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ABSTRACT
The radiation resistance mechanisms of nanoclusters (NCs) in oxide dispersion-strengthened (ODS) steels have been investigated. Molecular dynamics simulation has been used to investigate defect generation during the primary damage state of a displacement cascade in ODS steels for NCs of various radii and a range of primary knock-on atom (PKA) energies. Y2O3 NCs considerably enhance the radiation resistance of ODS steels by reducing the peak defect generation during the cascade within the Fe matrix. The NC also affects the morphology of the collision cascades, depending on PKA energy. At lower energies, the NC’s outer circumference act as a cessation point forming a dampened shockwave compared to a pure Fe system. At higher energies, the PKA energy is able to transfer through the NC, thus causing two smaller shockwaves in the Fe matrix. Along with the alteration of the cascade morphology, the NC boundary acts as a strong defect sink to absorb defects and defect clusters, leading to significant recombination of interstitials and vacancies away from the NC. The interfacial energy of the NCs with the Fe matrix increases with increasing diameter of the oxide NCs. The evolution of the NC is tracked through the primary damage state of a cascade, and the effects of ballistic dissolution play a key role in this evolution, most evident in the 2 nm NC.

I. INTRODUCTION
Next generation reactors require higher irradiation tolerant structural materials for more corrosive environments and higher temperatures. One of the popular candidate materials is oxide dispersion-strengthened (ODS) steels. ODS steels have been proposed as duct, cladding, and structural materials for fission reactors and as a material for the first wall and a blanket material in fusion reactors.1–3 There have been many experimental studies on the benefits of nanostructured ferritic alloys (NFAs). Several review papers on NFAs have shown that these materials have high yield strength and high irradiation resistance.4,5 NFA’s high irradiation resistance is due to nanoclusters (NCs) acting as successful sites in trapping defects and NC’s ability to act as a site for recombination.4,5 Although ODS steels, in theory, should have excellent irradiation resistance, ion irradiation experiments have shown conflicting results. A recent review paper published by Wharry et al. summarized the inconsistent experimental results from ion and neutron irradiation experiments on ODS steels.7 Depending on the material composition, consolidation and heat treatment performed, and the type of ions used for the irradiation, the results of the experiments varied greatly, with reports published stating that opposite trends are occurring in these NFAs.8

The irradiation stability of NCs is a major area of experimental research. As the NCs are one of the key factors in the strengthening of ODS steels, these materials have advantageous irradiation resistance properties for next generation reactors. The major ways of tracking the irradiation stability of NCs are in studying the evolution of size distribution, number density, volume fraction, the chemistry of these NCs, and the changes to the surrounding crystal structure as a function of dpa.9 Prior to irradiation, it has been shown, both experimentally and computationally, that NCs tend to be stable within the Fe matrix.9–10 After irradiation, irregularities have been observed in the NCs; atom probe topography (APT) is used to corroborate the dissolution of Y2O3 NCs, along with NC’s composition changing heavily after irradiation.11 Other experiments have shown an increase in the irregularities between the
Fe-matrix and the NC interface. Fundamental computer simulations play a key role in furthering the understanding of the irradiation evolution of ODS NCs.

Density Functional Theory (DFT) calculations have been used to understand the effects elements have on the formation and structure of NCs. Along with DFT calculations, higher order simulations, such as thermodynamic and kinetic Monte Carlo models, have shown the evolution of NCs during the heat treatment common in ODS steel manufacturing. All of these studies have advanced the understanding on the formation of NCs in ODS steels. In furthering the understanding of the primary damage state of cascades in NCs, studies have focused on pure Y2O3 damage, and the effect of an embedded NC on the damage evolution in an Fe matrix at very low energies.

Molecular Dynamics (MD) has been the main tool for looking at the primary damage state of displacement cascades. In this study, MD is employed to understand defect generation, damage morphology, radiation resistance, and enhanced defect recombination in ODS steels for NCs with various radii and a range of primary knock-on atom (PKA) energies. The simulations with various PKA energies provide significant insights into the relative stability of ODS steel NCs and their evolution within a displacement shockwave. Using MD to analyze the effects of displacement cascades on Y2O3 NCs furthers the understanding of the primary damage state of NCs with the surrounding α-Fe matrix in ODS steels and plays a key role in furthering the understanding of the irradiation properties of NCs.

II. SIMULATION APPROACH

All interactions between atoms are governed by an empirical potential, or force field, in MD simulations. The interatomic potential describing all of the interactions for the Y and O atoms in the simulations is developed by Hammond et al. and uses the form suggested by Buckingham. The Buckingham form uses a Born-Mayer potential to represent the Pauli repulsion of electron clouds at close distances coupled with a long range attraction term. The Born-Mayer potential is successful at modeling the equilibrium state of the structure; however, nonideal for nonequilibrium situations common in primary cascade simulations. To remedy this issue, a universal repulsive potential of the form, Ziegler, Biersack, and Littmark (ZBL) formula, is used to better model these non-equilibrium conditions. The ZBL potential and the Born-Mayer potential are connected via a polynomial equation; the basic setup is shown in Eq. (1). More information on the specific cutoffs for each atom pair and the dE term, a term to connect the two different potentials, can be found in Hammond’s paper. The Fe potential used for these simulations is an EAM potential developed by Marinica et al., which is shown to agree with experimental results and DFT calculations. The potentials have been previously employed to model the irradiation properties of α-Fe.

