (SP-CSAs), including high entropy alloys (HEAs), contain multiple principal elements present in almost equal proportions. Thermodynamically such compositions could be readily rendered uninteresting due to intuitive phase separation; however, their high mixing entropies facilitate formation of random solid solutions with rather simple crystal structures. Interestingly, these alloys also possess exceptional mechanical properties such as high strength-weight ratio, wear resistance, high-temperature strength and structural stability, fracture resistance, etc. that are significantly improved over conventional alloys [1–5]. In the addition, these alloys have attracted interest in physical properties such as electrical, magnetic and thermal, thus opening a new research field in metallic materials [6].

These alloys could be exciting new candidate materials for nuclear power applications, particularly if the enhanced mechanical properties could be coupled with high radiation tolerance for their use as structural materials in nuclear reactors. The inherent disorder (or random arrangement of different elements) could actually help in mitigating damage by annihilation of Frenkel pairs at very early stages of cascade events. With this proposition in view, Ni-based fcc SP-CSAs are the key alloys of interest where understanding radiation response of different compositions in order to design inherently radiation tolerant materials is currently the main focus [7,8].

These SP-CSAs are however new, and even the most fundamental radiation damage studies, such as point-defect evolution processes are lacking. Therefore, given the alloy complexity and the lack of literature, here we focus on simpler, binary NiFe and NiCr random alloys. In this work, using molecular dynamics (MD) simulations, we elucidate clustering mechanisms and kinetics of point defects in NiFe and NiCr, and compare them to that in pure Ni. We show that radiation induced point defects lead to formation of interstitial dislocation loops and stacking fault tetrahedra (SFT) in both pure Ni and the two alloys in good agreement with our

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experimental observations. We find that point defect diffusion is significantly slower in the alloys than in pure Ni. We also show that the sizes of defect clusters are significantly different between pure Ni and the alloys.

2. Methodology

The interaction of electrons, ions and neutrons with materials results in radiation damage primarily due to the displacement of atoms by elastic collisions, which can results in widely separated singular displacement events (e.g., electrons, protons, slow neutrons) and highly localized collision cascades (e.g., heavy ions and fast neutrons). Both processes result in the accumulation, diffusion and interaction of radiation-induced defects. These atomic level radiation damage production processes are conventionally modeled by giving a primary knock-on atom (PKA) kinetic energy that is dissipated in single or multiple elastic collision events, depending on energy, with higher PKA energies leading to a cascade of elastic scattering collisions. The evolution of such radiation-induced defects is the result of the accumulation and diffusion of the defects over time. This process can be broken down primarily into two stages, i.e., ballistic (e.g., collision) stage and kinetic (e.g., relaxation) stage. During the ballistic stage, a large number of atoms can be displaced in collision cascades, as shown by the peak in Fig. 1 by a heavy ion, or isolated Frenkel pairs are created in the case of light particles (e.g., electrons or protons). Overtime, many of these radiation-induced defects, whether produced in isolation or in cascades, recombine and annihilate during the kinetic stage leaving only a few surviving defects behind.

Ideally both stages can be computationally captured via MD simulations, however, to accurately capture atomic forces during the ballistic stage when the atoms come very close to each other, the iterative time step needs to be decreased significantly. As a result, the ballistic stage alone is computationally expensive, and most of the simulations cannot reach the longer time associated with defect diffusion and the kinetic processes that lead to addition defect clustering and evolution. This expense is exceedingly noticeable while modeling multi-component systems. Hence, traditionally MD simulations have largely remained limited to the ballistic stage, and the clustering and diffusion events occurring from surviving isolated defects or small clusters over longer time scales have commonly been captured using additional methods such as kinetic Monte Carlo (KMC) or temperature accelerated dynamics (TAD) [10,11]. Although these methods can model up to experimental time scales, they remain fundamentally limited by the diffusion events that are defined by input parameters, thus making these methods highly probabilistic in nature.

