Suppression of vacancy cluster growth in concentrated solid solution alloys

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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 $\text{Ni}_{0.5}\text{Fe}_{0.5}$. [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 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 $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$, suggesting that $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$ 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.

Vs 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 Vs 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-symmetry 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 Vs 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 Vs in CSAs predicted from MD simulations, Ni, $\text{Ni}_{0.8}\text{Fe}_{0.2}$ and $\text{Ni}_{0.8}\text{Cr}_{0.2}$ were irradiated at room temperature with 1.5 MeV Mn

ions at 7° off the $\langle 001 \rangle$ direction to avoid channeling effects. Cascade damage accumulation at irradiation fluences of up to $2 \times 10^{14} \text{ cm}^{-2}$ and $1 \times 10^{16} \text{ cm}^{-2}$ was studied; and the resulting disorder profiles, mainly interstitial-type loops and vacancy-type SFT, were determined from Rutherford backscattering spectrometry (RBS) along the $\langle 001 \rangle$ 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 vacancy numbers n ($n=1, 9$, and 13) in Ni, $\text{Ni}_{0.5}\text{Fe}_{0.5}$, $\text{Ni}_{0.8}\text{Fe}_{0.2}$, $\text{Ni}_{0.8}\text{Cr}_{0.2}$, and $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$ at 1000 K is presented in Fig. 1. These particular vacancy numbers are chosen to illustrate the trend of diffusion coefficients in different systems with increasing VC sizes. A relatively high temperature of 1000K is employed to make the diffusion observable on the MD timescale.

For a single vacancy, the MSD is smaller compared to large VCs, indicating that the diffusion of large VCs is faster than the diffusion of a single vacancy. In addition, a linear dependence of the MSD on time is observed. The diffusion coefficients, determined by the slopes of these curves, suggest that the diffusion of a single vacancy is much slower in Ni than in $\text{Ni}_{0.5}\text{Fe}_{0.5}$, $\text{Ni}_{0.8}\text{Cr}_{0.2}$ and $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$. It is

interesting to note that the diffusion of a single vacancy in $\text{Ni}_{0.8}\text{Fe}_{0.2}$ is slower than in $\text{Ni}_{0.5}\text{Fe}_{0.5}$ as well as in pure Ni (Fig. 1(a)). This finding is related to the site percolation threshold in binary *fcc* alloys, which is around 21%. [30] In this case, the vacancy diffusion is limited because long-range connectivity within the Fe content does not exist.

Diffusion coefficients are lower in Ni than in the CSAs when n is less than 9, but they become comparable at $n \sim 9$. For $n=13$, lower diffusion coefficients in the CSAs are observed. These results suggest that the diffusion of large VCs in CSAs is greatly suppressed compared with their diffusion in pure Ni. More important, the diffusion coefficient of VCs can be significantly modified by changing the alloying element and concentration. For example, a comparison between $\text{Ni}_{0.8}\text{Fe}_{0.2}$ and $\text{Ni}_{0.5}\text{Fe}_{0.5}$ in which 20% and 50%, respectively, of the Ni atoms in pure Ni are substituted with Fe atoms, finds that the diffusion coefficient of large VCs with $n=13$ is much reduced in $\text{Ni}_{0.5}\text{Fe}_{0.5}$. A significant alloying impact is also evident in $\text{Ni}_{0.8}\text{Fe}_{0.2}$ and $\text{Ni}_{0.8}\text{Cr}_{0.2}$, where 20% of the Ni atoms in pure Ni are replaced with Fe or Cr, respectively. Here, the enhanced diffusion of small VCs, such as $n=1$ and 9, is clearly shown in $\text{Ni}_{0.8}\text{Cr}_{0.2}$. Modulation of the diffusion coefficients is further achieved in ternary $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$, in which half of the Ni atoms in $\text{Ni}_{0.8}\text{Cr}_{0.2}$ are further substituted with Fe atoms, as shown by the lowest slope in Fig. 1(b) and 1(c). A more detailed comparison of diffusivities will be discussed later.