\[
V(r) = \begin{cases} 
V_{\text{ZBL}}(r) + dE & r \leq r_1 \\
\sum_{n=0}^{2} a_n r^n & r \in (r_1, r_2) \\
V_{\text{Buckingham}}(r) f_{\text{cut}}(r) & r \geq r_2 
\end{cases}
\] (1)

The MD simulations are performed using large-scale atomic/molecular massively parallel simulator (LAMMPS). The details for setting up a Y2O3 NC in a Fe matrix are described in Ref. 10, and only the central principles are described here. Initially, a Y2O3 NC is placed at the center of a Fe matrix, and the system is relaxed using the conjugant gradient method to release the accumulated stress. Then, the systems’ temperatures are raised to 100 K and equilibrated for 15 ps with the NVT (constant atoms, volume, and temperature) canonical ensemble, thus, achieving the minimum energy configuration of the Y2O3 NCs. The cascade simulations took place at a temperature of 100 K, with the system being equilibrated over 50 000 time steps for a total time of 50 ps to achieve an equilibrium state of the phonons. The temperature is set to 100 K to better illustrate the effects of the NC, without the benefit of temperature, during the primary damage state of a cascade. As research has shown that at higher temperatures, the peak damage is not significantly changed; however, the key difference being at higher temperatures, the thermal peak period is increased leading to increased migration of defects.

The PKA is placed 1.5 nm away from the outer region of the NC and given a velocity toward the center of the NC. Figure 1 shows the standard setup, prior to the PKA receiving the additional kinetic energy. These simulations are computationally expensive due to the necessarily small-time steps taken so that the relatively high change in the energy of the system is maintained and no atoms are lost. In the simulations, an adaptive time step is required with the following limiting factors: \(10^{-8}\) to \(10^{-3}\) ps time limit and a max distance traveled of 8 to 2 pm, decreasing with increasing PKA energy. A strict adaptive limit is set for the first picosecond of each simulation, and after the peak energy transfer from the PKA to the box, the adaptive limit is loosened to allow shorter run times for the remainder of the cascade simulation. The max distance is increased to 8 pm for all simulation cases after 1 ps; the simulations are halted at a time of at least 20 ps, when the damage state is at

![FIG. 1. An illustration of the Fe-NC system after equilibration with the non-PKA Fe atoms removed. The Fe PKA is given kinetic energy and a direction toward the center of the NC to simulate a displacement cascade within the NC.](image)
equilibrium. Through the use of an adaptive time step, the simulation is able to run smoothly over the entire run time, and also decreases the run time, while preserving accuracy. All simulations are carried out for a fixed volume and fixed number of atoms, with a periodic boundary condition along the three dimensions. A proper MD-cell size is chosen so as to avoid overlap of the displacement cascade with itself due to the periodic condition. For a displacement cascade with a 10 keV kinetic energy PKA, a crystal of ~250 000 atoms (50 × 50 × 50 Fe unit cells) for the 2 nm and 4 nm NC systems and a crystal of ~430 000 (60 × 60 × 60 Fe unit cells) for the 6 nm NC system are employed for this simulation.

A key tool in evaluating the number of Frenkel pairs created during a primary cascade simulation is Wigner-Seitz defect analysis. Wigner-Seitz defect analysis consists of comparing the location of atoms in an initial image of the Fe-NC system to the system as the PKA disperses energy into the surrounding system. By comparing the α-Fe NC system to the perfect system, postequilibration, the interstitials, antisites and vacancies are recorded based on the differences. Along with calculating the Frenkel pairs via Wigner-Seitz analysis, Ovito is also used to illustrate the simulation box, and track the formation of defects and defect clusters in the system.  

III. RESULTS

A. Nanocluster structure

For small diameter NCs, less than 2 nm, the preferential structure of Y2O3 is a disordered structure and with increasing diameter, the NCs formed a core-shell structure in the Fe matrix. Y2O3 NC core has the standard cubic Y2O3 structure, bixbyite type, surrounded by a shell of amorphous Y and O atoms. Surrounding the NC, the Fe atoms are displaced from their lattice sites. A core-shell structure for Y2O3 NCs is validated both experimentally and computationally via first principle calculations. In furthering the understanding of the NC structure, the next step is to understand the effects of NCs on the evolution of the primary damage state of a cascade in the Y2O3 NC and in the surrounding Fe matrix. In this study, a Fe PKA interacts with a NC at various energies, and the results are compared to a perfect bcc Fe system at the same PKA energies.

