Kinetics of interstitial defects in α -Fe: the effect from uniaxial stress

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ABSTRACT

Understanding defect kinetics in a stress field is important for multiscale modeling of materials degradation of nuclear materials. By means of molecular dynamics and molecular statics simulations, we calculate formation and migration energies of self-interstitial atoms (SIA) and SIA clusters (up to size of 5 interstitials) in α Fe and identify their stable configurations under uniaxial tensile strains. By applying uniaxial stress along [111], $\langle 111 \rangle$ oriented single SIA defects become more stable than $\langle 110 \rangle$ oriented SIA, which is opposite to stress-free condition. Diffusion of single SIA defects under [111] tensile stress is facilitated along [111] direction and the diffusion becomes one dimensional (1D). For SIA clusters, their diffusion under zero stress has gradual transition from three dimensional (3D) for small clusters to one dimensional (1D) for large clusters. Under the tensile stress along [111], the 3D to 1D transition is accelerated. For large SIA clusters, the stress effect is quickly saturated with less diffusivity enhancement in comparison with small SIA clusters.

Keywords: molecular dynamics simulations, strain, defect clustering, α Fe

1. Introduction

Numerous theoretical studies have been performed to obtain formation/migration energies of defects in alpha Fe by using molecular dynamics (MD) simulations, density function theory (DFT) and kinetic Monte Carlo simulations [1-11]. The defect kinetics are needed for multiple scale modeling of defect evolution in stainless steels for reactor applications. Majority of previous studies were limited to stress-free conditions. A few simulation studies considered the effect of stress on the defect generation and defect clustering in the early stage of displacement cascade formation and evolution [12-14]. A few limited atomic scale simulations were performed to compare the diffusion behaviors under stress [4, 15, 16]. Ab-initio calculations were employed to investigate the stability and mobility of single SIA in alpha Fe under external deformation [17]. Previous studies also considered the changes of energy barrier for impurity diffusion under stress [4, 16, 18].

For all previous studies on stress effects on defect formation and defect migration in alpha Fe, they were limited to single self-interstitial atom (SIA). The lack of knowledge on SIA clusters (containing more than one SIA) has been an issue for multi-scale modeling of defect development under conditions combining neutron damage and stress. One example is creep deformation of fuel cladding when strong fuel and fuel cladding interactions develop under high burnup. Understanding SIA and SIA clusters' behaviors are important to develop multiscale modeling approaches involving the coupling of microstructural changes and mechanical property changes. Small SIA clusters form in damage cascade stage and they are important precursor defects to form large stable defect clusters. In this study, we investigate the formation and migration energies of SIA defects from single SIA to 5-SIAs under uniaxial strain. The present study considers large strain up to 3%, which is not uncommon for strain concentrated region at a cracking tip, which is one important degradation phenomenon for reactor in-core components. Stable defect configurations, defect formation/migration kinetics and diffusion mechanisms are discussed.

2. Methodology

We use LAMMPS [19] code for both molecular dynamics (MD) and molecular statistics (MS) simulations. MD simulations are used to simulate defect migration to extract diffusivity and diffusion energies as a function of temperatures. MS simulations are used to calculate defect formation energies of different defect configurations. The Fe interatomic potential used in the present study is an embedded atomic potential type and was developed by Mendelev *et al.* [20]. The potential was obtained by fitting to both ab initio total energy calculation results and experimentally measured properties. The supercell contains about 512000 Fe atoms and periodic boundary condition is applied. In order to obtain migration

energies, MD simulations are performed at temperatures from 350K to 600K, which are high enough to obtain sufficient defect mobility but low enough to avoid dissociation of defect clusters. Under a given temperature the cell is first relaxed in NVT ensemble, then following by NPH ensemble. A stress is applied to the cell along [111] direction, to maintain a constant strain. After stress loading, a thermal relaxation is allowed in NVT ensemble. The cell stretching along [111] direction leads to shrinkage in both $[1\bar{1}0]$ and the $[11\bar{2}]$ directions through a Poisson's ratio of about 0.29. The time step during simulation is fixed to 0.2 fs. After equilibration to a fixed temperature, a NVE microcanonical ensemble is used to simulate defect migration for a time period up to 40 ns. The visual tool, OVITO [21], is used to record diffusion trajectories.