While the MSD of a single vacancy in different systems is rather smooth, the MSD for the large VCs presented in Fig. 1 with $n=9$ and 13 exhibits some plateaus, especially in pure Ni. The MSD increases for a while and then remains almost unchanged with time. These features indicate stagnation stages during the time intervals. By inspecting the atomic configurations, it is found that the VCs are dissociating. The dissociation happens more easily in pure Ni than in CSAs. After the dissociation, the main VC moves, leaving one vacancy behind. This process continues for several nanoseconds and creates several discrete vacancies in the cell. Since a single vacancy is almost immobile, as shown in Fig. 1(a), its position does not change with time. On the other hand, small VCs have relatively larger mobilities and they can

randomly diffuse in the cell until they encounter a discrete vacancy and recombine. To better illustrate this process, we take a perfect stacking fault tetrahedron with $n=15$ as an example. It is generally assumed that perfect SFT are immobile after creation. However, considerable mobility is found in this study based on the potential used, because of dissociation. The MSD is given in Fig. 2, together with the evolution of the defect structures.

It can be seen from Fig. 2 that the perfect tetrahedron initially diffuses with a low mobility until it decomposes into a single vacancy and an interstitial-vacancy cluster at about 95 ns. After that, the slope of the MSD changes to a larger value, which means that the defective cluster has a larger diffusivity than the perfect tetrahedron. The faster diffusion of this cluster occurs as a result of its randomly traveling until it comes across the isolated vacancy and becomes a perfect tetrahedron again. Consequently, the MSD becomes flat as the diffusion slows at around 180 ns. Decomposition occurs again at around 217 ns, and the slope of the MSD again becomes steep. The dissociation leads to some non-linear characteristics in the MSD. As a result, a long simulation time is required to calculate the diffusion coefficient from the MSD to minimize the uncertainty induced by these local non-linear features. In this study, simulations of more than 1 μ s were carried out to extract the diffusion coefficients, as shown in Fig. 1.

The normalized self-diffusion coefficient D^* at 1000 K calculated from the MSD is summarized in Fig. 3. It shows that the D^* of small VCs is much lower in pure Ni than in alloys. In contrast, adding Fe or Cr significantly enhances the diffusion of small clusters. Nevertheless, the D^* in the CSAs decreases with increasing cluster size, and finally it becomes much lower than in pure Ni. This is more noticeable in $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$, where the diffusion coefficient decreases more remarkably with increasing VC size. The threshold vacancy number for this transition is around 7-13 in different CSAs. This observation indicates that large VCs are less mobile in CSAs than in pure Ni. Therefore, these VCs will stay in their original locations for a long time after they are created by, for example, cascade collisions induced by ion

irradiation. Hence, the growth of VCs by the incorporation of additional vacancies or small clusters[10] is suppressed in these CSAs, especially in $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$.

Diffusion coefficients shown in Fig. 3 also indicate that small VCs migrate faster than a single vacancy. This is related to the fact that the barrier for the movement of a defect in the surface of a cluster is relatively small.[18] Consequently, the rearrangement of surface defects is greatly facilitated and the movement of VCs is favored. The high mobility of small VCs suggests that these clusters can diffuse quickly and coalesce with other clusters to create even larger clusters. The process is captured by the MD simulations, as also observed in a previous study.[18] Figure 3 further demonstrates that the mobilities of perfect SFT are rather low ($n=15, 21$, and 28). However, it shows that SFT with 15 vacancies still have relatively large mobilities in Ni and $\text{Ni}_{0.8}\text{Fe}_{0.2}$ because of the dissociation of the SFT, as illustrated in Fig. 2. However, this behavior is not observed in $\text{Ni}_{0.5}\text{Fe}_{0.5}$, $\text{Ni}_{0.8}\text{Cr}_{0.2}$ and $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$. In those alloys, most of the VCs bind together and the MSD exhibits a smooth dependence on the simulation time, as is evident in Fig. 1. Figure 3 also shows that the diffusion of VCs in Ni and $\text{Ni}_{0.8}\text{Fe}_{0.2}$ is very similar. The whole diffusion trajectories of perfect SFT with 15 vacancies in Ni and its CSAs are provided in the Supplementary movies.

The observed dissociation of VCs may depend on the potential used. To make sure the earlier conclusion is a general trend in CSAs, the results in pure Ni using a different potential developed by Mishin et al. are also presented in Fig. 3. Using this potential, the VCs do not dissociate during the simulations in the temperature range considered. Hence the Mishin potential provides a good cross-check of the results obtained from the Bonny potential. It can be seen that the variation in diffusion coefficients, along with the number of vacancies, is similar for both potentials. Nevertheless, the Mishin potential yields a rather low diffusivity for a perfect tetrahedron with 15 vacancies. This is expected because perfect SFT rarely move once they are formed. This comparison suggests that both potentials support the conclusion that the

diffusion of VCs is slower in CSAs than in pure Ni when the number of vacancies is high, as can be seen in Fig. 3.