Table I shows the simulation conditions that are run for this study, and NCs of various sizes are compared. The small NCs are the focus of this work, as small NCs play a key role in the strength of ODS steels. Three NC diameters are investigated in the current work: 2 nm, 4 nm, and 6 nm. Three PKA energies are investigated in the current work: 1 keV, 5 keV, and 10 keV for each NC. All of the simulations are run at a temperature of 100 K to better understand the effects the inclusion of NCs have on the Fe matrix, by limiting the migration of defects due to the temperature. In this research, the irradiation stability and evolution of damage of the Y2O3 NCs, along with the effect NCs have on the Fe Frenkel pair evolution in the Fe matrix, are the primary concerns. In Figure 2 the key regions of interest in this study; the Fe matrix, the atoms directly surrounding the Y2O3 NC and the Y2O3 NC itself. Through the location and clustering of the defects, the effects of adding a Y2O3 cluster are analyzed. By combining the information obtained from Wigner-Seitz defect analysis and cluster analysis to compare the number of Frenkel pairs at the peak damage and at the final damage state, after equilibration at the end of the simulation; the effect of adding NCs to primary cascade evolution is further understood.

B. Fe matrix

Understanding how NCs affect defect evolution in the Fe matrix is an imperative step in understanding the role NCs have in strengthening the irradiation resistance of ODS steel. The first step to explore is how the NCs affect the evolution of the defect populations within the Fe matrix. For low energies (1–10 keV), the primary cascade damage for pure α-Fe has been heavily investigated using MD. Figure 3 shows a standard displacement cascade for a 10 keV pure Fe PKA. The PKA interacts with the surrounding atoms, with the damage being dispersed within a single shockwave. The shockwave consists of an inner sphere of vacancies and an outer shell of interstitials. After the thermal peak, recombination dominates and within a few picoseconds, the system reaches an equilibrium state, with a few remaining Frenkel pairs. In comparison to

<table>
<thead>
<tr>
<th>NC diameter (nm)</th>
<th>No. of cases</th>
<th>Temp (K)</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>16</td>
<td>100</td>
<td>1, 5, and 10</td>
</tr>
<tr>
<td>2</td>
<td>16</td>
<td>100</td>
<td>1, 5, and 10</td>
</tr>
<tr>
<td>4</td>
<td>16</td>
<td>100</td>
<td>1, 5, and 10</td>
</tr>
<tr>
<td>6</td>
<td>16</td>
<td>100</td>
<td>1, 5, and 10</td>
</tr>
</tbody>
</table>
Fig. 3. Fig. 4 shows the evolution of the Fe defects with the inclusion of a NC and a 10 keV PKA for the 2, 4, and 6 nm NCs at 100 K. With the inclusion of a NC, the cascade morphology changes significantly with PKA energy. At lower energies, 1 and 5 keV, the NC acts as a cessation point for the cascade, causing a cascade to occur at the NC-Fe boundary. For the 10 keV PKA, a major difference occurs compared to the standard Fe shockwave. At this energy, the PKA is able to penetrate and transfer energy through the NC causing two smaller shockwaves on separate sides of the NC.

The NCs significantly change the damage morphology of cascades within the Fe matrix, via the NC acting as a cessation point for the peak damage state in the Fe matrix or splitting up the shockwave, depending on the PKA energy. At all energies, the NC preempts the shockwave and due to the higher coordination number of cubic Y₂O₃ causes the peak damage state to occur at smaller times compared to a pure Fe system. The decrease in time for the peak damage state increases with increasing NC diameter. At low energy, 1 and 5 keV, it shows that the NC preempts the shockwave, causing less damage to be transferred to the Fe matrix. At 10 keV, where the PKA energy is able to transfer through the NC, the decrease is more noticeable for the 4 and 6 nm NC simulations, with the 2 nm NC being the most similar to a pure Fe matrix. Since the shockwave at 10 keV is larger than the 2 nm NC, the 2 nm NC has less of an effect on the shockwave formation within the Fe matrix. Because of the higher coordination number of cubic Y₂O₃ compared to α-Fe, the NCs are able to quickly disperse the PKA energy to the system (specifically to the NC itself),
causing a significant drop in the energy dispersed back into the Fe matrix and causing the Fe shockwaves to occur and disperse quicker compared to a pure Fe system.

The Fe defect population is broken into two regions based on Fig. 2, the defects that populated the Fe matrix, and the defects that populated the Fe-NC interface region. Figure 5 shows typical cases for the Fe matrix defect evolution for each of the NC diameters compared to a pure Fe matrix at a PKA energy of 10 keV. The cases chosen are cascade simulations that behaved closest to the average values at both the peak and final defect population for each NC diameter. Figure 5 shows that the peak damage within the Fe matrix significantly decreases with increasing NC diameter. However, this significant decrease in Fe defects is less visible in the final number of point defects within the Fe matrix. The vacancy population for the 2 nm and 4 nm NCs are very similar to pure system at 20 ps; whereas the 6 nm shows a noticeable drop in vacancy population at the end of the simulation. Unlike the vacancy population, the final interstitial population is NC diameter dependent. For the 2 nm case, the population of interstitials is very similar to the pure Fe system. However, in the 4 nm and 6 nm cases, there are a large drop in number of defects, with the 4 and 6 nm interstitial populations behaving similarly.