The migration energies of SIAs can be obtained by both MS and MD methods. For MS approach, the migration energies represent difference of total energy between the initial SIAs configuration and the SIAs at saddle points. Nudged Elastic Band (NEB) method is used to study the kinetics of transition from the initial position to the final position [22-24]. The MD approach is more computational costly but it provides atomic-level details of SIAs' dynamics. The MD approach does not pre-select migration paths, and the migration energies are obtained from temperature dependence of diffusivities. Diffusivities are determined by

$$D_N(T) = \frac{\overline{R^2}(T)}{2n_d t}, \quad (1)$$

where $\overline{R^2}(T)$ is the mean-square displacement of the SIAs, n_d is the dimensionality of the motion (1 to 3), and t is the simulation time. The migration energies are extrapolated from Arrhenius expression,

$$D_N(T) = D_{0,N} e^{-E_m/k_B T}, \quad (3)$$

where k_B is Boltzmann's constant, E_m is the migration energy, and $D_{0,N}$ is a pre-factor. The data analysis follows the independent interval method in order to alleviate issue from limited computational time [3, 8, 9, 25, 26]. In this approach, the total simulation time is divided into K short time segments. For each time interval of $\tau_K (= \frac{t}{K})$, the square displacement $\overline{R_l^2}$ is calculated and the diffusion coefficient is obtained by considering the average of their square,

$$D_N(T) = \frac{1}{K} \sum_{l=1}^K \frac{\overline{R_l^2}}{2n\tau_K}(T) \quad (4)$$

Care must be taken to appropriately select τ_K , as discussed by Terentyev *et al.* [8]. The time interval τ_K must be long enough to include local correlations of defect motion (i.e. allowing diffusion in back and

forth directions for 1-D diffusion), but not too long to significantly increase total computation time. Previous study show that obtained diffusivity decrease with increasing τ_K , and become saturated for longer τ_K . The shortest τ_K to reach such saturation should be used in order to minimize statistic error and maximize computational efficiency [8]. In the present study, we use τ_K of 3.3 to 400 ps depending on the defect sizes and temperatures.

3. Result and discussion

Many previous studies have been performed to understand diffusion mechanisms and obtain defect kinetics of SIAs and SIA clusters in α -Fe and other metals [1, 2, 5, 6, 8-11, 27-33]. All these studies are limited to zero-strain conditions. Table I, II and III compare the structures of small SIAs, formation energies and migration energies from the present study with selected previous works (Table I and II for 0% strain), although the major focus of the present work is for non-zero strain conditions.

Table I shows the structures of small SIAs identified from the MD simulations. The OVITO is used for visualization [21]. The structures of 1-5 SIAs under strain free condition are consistent with previous studies [1, 9, 31]. For 1-SIA and 2-SIA, only $\langle 110 \rangle$ is observed during migration. For 2-SIA, $\langle 110 \rangle$ sessile configuration is observed, which is featured with low diffusivity and high migration energy barrier at low temperatures (less than 500 K). $\langle 110 \rangle$, $\langle 111 \rangle$, and sessile configurations are observed during the migration for 3-SIA and 4-SIA. The 4-SIA sessile configuration is relatively immobile. For 5-SIA, only $\langle 111 \rangle$ is observed. Defect configurations may change at different temperatures and under different simulation times [9]. More detailed descriptions of SIA structures under strain free condition can be founded in previous studies [1, 2, 9, 31]

The main differences between strain free and $[111]$ strained conditions are alinement directions of defects. The 1 to 5 SIAs prefer to be aligned parallel to $[111]$. At 0% strain, the 1-SIA can take any $\langle 110 \rangle$ configuration. However, under $[111]$ strain, only specific $\langle 110 \rangle$ configuration is preferred, which has small angle away from the $[111]$ strain direction. Unlike strain free condition, 1-SIA under the strain prefer to have $\langle 111 \rangle$ configuration aligned with $[111]$ direction. For 2-SIA, $\langle 110 \rangle$ and $\langle 111 \rangle$ configurations are observed. However, under high strain, the sessile configurations are not observed. The absence of sessile configurations leads to high diffusivities. For 3-SIA, $\langle 110 \rangle$, $\langle 111 \rangle$ and sessile configuration are all observed. The sessile configuration is observed under 1% strain at 400 K. The sessile configuration is highly immobile, similar to the 4-SIA sessile configuration at zero strain. For 4 and 5-SIAs, only $\langle 111 \rangle$ configuration is observed under strain. As strain increases, small SIAs prefer to have $\langle 111 \rangle$ rather than $\langle 110 \rangle$ configurations.

Table I. Observed configurations of 1-SIA to 5-SIA under strain free and strains along $[111]$.

	0% strain		Tensile strains along $[111]$: 1 to 3 %	
1-SIA	$\langle 110 \rangle$		$\langle 110 \rangle$
	-	-	$\langle 111 \rangle$
2-SIA	$\langle 110 \rangle$		$\langle 110 \rangle$
	Sessile		$\langle 111 \rangle$
3-SIA	$\langle 110 \rangle$		$\langle 110 \rangle$
	$\langle 111 \rangle$		$\langle 111 \rangle$
	Sessile		Sessile
4-SIA	$\langle 110 \rangle$		-	
	$\langle 111 \rangle$		$\langle 111 \rangle$
	Sessile		-	
5-SIA	$\langle 111 \rangle$		$\langle 111 \rangle$

Table II. Formation energies in eV for 1-SIA to 5-SIA under 0% strain.

