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The role of nickel in radiation damage of ferritic alloys

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Abstract—According to modern theory, damage evolution under neutron irradiation depends on the fraction of self-interstitial atoms (SIAs) produced in the form of one-dimensional glissile clusters. These clusters, having a low interaction cross-section with other defects, are absorbed mainly by grain boundaries and dislocations, creating the so-called production bias. It is known empirically that the addition of certain alloying elements influences many radiation effects, including swelling; however, the mechanisms are unknown in many cases. In this paper we report the results of an extensive multi-technique atomistic level modeling study of SIA clusters mobility in body-centered cubic Fe–Ni alloys. We have found that Ni interacts strongly with the periphery of clusters, affecting their mobility. The total effect is defined by the number of Ni atoms interacting with the cluster at the same time and can be significant, even in low-Ni alloys. Thus a 1 nm (37SIAs) cluster is practically immobile at $T < 500$ K in the Fe–0.8 at.% Ni alloy. Increasing cluster size and Ni content enhances cluster immobilization. This effect should have quite broad consequences in void swelling, matrix damage accumulation and radiation induced hardening and the results obtained help to better understand and predict the effects of radiation in Fe–Ni ferritic alloys.
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Keywords: Dislocation loops; Fe–Ni alloys; Diffusion mechanism; Radiation effects

1. Introduction

Growing energy needs demand significant improvements in the efficiency of energy production, including the usage of nuclear power plants [1]. Predicting the behavior of structural materials under operational irradiation conditions is crucial for safe, long-term operation [2]. However, extending the lifetime of nuclear plants makes direct experimental predictions impossible for the experiments must last decades to correctly reproduce the corresponding radiation effects. Therefore, theoretical and computational approaches with predictive capabilities have become commercially important applications. Any practically useful prediction that exceeds available experimental data should be (a) based on the best available theory and (b) realized as a computational approach through the models of necessary scales. The modern approach to generalized radiation damage theory, described in Ref. [3], is able to explain qualitatively and, in many cases, quantitatively, radiation-induced phenomena such as the effects of recoil energy spectra and grain boundaries on swelling; void ordering and void lattice formation; and radiation-induced segregation and radiation growth in anisotropic materials [3–10]. At present this theory is the most promising candidate to provide the basis for predictive modeling of materials behavior under irradiation. According to Ref. [3], in the commercially important case of cascade-type damage (neutron, ions) and for a material with a low dislocation density, ρ_d, the steady state rate of swelling S is independent on the irradiation dose ϕ and can be expressed as:

$$\frac{dS}{d\phi} \approx (1-\varepsilon_r)\varepsilon_i^g k_c^2/(k_c^2 + Z_V^d \rho_d) \approx (1-\varepsilon_r)\varepsilon_i^g \qquad (1)$$

where $(1-\varepsilon_r)$ is the fraction of NRT-estimated defects (for NRT standard, see Ref. [11]) surviving after cascades, ε_i^g is the fraction of self-interstitial atoms (SIAs) created in the form of one-dimensional (1-D) glissile clusters, k_c^2 is the total sink strength in the system and $Z_V^d \approx 1$ is the dislocation efficiency for absorbing vacancies. The swelling rate is proportional to ε_i^g because 1-D glissile clusters have a very low interaction cross-section with other defects. As a result they do not recombine with vacancies in the bulk but have a long mean free path (of the order of μm) and usually are absorbed by grain boundaries or dislocations, which increases the vacancy supersaturation. This is called "production bias" and it appears exclusively due to (a) the direct formation of SIA clusters under cascade damage conditions and (b) the ability of interstitial clusters to glide one-dimensionally.

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Production bias together with the conventional "dislocation bias" defines radiation swelling and many other radiation effects. Eq. (1) looks quite simple but describes the most important consequence of cascade-type irradiation and has successfully explained the recoil energy effect on swelling [4] from the contribution of the production bias. Eq. (1) presents the limit case when SIA clusters glide quickly, i.e. the known case of perfect pure metals. In the more general case, i.e. when the cluster's glide speed can be affected by the local environment, the production bias contribution depends on the cluster mean free path that, in its turn, is defined by the cluster diffusivity. It is therefore obvious that by affecting the formation of glissile clusters and/or their 1-D motion one can control a significant contribution to the radiation swelling and other related effects. Note that the addition of certain alloying elements has been successfully used over years to control many radiation effects, including swelling, in a purely empirical way.

In this paper we report our results from a study of the effect of nickel impurity in iron on 1-D glide of interstitial clusters/dislocation loops. There are several reasons for the choice of this system. First, ferritic alloys are widely used structural material in nuclear energy. Second, Ni is known for its ability to stabilize gamma-Fe phase in stainless steels. However, the knowledge of its effect on radiation resistance in ferritic steels is rather limited: there is some evidence that Ni affects Cu-precipitate evolution [12], dislocation loop populations [13], mechanical properties [14] and, possibly, swelling [15]. Third, the set of interatomic potentials available, Fe–Fe [16], Fe–Ni [17] and Ni–Ni [18], provides a very realistic description of Ni in ferritic alloys due to a thorough fitting to a number of *ab initio* properties in the Fe–Ni system. Moreover, a recent multi-technique *ab initio* study of Ni–impurity interactions with screw and edge dislocations in Fe [19][1] has confirmed the high accuracy of the Fe–Ni potential from Ref. [17]. This is because Ni in Fe is in a paramagnetic state and the magnetic effects, which complicate the description of Cr or Mn impurity in Fe, are negligible for Ni.