The results presented in Fig. 3 indicate that $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$ provides the most effective alloying composition to suppress VC growth, since the VC diffusion coefficients decrease rapidly as the size of VCs increases. This is a synergistic effect of the Fe and Cr alloying elements. Indeed, comparison with binary alloys suggests that $\text{Ni}_{0.5}\text{Fe}_{0.5}$ and $\text{Ni}_{0.8}\text{Cr}_{0.2}$ have the most influence on VC mobility. Diffusion is much faster for small VCs and slower for large VCs, a finding also reflected in the activation energies. The calculated D^* at different temperatures is used for the Arrhenius fit: $D^* = D_0 \exp(-E_a/k_B T)$, where k_B is the Boltzmann constant and D_0 is the pre-exponential factor. The fitting results for $n=1$ and 13, as well as the obtained activation energies E_a for different VCs, are displayed in Fig. 4. Note that the dependence of $\log D^*$ on $1/T$ can be represented well by a linear relation for these clusters. Figure 4 reveals a high activation energy for $n=15$, since it is a perfect stacking fault tetrahedron that is rarely mobile. This result also suggests that VCs of moderate size have smaller activation energies than either single vacancies or large VCs. In addition, it is indicated that $\text{Ni}_{0.5}\text{Fe}_{0.5}$ and $\text{Ni}_{0.8}\text{Cr}_{0.2}$ have the largest effect on E_a , since the E_a in both alloys are lowest for $n=1$ and highest for $n=13$ and 15. Therefore, these two alloys are more effective in inhibiting VC growth owing to the alloying effect of Fe and Cr.

The results presented show that the VC growth is suppressed in CSAs because the mobilities of VCs are modified. This is in line with experimental observations of vacancy-type defects in pure Ni and CSAs.[15,26,27] Consequently, these alloys are expected to be more resistant to vacancy-related effects, such as volume swelling.

3.2 Features of different EAM potentials

It is a common concern in classical MD simulations that interatomic potentials may significantly influence the results obtained. For the Ni-Fe-Cr system studied in this work, there are two potentials available, both developed by Bonny et al.[21,31] and denoted here as Bonny2011 and Bonny2013. Both potentials were fitted to a wide range of pure metal and alloy properties. However, these two potentials predict different results regarding the stable structures of VCs. Whereas Bonny2013 yields perfect SFT as the most stable VCs when the number of vacancies is a magic number, Bonny2011 predicts triangular [111] Frank loops as the most stable configurations. Since SFT are generally thought to be the most stable forms of VCs in *fcc* metals such as Ni, and SFT are also observed experimentally in some CSAs,[4,15,19] Bonny2013 is selected in this work to study the diffusion properties. Although it predicts SFT are the most stable VCs at 0 K, the dissociation of SFT at temperatures higher than 800 K is observed. On the other hand, the Mishin potential predicts that SFT are stable even at high temperatures. This leads to predictions of different diffusion behaviors for VCs. Nevertheless, the conclusion that large VCs exhibit low mobility in CSAs is confirmed by the cross-check between these two potentials.

The observed different stabilities of SFT are investigated by comparing their binding energies. The results calculated from the Bonny2013 and Mishin potentials are displayed in Fig. 5. The binding energy of a stacking fault tetrahedron (E_b^n) is calculated as $E_b^n = E_f^1 - E_f^n$, where E_f^1 and E_f^n are the formation energies of a single vacancy and a tetrahedron with n vacancies, respectively. The formation energy is given by $E_f^n = \frac{1}{n} [E(N - n) - E(N) + \frac{nE(N)}{N}]$, where $E(N)$ and $E(N - n)$ is the total energy of a perfect system containing N atoms and a defective system containing n vacancies, respectively. The formation energy defined here is normalized by the number of vacancies. The results at 0 K provided in Fig. 5(a) suggest that these two potentials give similar binding energies for SFT. Nevertheless, the Mishin potential results in slightly higher binding energies compared with Bonny2013. For SFT with more than 10 vacancies, higher binding energies are always found in perfect SFT ($n=10, 15, 21, 28\dots$). Since the

binding energies at 0 K are similar for both potentials, the different behaviors of SFT at high temperatures come from the different thermodynamical quantities based on these two potentials.