Defect size	Config.	Present work	Anento <i>et al.</i> [9]	Terentyev <i>et al.</i> [8]	Marinica <i>et al.</i> [31]	Medeleev <i>et al.</i> [20]	Willaime <i>et al.</i> [32]
1	<110>	3.52	3.57	3.52	3.52	3.52	3.75
	<111>	4.02	3.97	4.01	3.94	4.02	4.45
2	<110>	6.19	6.28	6.23	6.21	6.21	6.68
	<111>	6.73	6.86	6.76	6.74	6.74	7.43
3	<110>	8.85	9.00	8.87	8.84	8.84	9.49
	<111>	9.37	9.39	9.36	-	9.37	10.01
4	<110>	10.92	11.04	11.05	10.94	11.05	11.66
	<111>	11.07	11.18	11.22	11.14	11.36	11.77
5	<110>	13.24	14.12	13.42	-	13.25	14.18
	<111>	13.20	13.47	13.49	-	13.36	13.88

Table III. Migration energies in eV (diffusion prefactors in $10^{-8} \text{m}^2/\text{s}$) for 1-SIA to 5-SIA under 0% strain.

Defect size	Present work	Anento <i>et al.</i> [9]	Terentyev <i>et al.</i> [8]	Soneda <i>et al.</i> [11]	Marinica <i>et al.</i> [31]	J. Marian <i>et al.</i> [4]	Fu <i>et al.</i> [6]
1	0.30(230)	0.27(83.1)	0.31(44.2)	0.167(20.9)	0.31	0.127 (19.4)	0.34
2	0.41(1970)	0.36(351.6) 0.44(4267) for (T ≤ 500 K) 0.19(19.7) for (T > 500 K)	0.33(243)	0.083(7.32)	0.33	0.084 (14.8)	0.42 0.43
3	0.13(11)	0.14(12.1)	0.14(3.35)	0.065(5.57)	0.33-0.38	0.074 (14.2)	0.43
4	0.14(17)	0.15(12.3)	0.16(3.11)	0.100(7.93)	0.05-0.14	0.07*	-
5	0.06(9)	0.05(8.6)	0.007(0.43)	0.066(4.82)	-	0.067*	-

* The migration energy is deduced from their fitting equation.

Table II and III show formation energies and migration energies of 1-SIA to 5-SIA, respectively. Formation energies of 1-SIA to 5-SIA are in good agreements. With increasing defect cluster sizes, formation energies are increasing. In consistent with these previous studies as shown in Table II, the present study shows that $\langle 110 \rangle$ configurations are energetically more favorable than $\langle 111 \rangle$ configurations. For SIAs of size from 1 to 3, the energy difference between $\langle 111 \rangle$ and $\langle 110 \rangle$ is about 0.5 eV. As for the migration energies (as shown in Table III), they are generally decreasing with increasing sizes. Migration energies obtained from Soneda *et al.* and Marian *et al.* have relatively large differences from others, due to different potentials used [4, 11]. In generally, our results are in good agreements with previous studies [8, 9, 31].

Figure 1(a-d) compare the defect formation energies as a function of uniaxial tensile strain along $[111]$ for 1-SIA to 5-SIAs, respectively. Both $\langle 111 \rangle$ and $\langle 110 \rangle$ configurations are considered. All defect formation energies decrease with increasing strain, but the magnitudes of changes are different. $\langle 111 \rangle$ configurations are more sensitive to strain than $\langle 110 \rangle$. Hence, the energy gap between $\langle 111 \rangle$ and $\langle 110 \rangle$ configurations is reduced with increasing strain. As shown in Fig. 1a, the formation energies of $\langle 111 \rangle$ and $\langle 110 \rangle$ 1-SIA under zero strain are 4.02 eV and 3.52 eV, respectively. At 3% strain, these two energies reduce to 3.19 eV and 3.14 eV, respectively. For 2-SIA, $\langle 110 \rangle$ configuration is more favorable at zero strain. With increasing tensile strain, the energy gap between $\langle 110 \rangle$ and $\langle 111 \rangle$ configurations are reducing. At the highest strain of 3%, as shown in Fig. 1b, the formation energy of $\langle 111 \rangle$ 2-SIA is lower than $\langle 110 \rangle$ 2-SIA. For 3-SIA, switching of favorable configurations from $\langle 110 \rangle$ to $\langle 111 \rangle$ occur at 2% strain, as shown in Fig. 1c. For 4-SIA, such change occurs at even smaller strain $< 0.5\%$ (Fig. 1d). For the largest defect 5-SIA, $\langle 110 \rangle$ and $\langle 111 \rangle$ configurations are comparable in formation energies for 0% strain. But adding even small strain makes $\langle 111 \rangle$ configuration more favorable (Fig. 1e). For small defect clusters (1-SIA to 3-SIA), the formation energies of both configurations show almost linear dependence on strain. For large defect cluster such as 5-SIA, formation energies of $\langle 110 \rangle$ configuration deviate from Arrhenius temperature dependence and show an accelerated energy drop at higher strain ($>2\%$). It is worthy of the note that the trend of formation energy changes upon stress can be predicted by the elastic interaction between defects and applied stress fields. However, such estimation is difficult to consider the complexity caused by anisotropic strain relaxation associated with defect clusters. For multiscale modeling, it is necessary to have quantitative kinetics information as shown in Fig. 1.