Figure 6 shows average results for the point defects at the peak and final damage states, as a function of NC diameter and PKA energy, based on all the cases run. With an increasing NC diameter, there is a fairly significant decrease in the number of interstitials and vacancies at the peak damage state in the Fe matrix. The decrease in the Fe matrix peak defect population is also strongly associated with the NC size. As the 2 nm case is the smallest, a key aspect being that the 2 nm is smaller than the cascade shockwave, the PKA energy is easily transferred through the NC, such that the 2 nm diameter most resembles a pure Fe system, having nearly the same amount of point defects at the peak damage state. With increasing diameter, more and more energy is transferred to the NC, causing significant drops in the number of defects at the peak damage state within the Fe matrix. With significant drops in the number of defects at the peak damage state, within the Fe matrix, the final damage state varied with NC diameter.

In looking at the averaged final damage state in Fig. 6 (ii) and (iv), the NC diameter has a noticeable effect on the number of defects. The NC has different effects on the interstitials and vacancies. The NC has different effects on the interstitials and vacancies. The vacancy population is less effected by the introduction of a NC, with only a slight drop in the final number of vacancies for the 2 and 4 nm NCs and a more noticeable drop for the 6 nm NC. The interstitial population has a more pronounced result. The 2 nm NC is nearly identical to a pure Fe system, whereas both the 4 and 6 nm NCs dropped the final number of point defects to roughly a 1/3 of a pure Fe matrix. The increased vacancy population is likely an effect of the difference in migration energy between the interstitials and vacancies. With interstitials having significantly lower migration energies, their migration is possible in a MD simulation, whereas
Defect production efficiency is a common parameter in describing and comparing the primary damage of a cascade. Defect production efficiency is defined as the ratio of the surviving number of Frenkel pairs to the number of displacements predicted by the Norgett-Robinson-Torrens formula (NRT). Equation (2) is used to calculate the remaining number of Frenkel pairs based on the NRT model, where $E_d$ is the average threshold displacement energy, a value of 40 eV is used for Fe following the standards introduced by the American Society for Testing and Material (ASTM) and $E_{PKA}$ is the energy of the PKA.

$$v_{NRT} = \frac{0.8E_{PKA}}{2E_d}$$

The major takeaway for the inclusion of a NC in the Fe system is dependent on NC diameter, PKA energy, and defect type. For vacancies, there is a significant drop at lower energies for all three NCs in this study. However, at a PKA energy of 10 keV, only the 6 nm NC shows a drop in the vacancy point defect production efficiency. In the case of interstitials, however, the 2 nm NC behaves unlike the 4 nm and 6 nm NCs. With increasing PKA energies, from 1 keV to 10 keV, the defect production efficiency increases with increasing PKA energy, at 10 keV the 2 nm NC has the same ratio as a pure Fe matrix. The 4 nm and 6 nm NCs show a significantly smaller volume the 2 nm NC has compared to the larger NCs. With increasing PKA energy, the 2 nm NC is similar in size to a 10 keV shockwave, the effect of the NC on the Fe matrix is completely diminished for interstitials. The difference in the interstitial ratio between the 2 nm and the larger NCs is due to the volume ratio of the displacement shockwave to the NC. As the 2 nm NC is similar in size to a 10 keV shockwave, the effect of the NC on the Fe matrix is completely diminished for interstitials.

The defect production efficiency is plotted vs PKA energy in Fig. 7. The introduction of a NC in the Fe system is dependent on NC diameter, PKA energy, and defect type. For vacancies, there is a significant drop at lower energies for all three NCs in this study. However, at a PKA energy of 10 keV, only the 6 nm NC shows a drop in the vacancy point defect production efficiency. In the case of interstitials, however, the 2 nm NC behaves unlike the 4 nm and 6 nm NCs. With increasing PKA energies, from 1 keV to 10 keV, the defect production efficiency increases with increasing PKA energy, at 10 keV the 2 nm NC has the same ratio as a pure Fe matrix. The 4 nm and 6 nm NCs significantly decrease the interstitial ratio at all three PKA energies. The difference in the interstitial ratio between the 2 nm and the larger NCs is due to the volume ratio of the displacement shockwave to the NC. As the 2 nm NC is similar in size to a 10 keV shockwave, the effect of the NC on the Fe matrix is completely diminished for interstitials.

The major takeaway for the inclusion of a NC in the Fe system is that there is an energy dependence on the effects NCs have on the Fe matrix defect population. At low energies (1 and 5 keV), the NC acts as a cessation point for the shockwave, in this situation, the number of point defects is decreased for both the interstitial and vacancy population. The interstitial population drops more than the vacancy population. At higher energies (10 keV), the effect of NC diameter is more evident. As the 2 nm NC compares greatly to the pure Fe system and the 4 nm and 6 nm with the cascade being broken into two smaller shockwaves resemble a new phenomenon. The two smaller shockwaves caused by the 4 nm and 6 nm NCs leads to a noticeable drop in interstitials and vacancies in the Fe matrix. Although at 10 keV, the population of Fe vacancies compared to interstitials within the Fe matrix is higher, likely a result of the mobility of vacancies vs interstitials. During the shockwave, the vacancies occur primarily within the center of the shockwave, with some occurring near the fringes of the shockwave. After the initial wave of recombination with nearby vacancies, some interstitials migrate to the NC.