In this work we carried out an extensive study of interstitial cluster/dislocation loop mobility in Fe–Ni alloys using different atomic level techniques to discover the strong effect of Ni on SIA cluster trapping and reducing their mobility, and used the results to predict qualitatively some properties of irradiated Fe–Ni ferritic alloys. The structure of the paper is as follows. In Section 2 we describe briefly the atomistic techniques used. The results describing Ni effects on interstitial cluster mobility are presented in Section 3 and the predicted effects under irradiation are summarized and discussed in Section 4.

2. Modeling approaches

We have used molecular dynamics (MD) and molecular statics (MS) techniques. Conventional MD was used to model thermally activated motion, e.g. diffusion of interstitial clusters in body-centered cubic (bcc)-Fe with different Ni content, from 0.8 to 2.5 at.% over a wide temperature range from 300 to 900 K. A detailed description of our diffusion modeling approach for clusters performing 1-D motion can be found in previous work on pure Fe [20,21] and Fe–Cr alloys [22]. The system size for the MD study varied from ~15,000 to ~45,000 mobile atoms depending on the cluster size (NSIA), which varied in the range of 7–61 SIA. We have modeled diffusion up to a few hundred nanoseconds to accumulate statistically significant number of cluster jumps and long enough trajectories. The MD results were used to determine the diffusion coefficients by different methods as described in Ref. [23] and applied in Refs. [20,21]. According to Ref. [23], the cluster diffusion coefficients can be obtained by treating cluster jumps and their correlations or decomposing their trajectories according to jumps (trajectory jump decomposition (TJD)) or time segments (trajectory time decomposition (TTD)). If the simulation of each trajectory is long enough, all diffusion coefficients are similar, as demonstrated in Refs. [20–23]. Ni atoms introduce new correlations in the motion of SIA clusters. This makes treatment of one-dimensional cluster motion more complicated for the segments in the trajectory decomposition should be longer to include these correlations. For example, TTD treatment for 19SIA cluster at 500 K converges with segments above ~30 ps [21]. Treatment of the same cluster trajectory in Fe–0.8 at.% Ni alloys demands segments of ~280–300 ps that is 10 times longer. Therefore to achieve the same accuracy of the cluster diffusion coefficient in an alloy one needs to increase the modeling time by the corresponding factor. The effective migration energy, E_m^*, can then be estimated from the Arrhenius-type plot. We also have analyzed particular mechanisms using visualization methods.

MS simulations were carried out in systems from ~230,000 to ~627,000 atoms. MS was used to calculate the cluster–Ni interaction energy by relaxing particular configurations. However, particular impurity distributions can be quite different and lead to quite different total interaction energies and migration barriers, especially in concentrated alloys. We therefore used two MS-based approaches. In the one, the elementary contributions to the total energy state of the cluster in the alloy were calculated by locating a Ni atom in a position in the atomic rows normal to the cluster habit plane. Cluster-Ni binding energy vs. row position and distance to the cluster habit plane were obtained.

In the other, we sampled different configurations over a particular Fe–Ni alloy using the approach developed in Ref. [22]. In this model, a crystal, with randomly distributed Ni atoms with a particular concentration, C_{Ni}, was simulated. The SIA cluster was then inserted in the crystal and relaxed. Next, the cluster position was shifted by the Burgers vector, $\boldsymbol{b} = \frac{1}{2}\langle 1\,1\,1\rangle$, and the system was relaxed again. The process continued until all the configurations along the $\langle 1\,1\,1\rangle$ directions were tested. If the sampled path is long enough, all the possible configurations are tested. Here we studied $N = 1000$ positions (crystal length along $\langle 1\,1\,1\rangle$ direction was $1000|\boldsymbol{b}| \approx 250$ nm). The effective binding energy, $E^{eff}_{NSIA}(C_{Ni})$, depends on the temperature T and is defined as:

$$E^{eff}_{NSIA}(C_{Ni}) = kTln\left[\frac{\sum_i^N \exp(E_i - E_{min}/kT)}{N}\right] \quad (2)$$

where k is the Boltzmann constant, E_{min} and E_i are the minimum and the current potential energies calculated after each relaxation in the system containing an NSIA cluster

[1] The pseudopotentials + plane wave DFT approach was used to verify LSMS in modeling Ni and Cr interacting with defects in Fe in small, ~100–200 atoms, systems. LSMS + MS was then used to model interaction with edge and screw dislocations in ~1200 atoms system.