Fig. 1. Formation energies of $\langle 111 \rangle$ and $\langle 110 \rangle$ SIA defect clusters as a function of strain for cluster containing (a) single SIA, (b) 2 SIAs, (c) 3 SIAs, (d) 4 SIAs, and (e) 5 SIAs.

Fig. 2. Schematics of migration mechanisms for 1-SIA.

Under zero strain condition, there are two possible diffusion mechanisms for SIAs in α -Fe. Figure 2 shows the migration mechanisms of A and B for 1-SIA. In the first mechanism (denoted as mechanism A), $\langle 110 \rangle$ SIA(s) reorient to $\langle 111 \rangle$ and then diffuse along $\langle 111 \rangle$ direction. The migration energy is mainly contributed by the energy barrier required by reorientation [1, 8, 10, 11, 29, 34, 35]. In the second mechanism (denoted as mechanism B) which was first proposed by Johnson [36], $\langle 110 \rangle$ SIA(s) rotate rigidly and translate to other $\langle 110 \rangle$ SIAs on a different plane. Recent DFT and MD studies suggested that mechanism B is more favorable [5, 8, 9, 31]. One of interesting feature of the mechanism A is that 1-SIA can migrate with 1D resulted from the small formation energy difference between $\langle 111 \rangle$ and $\langle 110 \rangle$ configurations [8]. After one jump with the mechanism A, 1-SIA may translate consecutively without returning $\langle 110 \rangle$ configuration due to small energy barrier for 1D migration [1, 10, 27, 34]. The mechanism B, on the other hand, leads to 3D migration [8, 9, 31].

We calculate the energy barriers for the diffusion mechanisms A and B, as shown in Fig. 3a and b, respectively. Energy barriers are calculated by using Molecular Statics (MS) and Nudged Elastic Band (NEB) methods [22-24]. Fig. 3a plots the energy barriers for reorientation of $\langle 110 \rangle$ to $\langle 111 \rangle$ configuration in the mechanism A. Previous DFT calculation showed such reorientation is unlikely due to relatively high energy barrier (~ 0.7 eV) [5, 32]. In consistency, the present study obtains an energy barrier of 0.50 eV for $\langle 110 \rangle$ to $\langle 111 \rangle$ reorientation involved in the mechanism A, and 0.31 eV for $\langle 110 \rangle$ to $\langle 110 \rangle$ migration involved in the mechanism B. In comparison, F. Willaime et al. obtained 0.50 eV and 0.31 eV by using MD simulations, and 0.76 eV and 0.34 eV by using DFT calculations [5, 32]; Terenvey et al. obtained 0.57 eV and 0.34 eV by using MD simulations for mechanisms A (reorientation only) and

B, respectively [8]. All these calculations show that mechanism B is energetically favored (0.31 eV). As the strain along [111] increases, the energy barriers decrease for both mechanisms and the barrier difference between two mechanisms is reduced as well. At 3% strain along [111], the two mechanisms become comparable (0.214 eV vs. 0.184 eV). The energy barrier, 0.184 eV, is higher than the MD result, 0.07 eV (to be shown).

Fig. 3. The energy barriers of 1-SIA for (a) the migration mechanism A and (b) the migration mechanism.

Fig. 4. 1-SIA migration trajectories under different uniaxial tensile strains along $[111]$ direction at 500 K.