### C. Fe-Y$_2$O$_3$ NC boundary

The inclusion of a NC shows a noticeable drop in interstitials and a drop for vacancies, for the larger diameter NCs in the Fe matrix; however, a significant population of interstitials and vacancies forms along the outer shell of the NC after the shockwave and during the cascade relaxation process. As shown in Fig. 4, the number of defects surrounding the NC or at the interface increases until a saturation number of defects is reached. Figure 8 shows how the Fe defect populations directly surrounding the NC evolve over time for a 10 keV PKA based on the NC diameter. The interstitials are broken into three smaller groups: pure Fe interstitials, Fe-Y antisites, and Fe-O antisites; where an antisite is defined as an atom sitting on the lattice site of another atom type; in this case, Fe on O lattice sites and Fe on Y lattice sites. Figure 9 shows a plot of the number of Frenkel pair defects as a function of the surface area of each NC. With increasing PKA energy, the following trends are evident: a plateau in the number of defects per surface area surrounding the NC is reached for the 2 nm NC, a characteristic of the significantly smaller volume the 2 nm NC has compared to the 4 nm and 6 nm NC diameters. Whereas the 2 nm NC defect population plateaus, the 4 nm and 6 nm NCs show an increasing number of point defects as a function of surface area with increasing PKA energy, fairly linearly for both the vacancy and interstitial populations, with the 4 nm NC having a slightly higher concentration of Fe defects surrounding the NC.

![Fig. 7. A ratio of the number of surviving defects to the predicted value using the NRT model for pure Fe vs Fe matrix with the inclusion of a NC; (i) Fe vacancies and (ii) Fe interstitials.](image-url)
In evaluating the evolution of Fe antisites, the following is very evident: with increasing PKA energy the number of Fe antisites within the NC increases with increasing NC diameter and PKA energy. Figure 10 plots the average, final antisite population for the 3 NC diameters in this study, the fraction of Fe interstitials that are characterized as antisites within the NC, and the fraction of NC lattice sites filled with Fe antisites. The PKA energy is able to knock significantly more O atoms off of their lattice sites vs the Y atoms, as expected based on the atomic weights of these two elements. The 2 nm case sees the least amount of change in antisite population, with increasing energy, due to the limited size of the NC, and the energy being able to more easily transfer through the NC back to the Fe matrix. The fraction of interstitials that are antisites follows a similar trend with energy; with the fraction increasing with PKA energy and NC diameter. With increasing energy, more Fe atoms are able to penetrate and interact with the NC atoms,
and with increasing NC diameter the number of NC lattice sites significantly increases, giving the Fe atoms a much higher chance of interaction. The effect of increasing NC radii is shown in the final plot, Fig. 10 (iv), with the fraction of Fe antisites on NC lattice sites significantly increasing with decreasing radii.

The Fe defects surrounding the NC over the course of the simulation show that during the primary damage state of a cascade, the formation of a defect sink is occurring at the Fe matrix-NC boundary. In furthering the understanding of the sink formation along the NC boundary, cluster analysis is used to look at the clustering in this region. Figure 11 shows the average number of vacancy and interstitial clusters in relation to the surface area of the NCs. Unlike the Fe matrix, where little to no clustering occurs, the Fe atoms interacting with the NC cause a significant

**FIG. 10.** Plots showing key information for the Fe antisites within the NCs; (i) number of Fe-Y antisites, (ii) number of Fe-O antisites, (iii) the fraction antisites to the interstitial population, and (iv) the fraction of Fe antisites to NC lattice sites.

**FIG. 11.** Distribution of the clusters surrounding the NC based on the NC diameter for a 10 keV cascades; (i) vacancy clusters and (ii) interstitial clusters.
amount of clustering around the NC. The cascade increases the irregularities along the Fe-NC interface, causing a sink to form at the NC boundary. Within the primary damage state, as shown in Fig. 12, the population of defects at the NC boundary is biased toward the Fe interstitials.

By taking into account the surface area of the NCs, the effect of NC diameter did not have a noticeable effect on the clustering of Fe interstitials and vacancies. In a pure bcc system, the clustering occurs primarily during the thermal spike and then shortly afterwards via the diffusion of interstitials and vacancies. In the case of a Fe-NC system, most interstitials and vacancies are generated near the boundaries of the NC during the shockwave. After the peak damage state, the Fe interstitial atoms in the matrix preferentially migrate toward the NC. An interesting phenomenon, shown in Fig. 4, is the migration of defects after the peak damage state. The interstitials and vacancies, during the relaxation process of the cascade, surround the NC forming a uniform-thin layer of defects. These point defects are not limited to the shockwave volume, but over time begin to surround the entire NC. This property shows that Y₂O₃ NCs act as a key factor in irradiation resistance of the Fe matrix. Fe antisite evolution shows that within the NC, there is very little difference in the peak number of antisites during the peak damage state and the number of antisites at the end of the simulation. The antisites that are able to penetrate into the NC during the primary shockwave are maintained and play a key role in the evolution of the NC.