To further analyze the temperature effects, the binding energy of a perfect stacking fault tetrahedron with $n=15$ is calculated by taking the lattice thermal expansion into account. This method was successfully implemented to analyze the temperature-dependent defect energetics in Ni.[33,34] Although it does not include the effects of lattice vibrations induced by temperature, this approach provides reasonable results for the temperature dependence of binding energies. Note that these two potentials result in strikingly different thermal expansion coefficients (α), defined as $(a-a_0)/a_0$, where a and a_0 are the lattice constants determined at non-zero temperatures and 0 K, respectively. According to α , different lattice parameters correspond to different temperatures. The melting temperatures (T_m) are 1530 K and 1700 K, based on the Bonny2013 and Mishin potentials, respectively.[32] For comparison, the temperature dependence of E_b^{15} as a function of T/T_m is shown in Fig. 5(b), indicating that the binding energy decreases with increasing temperature. In particular, the decrease based on the Bonny2013 potential is more pronounced than the decrease based on the Mishin potential. The results at 1000 K presented in Fig. 5(a) show that the binding energies of SFT based on Bonny2013 become rather low, and the low binding energies lead to instability of the SFT during the MD simulations. In particular, the binding energy differences between SFT with vacancies of adjacent numbers become much smaller. On the other hand, the SFT are still stable at high temperatures based on the Mishin potential because of their high binding energies.

3.3 Experimental evidence

To validate the MD results and further evaluate the feasibility of using the Bonny2013 potential to predict VC behaviors, ion irradiation-induced disorder in CSAs is compared under two different fluences, $2 \times 10^{14} \text{ cm}^{-2}$ and $1 \times 10^{16} \text{ cm}^{-2}$. The corresponding displacements per atom (dpa) at the damage peak are ~ 0.27 and

13.5, respectively. Irradiation-induced disorder is determined from RBS along the $\langle 001 \rangle$ channeling direction.[28] The RBS yield is mainly attributed to the dechanneling ions due to lattice distortions resulting from both interstitial loops[35] and SFT.[36] The disorder level at 1.0 is equivalent to an amorphous state in which no crystallinity remains, whereas the defect-free crystal has a disorder level of zero. Owing to the relatively low-dose irradiation, damage accumulates and leads to increasing lattice strain/distortion, which peaked at ~ 360 nm, as shown by the data points in Fig. 6(a) and by the unfilled arrow in Fig. 6(c). After further irradiation, irradiation-enhanced defect migration driven by the accumulated strain at the damage peak region is clearly observed, as shown in Fig. 6(b) and the three filled arrows in Fig. 6(c). SFT and interstitial loops can move both toward the surface and deeper into the sample under ion irradiation. Whereas defects migrating to the surface anneal at the surface, defects migrating into the samples cause a buildup of local strains that drives the defects even deeper under prolonged irradiation.[19] In a recent experimental study of 3 MeV Au-irradiated Ni, $\text{Ni}_{0.5}\text{Co}_{0.5}$, and $\text{Ni}_{0.5}\text{Fe}_{0.5}$ using cross-sectional transmission electron microscopy (TEM) and ion channeling, Lu et al.[19] reported that the defects migrate much deeper in Ni than in the two binary alloys, suggesting higher defect mobility in Ni. Their TEM results showed that SFT and interstitial loops coexist in the same damage region.[19] Moreover, their results demonstrated large depth shifts in the damage peak position that are also found in the present study. Since the densities of Ni and its solid solution alloys are similar,[29] the implantation ranges of Mn in these samples should be comparable. The large differences in observed damage depth must originate from the different diffusion properties of defects. Such irradiation-induced defect migration is clearly observed in Fig. 6(c), where the damage peak in Ni moves from ~ 360 nm to ~ 730 nm. But it is much less apparent in the binaries, from ~ 360 nm to ~ 560 nm in $\text{Ni}_{0.8}\text{Fe}_{0.2}$ and to ~ 600 nm in $\text{Ni}_{0.8}\text{Cr}_{0.2}$. The deeper disorder profile in Ni than in the alloys is consistent with the present MD simulation results for SFT diffusivities.