Figures 4(a-d) compare single SIA migration trajectories under different tensile strains of 0%, 1%, 2% and 3%, respectively. The z axis is $[111]$ direction, x axis is $[1\bar{1}0]$ direction and y axis is $[11\bar{2}]$ direction. The trajectories represent the movements of the center of mass of single SIA, used to calculate diffusion coefficient. As strain increases, the migration of 1-SIA gradually changes from 3D (Fig. 4 (a)) to 1D (Fig. 4 (d)). At 0% strain, the 1-SIA have only $\langle 110 \rangle$ configuration during the migration, which leads to 3D migration due to diffusion mechanism B. However, at 3% strain, deformation induced asymmetric structural distortion favors certain alignments of 1-SIA. That is, the 1-SIA has different formation energy for different $\langle 110 \rangle$ configurations. It causes $\langle 110 \rangle$ 1-SIA to choose specific $\langle 110 \rangle$ direction, which is aligned parallel to the strain direction $[111]$. Such alignment has been discussed before in simulation of

cascade creation [12]. The 1D migration is due to the preference of parallel alignment of $\langle 110 \rangle$ and $\langle 111 \rangle$ 1-SIA to $[111]$ strain direction. Although it is difficult to tell from Fig. 4, visual inspection at much smaller length scales show that for strain $> 2\%$, $\langle 111 \rangle$ 1-SIA may occasionally change to $\langle 110 \rangle$ 1-SIA. But $\langle 110 \rangle$ 1-SIA quickly changes back to $\langle 111 \rangle$ and re-start 1D diffusion. The time duration to stay as $\langle 110 \rangle$ configuration is much shorter at high strains. At highest strain of 3% the likelihood of $\langle 111 \rangle$ to $\langle 110 \rangle$ transition is further reduced. Diffusion is dominated by forward and backward movements along $\langle 111 \rangle$ axis without returning back to $\langle 110 \rangle$ configuration.

Fig. 5. Migration trajectories of (a) 2-SIA, (b) 3-SIA, (c) 4-SIA, and (d) 5-SIA under 0% strain at 500 K.

For defect clusters (from 2-SIA to 5-SIA), their diffusions under zero strain exhibit a transition from 3D (for small clusters) to 1D (for large clusters). Figure 5(a-d) plot the diffusion trajectories of 2-SIA to 5-

SIA clusters. The x, y, z axes correspond to directions of $[100]$, $[010]$ and $[001]$, respectively. All defect clusters show certain mixture of 3D and 1D diffusion. But their relative contributions to diffusion are sensitive to cluster sizes. For 2-SIA, as shown in Fig. 5a, diffusion favors 3D migration. For 3-SIA, diffusion is still a mixture of 3D and 1D, but the contribution from 3D is decreasing, as evidenced by relatively longer migration distance (or equivalently, longer time duration) for 1-D migration along $[111]$ axis, as shown in Fig. 5b. For larger clusters of 4-SIA and 5-SIA, as shown in Figs. 5c and 5d, 1D diffusion is dominant, featured by line-like diffusion trajectories. Defects may end its 1D migration by changing back to $\langle 110 \rangle$ configuration. But $\langle 110 \rangle$ SIAs immediately rotate into another $\langle 111 \rangle$ configuration and start another $[111]$ 1D diffusion. These motion of small SIAs are consistent with previous works under strain free condition [8, 9].

Fig. 6. Migration trajectories of (a) 2-SIA, (b) 3-SIA, (c) 4-SIA, and (d) 5-SIA under 3% strain at 500 K.

Adding tensile strain promotes 1D migration, and the likelihood to end 1D migration through transition of $\langle 111 \rangle$ to $\langle 110 \rangle$ configurations is significantly reduced. Under high strains (3%), no such configuration changes are observed. Figs. 6(a-d) plot the diffusion trajectories of 2-SIA to 5-SIA under 3% strain. The x, y, z axes correspond to directions of $[1\bar{1}0]$, $[11\bar{2}]$, and $[111]$, respectively. Regardless of cluster sizes, all diffusion trajectories are featured by single straight-lines along $[111]$ axis. The migration direction is matched to the strain direction and it shows fully 1D under 3% strain along $[111]$. Again like 1-SIA, the formation energy differences between $\langle 111 \rangle$ and $\langle 110 \rangle$ of the small SIAs are reduced compared to that of unstrained crystals (0%). Even, the $\langle 111 \rangle$ for small SIAs have the lowest formation energy depending on strains as shown in Fig. 1 (b) to (e). Plus, under 3% strains, all defects are aligned parallel to the strain direction $[111]$. Thus, the small SIAs show 1D fast diffusion to the applied strain direction. These migration behaviors under high $[111]$ strain are similar to large SIAs (sizes more than 7) under zero strain. The fast 1D migration and its relative low migration energies for large SIAs have been observed in previous works [1, 4, 8, 9, 11, 28].