D. Y₂O₃ NC

Figure 13 shows a visualization of the evolution of defects within the NC. Unlike the standard cascade evolution, the damage within the NC continues to grow until it later reaches a plateau level. The Fe antisites impeding into the NC play a key role in this evolution trait of NCs. The Fe antisites within the NC tended to stay in the NC after the initial shockwave, shown in Fig. 8. During the primary shockwave, the NC atoms are displaced. The standard displacement cascade is followed by a thermal peak where interstitials migrate to vacancies and recombination occurs; however, in the case of the NC, due to the combination of both Fe antisites and the incoherent boundary between the Fe matrix and the NC, recombination is considerably more difficult compared to a pure system. Figure 14 shows the evolution of the defect populations within the NC based on the diameter. The 2 nm and 4 nm NC show a slight amount of recombination, whereas the 6 nm NC never has an initial peak defect population early, but rather a gradual growth in the number of defects until reaching a plateau. In the case of increasing NC diameter, more and more PKA energy is transferred to the NC vs the Fe matrix. The key evolution in the primary damage state of a cascade during the NC is that over the course of each simulation, the NC reaches a plateau in damage and is unable to recover back to the NC’s standard cubic structure.
The PKA energies examined show two key ways that the PKA energy can be dispersed through the NC. At lower energies, the PKA energy instead of transferring through the NC, bounces off the NC, causing some damage to the NC, but the resulting shockwave has a minimal effect on the NC compared to higher PKA energies. In the 1 keV and 5 keV cases, the behavior is more comparable, where the 10 keV cases their own category. In these cases, the shockwave is contained to one side of the NC, with the cascade affecting a larger portion of the NC surface area for the 5 keV case compared to the 1 keV case. The PKA energy, rather than transferring through the NC, ricochets off of the NC and back to the Fe matrix. In the 10 keV simulations, the PKA energy is able to penetrate the NC, and the peak damage state occurs with two smaller shockwaves on opposite sides of the NC; however, significantly more energy is transferred to the NC in the 10 keV PKA case. With increasing energy, the initial number of Frenkel pair defects within the NC at the peak damage state increases, causing a larger defect population in the NC.

Figure 15 shows the fraction of NC defects compared to the number of NC lattice sites, for the 2 nm, 4 nm, and 6 nm NCs for the three PKA energies investigated in this study after the defects have reached their plateau population. With increasing PKA energy, the fraction of defects increases for all three cases when comparing the number of defects based on PKA energy and NC diameter. There is a plateauing amount of defects over the course of the cascade within the NC, with this plateau increasing with PKA energy. The plateauing effect within the NCs is due to incoherency between the α-Fe matrix and the cubic Y₂O₃ NC. The incoherency, along with the Fe boundary atoms, seems to cause the NC system to go into disorder. The NC loses its original core-shell structure and is not able to recover throughout the cascade relaxation process. The Y and O atoms stay within the original volume of the NC lattice sites, and a point defect sink quickly surrounds the NC. Although the damage leads to an increase in disorder of the NC, the NC’s atoms stay together, thus future cascades will have to interact with the newly formed point defect sink, prior to the NC.
IV. DISCUSSION

The radiation stability of NCs in ODS steels has been a major area of research for some time. Many neutron and ion irradiations have been conducted to further understand this phenomenon; however, these results have been inconclusive, due to various material compositions, consolidation techniques, and irradiation conditions. The results obtained in the present simulations may help the understanding of NC irradiation resistance. Our results show that with increasing PKA energy, the energy interacts with the NC in various ways. At low energies, the cascade energy ricochets off the NC and stays on one side. For the 10 keV cases, the energy is able to penetrate through the NC. There are a few key aspects to the inclusion of a NC.

A. Point defect populations in the Fe matrix

MD simulations of cascades in α-Fe with a NC have consistently shown fewer displaced atoms created at the end of a cascade compared to the standard empirical models. In comparing the results of a pure α-Fe system to that of the α-Fe NC system, the number of defects within the Fe matrix is dependent on quite a few characteristics: NC diameter, PKA energy, defect type, and PKA shockwave. The Fe matrix with a NC has a small drop in the number of vacancies compared to a pure Fe system. The small drop in vacancies is shown for all PKA energies and NC diameters, the drop increasing with NC diameter. In the case of interstitials, the NC diameter played a larger role. The 2 nm NC sees the interstitial increase with PKA energy and at 10 keV PKA energy there is no difference between the 2 nm NC system and a pure Fe matrix.