These experimental observations validate our MD results, although the dissociation of SFT based on the employed empirical potential may raise some uncertainties. For Ni, an independent potential is used to

evaluate the VC kinetics. Lower mobilities of larger VCs are observed based on both potentials. For the Ni-Fe-Cr system, the previous 2011 potential[31] predicts that VCs are in the form of vacancy platelets instead of SFT. We have tried to study VC diffusion using this potential and find that, for more than 5 vacancies, the clusters keep vibrating at the original places and no long-range diffusion is observed. For very small VCs (<5), this potential displays larger diffusivities in alloys than in pure Ni, which is in agreement with the present results based on the Bonny2013 potential. The main uncertainty comes from the SFT dissociation. As illustrated in the Supplementary movies, SFT are less likely to dissociate in the alloys, indicating that the results for alloys may be reliable. Moreover, these MD results for diffusivities are supported by experimental evidence. The conclusion of the present study, therefore, reflects a general property of CSAs.

4. Conclusions

MD simulations are carried out to study the diffusion of VCs in CSAs, including $\text{Ni}_{0.5}\text{Fe}_{0.5}$, $\text{Ni}_{0.8}\text{Fe}_{0.2}$, $\text{Ni}_{0.8}\text{Cr}_{0.2}$, and $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$. Diffusion of large VCs containing more than 7-13 vacancies is much slower in CSAs than in pure Ni, which suggests the effective suppression of large VC growth in these alloys. This is consistent with experimental observations that irradiation-induced damage appears much deeper in Ni than in $\text{Ni}_{0.8}\text{Fe}_{0.2}$ and $\text{Ni}_{0.8}\text{Cr}_{0.2}$ alloys. These results provide insight into different VC behaviors in these systems: whereas larger clusters are frequently observed in pure Ni, smaller VCs are found experimentally in CSAs. Furthermore, our MD work predicts that $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$ provides the most effective alloying environment for inhibiting VC growth. As effective reduction of large VCs or SFT formation is necessary to minimize the volume swelling and alleviate radiation hardening of structural materials, the present study on alloying effects in model concentrated solid solutions suggests an alternative pathway to control the formation and growth of VCs, especially SFT, in structural alloys.

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Supplementary movies

Supplementary movies of the diffusion of perfect SFT with 15 vacancies in pure Ni and alloys at 1000K considered in this work. Different alloys are denoted in the filenames. For pure Ni, the trajectories from two different potentials are given. In the movies, vacancies are represented by green balls and interstitials are represented by blue balls.

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Figure 1 MSD for vacancy clusters with different size N (1 (a), 9 (b) and 13 (c)) in Ni, $\text{Ni}_{0.5}\text{Fe}_{0.5}$, $\text{Ni}_{0.8}\text{Fe}_{0.2}$, $\text{Ni}_{0.8}\text{Cr}_{0.2}$ and $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{Cr}_{0.2}$ at 1000 K.

Figure 2 The MSD for a perfect stacking fault tetrahedron with $N=15$ in pure Ni at 1000K within a short time to illustrate the plateau features. The initial perfect tetrahedron is shown by orange balls with non-zero centro-symmetry parameters. During the diffusion simulations, vacancies are denoted by green balls while interstitials are represented by blue balls. These defects are obtained from Wigner-Seitz defect analysis.

Figure(s)

Figure 3 Diffusion coefficients of VCs with different sizes at 1000K. For pure Ni, the results from two different potentials are reported.

Figure(s)

Figure 4 Arrhenius fit for vacancy clusters with N=1 (a) and N=13 (b) as well as the determined activation energy (c) for different size of vacancy clusters.

Figure 5 (a) Binding energies of SFT in pure Ni with different number of vacancies at 0K and 1000K predicted by Bonny2013 and Mishin potential. (b) Temperature dependence of the binding energy for perfect SFT with 15 vacancies in pure Ni by taking the thermal expansion into consideration. The thermal expansion coefficient α is given in the insets. The temperature is given by T/T_m where T_m is the melting temperature of Ni predicted by the employed potential.

Figure 6 (a) Disorder profiles in Ni, $\text{Ni}_{0.8}\text{Fe}_{0.2}$ and $\text{Ni}_{0.8}\text{Cr}_{0.2}$ resulting from 1.5 MeV Mn irradiation to fluence of (a) $2 \times 10^{14} \text{ cm}^{-2}$ and (b) $1 \times 10^{16} \text{ cm}^{-2}$. The lines in (a) and (b) are the curve fit to the data to guide the eye and are summarized in (c) to clearly shown irradiation-induced damage migration under prolonged irradiation, as indicated by the distance between the corresponding arrows.

Graphical abstract