Figure 7(a-e) summarizes the diffusivities extracted from defect clusters of different sizes, from 1-SIA to 5-SIA, and under different strains, from 0% to 3%. The effects of strain on motilities are significant, particularly for small clusters. As shown in Fig. 7a, for 1 SIA, diffusivities increase with increasing strain. At 350K (corresponding to $1/kT=33.17 \text{ eV}^{-1}$), 3% strain enhances the diffusivity by more than two orders of magnitudes. The enhancements are less at higher temperatures. The temperature dependence shows an Arrhenius relation for a thermally activated process. The “apparent” activation energies are the largest for 0% strain, and are reduced with increasing strain, as reflected by slope changes of lines. We use “apparent” to emphasize that even diffusivities follow an Arrhenius expression governed by seemingly a single activation energy, the diffusion mechanisms behind can be a mixture of many different mechanisms. At 0% and 1% strain, 1-SIA defect diffuse dominantly through 3D mechanism B. At 2-3% strain, it diffuses through a mixture of mechanism A and B as shown in Figure 3. These diffusion differences are supported by strain-induced formation energy changes (as shown in Fig. 1) and are the fundamental causes of diffusivity changes in both their magnitudes and activation energies.

For defect clusters larger than 1-SIA, the strain-induced diffusivity enhancements are relatively less. Introducing small amount of strain quickly enhances diffusivity to a level comparable to that under large strain. A higher strain results in a higher diffusivity, but the enhancement is less. Hence, the effect of strain is quickly saturated. Diffusivities at the highest strain of 3% are comparable among different cluster sizes. Fundamentally this is due to the fact that relative diffusion contributions of gliding with less migration energies are already significant for larger clusters under 0% strain (as evidenced by 3D to 1D diffusion transition shown in Fig. 3). Under strain, 1D diffusion is further promoted but its additional

contribution to diffusivity enhancement is limited for large defect clusters. For the largest 5-SIA defect cluster, diffusivities under strain are very close to that without strain, since adding small amount of strain quickly lets $\langle 111 \rangle$ configuration become energetically favorable and diffusion is exclusively governed by 1D migration.

Fig. 7. Diffusion coefficients as a function of temperature under different tensile strain along $\langle 111 \rangle$ direction for defect clusters containing (a) 1-SIA, (b) 2- SIAs, (c) 3-SIAs, (d) 4-SIAs, and (e) 5-SIAs.

Table IV. Migration energies and diffusivity prefactors of SIAs defects.

Defect size	D_0 ($\times 10^{-8}$ m ² /s) and E_m (eV) at different tensile strains							
	0%		1%		2%		3%	
	D_0	E_m	D_0	E_m	D_0	E_m	D_0	E_m
1	230	0.30 ± 0.003	383	0.23 ± 0.005	91	0.14 ± 0.001	33	0.07 ± 0.001
2	1970	0.41 ± 0.008	58	0.14 ± 0.003	32	0.08 ± 0.002	28	0.07 ± 0.001
3	11	0.13 ± 0.003	11	0.06 ± 0.002	11	0.05 ± 0.001	11	0.05 ± 0.001
4	17	0.14 ± 0.004	14	0.07 ± 0.001	13	0.05 ± 0.002	12	0.05 ± 0.003
5	9	0.06 ± 0.002	9	0.05 ± 0.002	11	0.05 ± 0.002	8	0.04 ± 0.001

Table IV summarized the extracted migration energies and diffusivity prefactors of SIA defect clusters under different strains. Regardless of strain levels, migration energies of defect clusters in general become smaller with increasing cluster sizes. Under zero strain, diffusion energies change from 0.3 eV for 1-SIA defect to 0.06 eV for 5-SIA defect. Under 3% tensile strain, the energies change from 0.07 eV for 1-SIA defect to 0.04 eV for 5-SIA defect. For the defects of the same size, increasing strain reduces migration energies. But the energy reduction is less for large defect clusters. For example, for 1-SIA defect, migration energy changes from 0.3 eV under zero strain to 0.07 eV under 3% strain. In a comparison, the changes are 0.06 eV to 0.04 eV for 5-SIA defect. For the diffusivity prefactor, it abruptly increases for 2-SIA under strain-free (0%) condition. Similar trend has been reported in previous studies [8, 9]. The primary reason is the involvement of sessile configurations which are relatively immobile than other defect types.

It is worthy to point out that the present study has certain limits. Firstly, although the EAM potential used were able to reproduce qualitatively the DFT results, it may underestimate the formation energy difference between $\langle 110 \rangle$ and $\langle 111 \rangle$, which may impact on the diffusion properties predicted. Secondly, the present study is limited to strain effect along $[111]$ only, since this direction is most effective to influence the stability difference between $\langle 110 \rangle$ and $\langle 111 \rangle$ SIA configurations [12]. Studies on diffusion behaviors under an arbitrary strain direction is beyond the scope of the present study. The effect of other strains could be very different.