The lack of difference between a 2 nm NC and a pure Fe matrix at 10 keV shows the importance of the ratio of the volume of the shockwave to that of the NC. This ratio is an important aspect of a NCs impact on the Fe matrix in ODS steel. Figure 3 shows a standard cascade in a Fe matrix. The key aspect of the cascade is that the shockwave consists of an outer layer of interstitials and an inner sphere of vacancies. As the 2 nm NC is smaller than the shockwave, the interstitials created along the boundary of the shockwave are not affected by the inclusion of a NC. As some PKA energy is still transferred to the NC vs the Fe matrix, a drop in the total number of point defects is still visible for the 2 nm NC. The key aspect of the damage reduction in the Fe matrix is based on the ratio of the volume of a standard shockwave in the pure Fe to the volume of the NC. In comparing the ratio of the volume of a NC to the average volume of a 10 keV shockwave, respectively, for the 2 nm, 4 nm, and 6 nm, the ratios of the volumes are 0.1, 0.7, and 2.3.

In looking at the values of the ratio of the volumes, the 2 nm’s defect production efficiency is very interesting. In the case of the interstitials with increasing PKA energy, the defect production efficiency becomes identical to the pure Fe system. In recalling the morphology of a cascade, a core of vacancies surrounded by a shell of interstitials, the defect production efficiency makes sense. As the 10 keV PKA shockwave is able to completely encapsulate the 2 nm NC, the shell of interstitials occurs unaffected in the Fe matrix, causing the 2 nm NC to have the same defect production efficiency as the pure Fe matrix for Fe interstitials. For the larger NCs, the 4 nm and 6 nm diameter NCs, the significant difference is seen between the interstitial populations within the Fe matrix. The 6 nm NC has a slight decrease in the number of interstitials defect production efficiency to the 4 nm, but the same defect production efficiency for vacancies. The ratio between the NC and the shockwave is very important in limiting the amount of damage within the Fe matrix.

B. Fe-NC boundary

Four major theories have been proposed as controlling NC evolution due to irradiation: ballistic dissolution and irradiation-enhanced, homogeneous nucleation leading to either Ostwald ripening or inverse Ostwald ripening. Further experimental research has shown that the interfacial energy between the Fe atoms and the NC plays a key role in the evolution of the NC. The results in this study demonstrate that the cascade acts in two major ways depending on the PKA energy:

(1) At low energies, the PKA interaction with the NC is a hindrance to the shockwave’s growth, with the NC forcing the shockwave to occur earlier than in a NC free system, thus the NC causes less Fe defects within the matrix.

(2) With 10 keV PKA energy, the cascade energy is able to penetrate and transfer through the NC, causing the standard shockwave to be broken up into two tiny shockwaves on different sides of the NC.

All of the theorized concepts for NC evolution via irradiation occur over a significantly larger time scale than anything that is possible in MD. However, it is possible to understand precursors that could corroborate a potential long-term mechanism. Independent of PKA energy, there is an amorphization of the NC at all PKA energies, with the amount of disorder increasing with increasing PKA energy. Interfacial energy plays a key role in creating efficient point defect sinks. In comparing the amount of disorder caused by a cascade, the fraction of atoms in the NC displaced increases with decreasing radii. The NC disorder, caused by the cascade, reaches a plateau level during the relaxation process of the displacement cascade, with the plateau occurring over longer periods of time for increased NC radii.

A key mechanism that has been backed by experimental evidence is Ostwald ripening of the NCs. Ostwald ripening is the combination of the above phenomena and can either occur with large NCs breaking up into smaller NCs or the dissolution of small NCs leading to an increase in the average size of NCs and a decrease in the number density. In the MD results, there are three major factors that suggest the 2 nm NC is more likely to dissolve compared to the larger NC.

(1) The 2 nm NC has significantly more amorphization than the 4 or 6 nm NCs. Although no Y and O atoms are able to enter the Fe matrix during the primary damage state of a cascade, it is more likely that, with increased amorphization and over much larger time scales, diffusion and ballistic dissolution of NCs is possible, with the more disordered NCs likely to dissolve.

(2) The higher fraction of Fe antisites within the 2 nm NC compared to the 4 and 6 nm NCs suggests that over an extended period of time where many displacement cascades are interacting with NCs, the 2 nm NC will be dissolved at a much faster rate.
(3) When comparing the volume of a shockwave to the volume of a NC, the 2 nm NC is the only NC diameter is this study that is completely encapsulated by a shockwave, having a volume one tenth of a 10 keV shockwave. As 10 keV is a common PKA energy in reactor conditions, this suggests that the tiny NCs will experience ballistic dissolution over larger time scales.

The MD results shown here help to corroborate that the smaller NCs are more likely to dissolve via ballistic dissolution in comparison to the larger NCs. Based on the fraction of antisites within the NCs and the fraction of disorder within the NCs, the 2 nm NC has a significant increase in damage compared to the 4 nm and 6 nm NCs, suggesting that over the course of many dpa, the tinier NCs will dissolve at a faster rate. The MD simulations here are unable to see growth in NCs as only one NC is within each simulation and migration of defects would occur over much longer time scales. However, ballistic dissolution is a key part of Ostwald ripening, and the results shown may indicate that over the course of many dpas worth of damage that Ostwald ripening may be a key mechanism in long term evolution of NC number density and average NC radius.