Recently, another approach, a defect-driven method, has been tested that bypasses the ballistic stage, and focuses only on the kinetic stage [12–14]. Under this method, the focus is largely on capturing the evolution of the single point defects that are created by low-energy ballistic events (e.g., electrons and protons) or those that survive single or multiple cascade events. In this approach, instead of creating Frenkel pairs via time-consuming ballistic recoil events, they are randomly distributed at the start of the simulation. The disadvantage is that the processes inherent to more energetic ballistic events, such as the effects of shock waves, high displacement densities, and thermal spike on defect production, are not captured. Also not captured are the dependences of phase transformations, such as amorphization in ordered intermetallic compounds [15] on Frenkel pair concentrations or dose. However, such approaches that add Frenkel pairs with time are often limited to smaller simulation cell sizes and are unable to capture the slow evolution of microstructure when Frenkel pairs are added one-by-one with time. In the current approach, microstructure evolution is accelerated or driven by the larger Frenkel pair concentration, and the results from this approach have been fruitful in reproducing experimentally observed clusters and in revealing the underlying clustering mechanisms [16]. Similarly, radiation-induced grain growth has been captured in CeO_2 while elucidating the fast kinetics of grain growth observed experimentally [17,18]. Such interstitial clustering and irradiation induced grain growth had not been captured previously from the conventional cascade methods. This method has also captured defect clusters in other ceramic oxides, such as MgO, UO_2 and ZrO_2, and has replicated defect evolution observed from the conventional approach, thus validating the idea to bypass the ballistic stage for defect-driven processes [12,13,19]. Additionally, this approach is also devoid of the interatomic potential artifacts that may originate from the by Ziegler, Biersack, and Littmark (ZBL) scheme used to model forces of atoms that may come very close to each other during the ballistic stage [20]. It should be noted that this approach is not a surrogate to the traditional energetic cascade approach; it models the interaction and evolution of surviving defects from collision cascades, as well as the isolated defects that accumulate under irradiation with electrons and protons.

In this work, we apply this approach to metallic materials, and show that the nature of defect clusters reminiscent of fcc metals can be also be captured via this approach. We mainly model pure Ni, $Ni_{0.5}Fe_{0.5}$ and $Ni_{0.80}Cr_{0.20}$ materials using a $20 \times 20 \times 20$ fcc unit cell system consisting of 32,000 atoms. In alloys, the atoms are randomly placed to replicate the random nature of SP-CSAs. In most of the simulations, 1 at.% Frenkel pairs (i.e., 320 interstitials and vacancies) are randomly created, and the simulations are run for 5 ns at 300 K, unless mentioned otherwise. This concentration of defects is only slightly lower (factor of four) than that produced in MD simulations by the overlapping of 5 keV cascades in fcc $Ni_{0.5}Fe_{0.5}$ [21]. In this work, the interstitials are created by placing lattice atoms at octahedral site leaving behind vacant lattice sites. Thus, equal number of interstitials and vacancies are always created. We use the embedded atom method (EAM) based Bonny

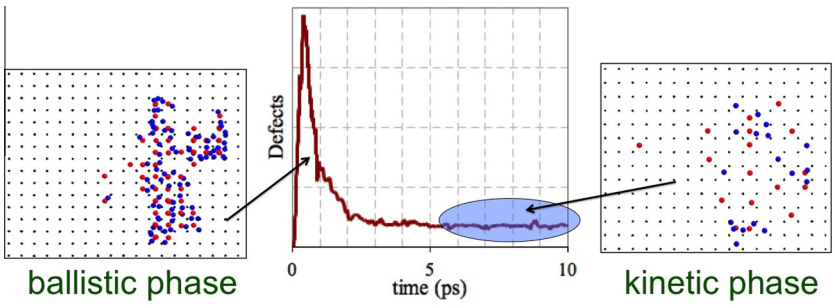

Fig. 1. Schematic showing generation of point defects forming a peak during the ballistic phase followed by vacancy-interstitial recombination during the kinetic phase leaving behind few surviving defects.