The studies are important for multiscale modeling. Particularly for object kinetic Monte Carlo simulations which treat defect clusters as point-like defects. Both diffusivity enhancement and migration energy reduction for large defect clusters suggest that SIA clusters play an important role in defect

clustering process, including agglomeration of small defects to larger ones, recombination of interstitial-type and vacancy-type defect clusters, and defect migration towards defect sinks such as grain boundaries. The dimensionality of SIAs' migration is important to defect clustering process [8]. This anisotropic diffusion causes big differences in microstructural evolution. Thus, the present study provides necessary fundamental understanding towards multiscale modeling.

Conclusion

We investigate the formation and migration energies of 1-SIA and SIA defect cluster under uniaxial [111] strain. Regardless of cluster sizes, both $\langle 111 \rangle$ and $\langle 110 \rangle$ oriented defects have reduced formation energies as a function of strain. Furthermore, $\langle 111 \rangle$ configuration is more sensitive to strain and its defect formation energies are reduced more significantly. For all defect cluster sizes, there are switching of stable configurations from $\langle 110 \rangle$ SIA to $\langle 111 \rangle$ SIA. The strain also reduces the energy barrier for $\langle 110 \rangle$ SIA(s) to rotate into $\langle 111 \rangle$ SIA, which makes 1D migration along [111] more feasible. For 1-SIA, the migration dimension is 3D under zero strain, but change from 3D to 1D gradually with increasing strain. For SIA clusters, even under zero strain, their diffusion exhibit gradual transition from 3D to 1D at large sizes. By applying [111] tensile stress such transition starts at smaller defect sizes.

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References

- [1] G.R.O. B.D. Wirth, D. Maroudas, G.E. Lucas, Energetics of formation and migration of self interstitials and self interstitial clusters in alpha iron, *Journal of Nuclear Materials* 244 (1997) 185-194.
- [2] Y.N. Osetsky, A. Serra, B.N. Singh, S.I. Golubov, Structure and properties of clusters of self-interstitial atoms in fcc copper and bcc iron, *Philosophical Magazine A* 80(9) (2000) 2131-2157.
- [3] Y.N. Osetsky, Atomistic Study of Diffusional Mass Transport in Metals, *Defect and Diffusion Forum* 188-190 (2001) 71-92.
- [4] J. Marian, B.D. Wirth, A. Caro, B. Sadigh, G.R. Odette, J.M. Perlado, T. Diaz de la Rubia, Dynamics of self-interstitial cluster migration in pure α -Fe and Fe-Cu alloys, *Physical Review B* 65(14) (2002).
- [5] C.C. Fu, F. Willaime, P. Ordejon, Stability and mobility of mono- and di-interstitials in alpha-Fe, *Physical review letters* 92(17) (2004) 175503.
- [6] C.-C. Fu, J.D. Torre, F. Willaime, J.-L. Bocquet, A. Barbu, Multiscale modelling of defect kinetics in irradiated iron, *Nature Materials* 4(1) (2004) 68-74.
- [7] K. Kusunoki, Molecular-Dynamics Study of Self-Interstitial Diffusion in bcc-Iron, *Materials Transactions* 47(8) (2006) 1906-1909.
- [8] D.A. Terentyev, L. Malerba, M. Hou, Dimensionality of interstitial cluster motion in bcc-Fe, *Physical Review B* 75(10) (2007).
- [9] N. Anento, A. Serra, Y.N. Osetsky, Atomistic study of multimechanism diffusion by self-interstitial defects in α -Fe, *Modelling and Simulation in Materials Science and Engineering* 18(2) (2010) 025008.
- [10] N. Soneda, T.D. de la Rubia, Defect production, annealing kinetics and damage evolution in α -Fe: An atomic-scale computer simulation, *Philosophical Magazine A* 78(5) (1998) 995-1019.
- [11] N. Soneda, T. Diaz de La Rubia, Migration kinetics of the self-interstitial atom and its clusters in bcc Fe, *Philosophical Magazine A* 81(2) (2001) 331-343.
- [12] D.J.B. F. Gao, P.E.J. Flewitt, T.A. Lewis, The influence of strain on defect generation by displacement cascades in α iron, *Nuclear Instruments and Methods in Physics Research B* 180 (2001) 187-193.
- [13] S. Di, Z. Yao, M.R. Daymond, F. Gao, Molecular dynamics simulations of irradiation cascades in alpha-zirconium under macroscopic strain, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 303 (2013) 95-99.
- [14] B. Beeler, M. Asta, P. Hosemann, N. Grønbech-Jensen, Effects of applied strain on radiation damage generation in body-centered cubic iron, *Journal of Nuclear Materials* 459 (2015) 159-165.
- [15] W.-L. Chan, R.S. Averback, Y. Ashkenazy, Anisotropic diffusion of point defects in metals under a biaxial stress field simulation and theory, *Journal of Applied Physics* 104(2) (2008) 023502.