C. NC irradiation resistance

When comparing the three NCs, all three of the diameters in this study show varying degrees of the same characteristic. In terms of cascade morphology, two common traits are observed, as described above. At low energies, the introduction of a NC causes a single shockwave to occur along the NC boundary, with the peak defect population decreasing with increasing NC diameter. At 10 keV, the cascade is able to transfer energy through the NC, causing two shockwaves to occur on opposite sides of the NC, with a significant decrease in the Fe defect population, which decreases further with increasing NC radii. In looking at the amount of defects remaining in the Fe matrix at the end of the simulation, the 4 nm and 6 nm equally outshine the 2 nm NC ability to hinder Fe defects in the Fe matrix. The 2 nm NC is too small, such that the PKA energy is easily transferred through the system, causing the NC to absorb very little of the PKA’s energy in comparison to the larger NCs. Although only relatively low PKA energies are investigated in this study, a key aspect is shown with the 2 nm NC. When a cascade shockwave is larger in size compared to a NC, the NC’s ability to decrease the amount of point defects in the NC and in the Fe matrix is greatly reduced during the primary cascade damage state.

The next similarity is in the formation of a defect sink surrounding the NC. The population of interstitial to vacancy populations is found to be independent of PKA energy and NC diameter with a value of 1:3, preferential toward interstitials. Along with having a similar ratio of defect populations, the NCs also have a very similar distribution of Fe defect clusters surrounding the NC, although the 2 nm and 4 nm NCs have a slightly higher concentration of Fe defects surrounding the NC compared to the 6 nm NC. In further analyzing the Fe interstitials, Fe antisites within the NC play a key role in the structural stability of the NC during a cascade. In a pure population sense, with increasing NC diameter, the amount of antisites significantly increases with increasing radii. In taking into account this fact, a better evaluation method is the fraction of NC lattice sites replaced with Fe atoms. This fraction increases significantly with decreasing radii. In comparing the population of defects surrounding the NCs, the 4 nm NC outshines both the 2 nm and 6 nm NCs, with a similar point defect population to the 2 nm NC and significantly smaller fraction of Fe antisites within the NC.

The next issue of interest between different radii is the evolution of the NC structure. Again, all three cases show a certain level of amorphization within the NCs, which increases with increasing PKA energy. With the severity of the amorphization increasing with decreasing radii, along with high amounts of amorphization, the NCs have an increased population of vacancies compared to interstitials. This increase is vacancy population is due to the inclusion of Fe antisites within the NC, easily pushing the O atoms off of their lattice sites. By taking into account the structural changes of the NC, the major difference is in the fraction of amorphization. As discussed in Sec. IV B, an irregular boundary between atoms creates a strong point defect sink. In combining the information about the surrounding Fe atoms and the structural changes in the NCs, the 4 nm NC again outshines the other two diameter NCs, with the 2 nm NC likely disappearing over longer time periods due to Ostwald Ripening, and the 6 nm NC having a smaller concentration of defects surrounding the NC.

Many features go into choosing an ideal NC diameter for ODS steels, with the goals of increasing the irradiation resistance and overall strength of the base material. In this study, the primary damage state of a cascade for NCs at 100 K is shown. Comparing the effect the size of a NC has on the primary damage state of the cascade a few key attributes that are a factor in all three NC diameters in this study: Fe atoms migrating toward the NC over the course of the simulation, the formation of a defect sink at the Fe-NC boundary, and the amorphization of the NC structure. The key trends shown for point defect evolution in the Fe-NC system based on the NC diameter are as follows:

- The ability for the NCs to significantly decrease the remaining population of Fe defects in the matrix is ultimately a factor in the volume of the shockwave compared to the volume of the NC, with the remaining number of defects decreasing with a decreasing in the ratio of shockwave to NC volume.
- The smaller NCs tended to have a higher concentration of Fe defects surrounding the NC, such that with decreasing radii the population of point defects surrounding the NC is optimized
- Amorphization of the NCs is seen in all three NC diameters, with the disorder increasing with decreasing radii. This phenomenon suggests that over longer time periods Ostwald ripening may be a key factor in the dissolution of smaller NCs.

V. CONCLUSIONS

In this study, displacement cascades have been simulated to interact with NCs of various diameters. New results help to further understand how cascade damage affects the evolution of defects in the Fe matrix surrounding NCs, the Fe-NC boundary, and the NC itself.

(1) There are two major pathways for shockwave growth with the inclusion of a NC. At low energies of 1 and 5 keV PKAs, the shockwave occurs at the initial impact site with the NC. However, at higher energies, the cascade energy is able to
penetrate through the NC and cause two smaller shockwaves in the Fe matrix on opposite sides of the NC.

(2) All the NCs experience amorphization via a displacement cascades, where the amorphization increases with PKA energy and the fraction of NC amorphization increases significantly with decreasing radii. Although there is an increase in amorphization of the NCs, the NC atoms remain together during the primary cascade damage.

(3) With decreasing radii, there is a significant increase in the amount of disorder within the NCs, but the 2 nm NC has a higher fraction of disorder compared to the 4 and 6 nm NCs. In combining the disorder with Fe antisites within the NC, the MD results suggest that the tinier NCs are more likely to dissolve via ballistic dissolution.

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REFERENCES