- [16] D.S. Tchitchekova, J. Morthomas, F. Ribeiro, R. Ducher, M. Perez, A novel method for calculating the energy barriers for carbon diffusion in ferrite under heterogeneous stress, *The Journal of chemical physics* 141(3) (2014) 034118.
- [17] Z. Chen, N. Kioussis, N. Ghoniem, D. Seif, Strain-field effects on the formation and migration energies of self interstitials in α -Fe from first principles, *Physical Review B* 81(9) (2010).
- [18] X. Li, C. Gao, X.L. Xiong, Y. Bai, Y.J. Su, Hydrogen diffusion in α -Fe under an applied 3-axis strain: A quantum manifestation, *International Journal of Hydrogen Energy* 40(32) (2015) 10340-10345.
- [19] S. Plimpton, Fast Parallel Algorithms for Short-Range Molecular Dynamics, *Journal of Computational Physics* 117 (1995) 1-19.
- [20] M.I. Mendelev, S. Han, D.J. Srolovitz, G.J. Ackland, D.Y. Sun, M. Asta, Development of new interatomic potentials appropriate for crystalline and liquid iron, *Philosophical Magazine* 83(35) (2003) 3977-3994.
- [21] A. Stukowski, Visualization and analysis of atomistic simulation data with OVITO—the Open Visualization Tool, *Modelling and Simulation in Materials Science and Engineering* 18(1) (2010) 015012.
- [22] B.P.U. Graeme Henkelman, Hannes Jonsson, A climbing image nudged elastic band method for finding saddle points and minimum energy paths, *Journal of chemical physics* 113 (2000) 9901-9904.
- [23] H.J. Graeme Henkelman, Improved tangent estimate in the nudged elastic band method for finding minimum energy paths and saddle points, *Journal of chemical physics* 113 (2000) 9978-9985.
- [24] A. Nakano, A space-time-ensemble parallel nudged elastic band algorithm for molecular kinetics simulation, *Computer Physics Communications* 178(4) (2008) 280-289.
- [25] M.W. Guinan, R.N. Stuart, R.J. Borg, Fully dynamic computer simulation of self-interstitial diffusion in tungsten, *Physical Review B* 15(2) (1977) 699-710.
- [26] G.R.O. B.D. Wirth, D. Maroudas, G.E. Lucas, Dislocation loop structure, energy and mobility of self-interstitial atom clusters in bcc iron, *Journal of Nuclear Materials* 276 (2000) 33-40.
- [27] M.V. Yu.N. Osetsky, A. Serra, S.I. Golubov, V. Priego, Computer simulation of vacancy and interstitial clusters in bcc and fcc metals, *Journal of Nuclear Materials* 251 (1997) 34-48.
- [28] Y.N. Osetsky, D.J. Bacon, A. Serra, B.N. Singh, S.I. Golubov, One-dimensional atomic transport by clusters of self-interstitial atoms in iron and copper, *Philosophical Magazine* 83(1) (2003) 61-91.
- [29] N. Soneda, S. Ishino, A. Takahashi, K. Dohi, Modeling the microstructural evolution in bcc-Fe during irradiation using kinetic Monte Carlo computer simulation, *Journal of Nuclear Materials* 323(2-3) (2003) 169-180.
- [30] G.J. Ackland, M.I. Mendelev, D.J. Srolovitz, S. Han, A.V. Barashev, Development of an interatomic potential for phosphorus impurities in α -iron, *Journal of Physics: Condensed Matter* 16(27) (2004) S2629-S2642.

- [31] MArinica, (2011).
- [32] F. Willaime, C.C. Fu, M.C. Marinica, J. Dalla Torre, Stability and mobility of self-interstitials and small interstitial clusters in α -iron: ab initio and empirical potential calculations, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 228(1-4) (2005) 92-99.
- [33] R.C. Pasianot, V.P. Ramunni, Small interstitials clusters migration in bcc metals: A Molybdenum model, Computational Materials Science 48(4) (2010) 783-789.
- [34] R.E.S.G.R.O.B.D. Wirth, Primary damage formation in bcc iron, Journal of Nuclear Materials 251 (1997) 49-60.
- [35] L. Malerba, Molecular dynamics simulation of displacement cascades in α -Fe: A critical review, Journal of Nuclear Materials 351(1-3) (2006) 28-38.
- [36] R.A. Johnson, Interstitials and Vacancies in α Iron, Physical Review 134(5A) (1964) A1329-A1336.