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ATOMISTIC AND SRIM APPROACH TO THE PROPERTIES OF POINT DEFECTS IN SINGLE CRYSTALLINE Mo

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Abstract

Molecular Statics (MS) simulations were performed to obtain information about the stability of point defects in single crystalline Molybdenum (Mo). Different interatomic potentials were utilized to determine the formation energies of interstitials and vacancy. Monte Carlo transport code stopping and range of ions in matter (SRIM) was used to understand the non ionizing energy loss (NIEL) phenomenon and number of Frenkel's pair induced with a dose of $5.29 \times 10^{16} \text{ions/cm}^2$ of He ions carrying energy of 300keV. Displacement per atom (dpa) and ionization of the specimen are investigated against depth. SRIM predicted a fall in NIEL, number of defects and ionization as the impinging ions penetrate deep into the sample. This study predicts the nature of Mo in a harsh radiation environment.

1. Introduction

Single crystalline Mo is a refractory metal which can be used as a structural material in PWR and BWR kinds of reactors due to its fascinating thermal, structural and mechanical features [1-5]. The quantification of the irradiation induced defects is of critical importance from the semiconductor to nuclear science. Kinchin Pease did the modelling of these induced damages such as displacement per atom (dpa) by taking into account the kinetic energy of the incident beam of ions [6]. When fission products are incident upon the reactor structural components it deteriorates their macroscopic properties and finally lead to Fukushima and Chernobyl like dreadful incidents. When a particle with threshold incident energy hit atoms it displaces the atom from its original position, leaving a vacant site. The recoiled atoms if received sufficient energy it can generate cascade collisions. Every knock on atom is responsible for the creation of a vacancy and come to rest as a self interstitial defect. The number of interstitials generation should be equal to the number of created vacancies and they are collectively known as Frenkel's pairs. The amount of energy required to generate the Frenkel's pair is the algebraic sum of the formation energy of interstitial and vacancy, i.e.,

$$E_f = E_i + E_v, \quad (1)$$

where E_i and E_v represent the formation energy for interstitial and vacancy. Several examinations on defect studies on bcc metals and their alloys have been performed [7-13]. In this manuscript we used five different interatomic potentials developed Mo. Updated values of formation energies acquired with different interatomic potentials were reported. SRIM calculations were utilized to study Frenkel defects, NIEL, ionization, implanted ions percent concentration, and displacement per atoms (dpa).

2. Methods and Interatomic Potentials

The MS simulations were carried out by High Performance Computing facility at the college of nuclear science and technology, Harbin engineering university, Harbin, China using large-scale atomic/molecular massively parallel simulator (LAMMPS) code. We used five distinct interatomic potentials of Mo to govern particle to particle interactions. The Zhou et al. [14], Starikov et al. [15], Ackland Thetford [16], Park et al. [17] and Chen et al. [18] potentials are design for pure Mo and can yield almost similar results to experimental work when tested at a specific temperature and pressure. The Starikov potential was selected specifically because of its close approximation of structural and thermodynamical properties. The Zhou potential was selected and it is an improved fitting for Mo's in order to better model the dislocation analysis and tensile stress of the said element. The Ackland's potential is the remedied version of the Finnis Sinclair potential. The Park potential is better to generate the values of defect formation, thermal expansion, surface and stacking fault energies and ideal shear strength in approximation to DFT calculations. SNAP is a machine learning potential and is also working in close agreement to the DFT values obtained for elastic constants, melting point, phonon spectra, surface energies, grain boundary energies, etc.

We created in each case a supercell (10 × 10 × 10) with 2001 atoms of Mo and a structural relaxation was performed. The lattice constant for Mo was set to 3.18Å at 0K. Periodic boundary conditions were applied in all the three axes. All the calculations were made at a temperature of 800K. The interstices in our examination are the <111> dumbbell, <110> dumbbell, and <111> crowdian configurations. A dumbbell and crowdian configurations are constructed at one of these sites by adding a Mo interstitial atom, i.e., 3 systems each with one Mo self interstitial.

3. Results and Discussions

In Figure 1, the positions specified with green spheres are representing the interstitial positions while in the red spheres showing the atomic positions of Mo. There are 32 possible interstitial positions in a bcc Mo system.

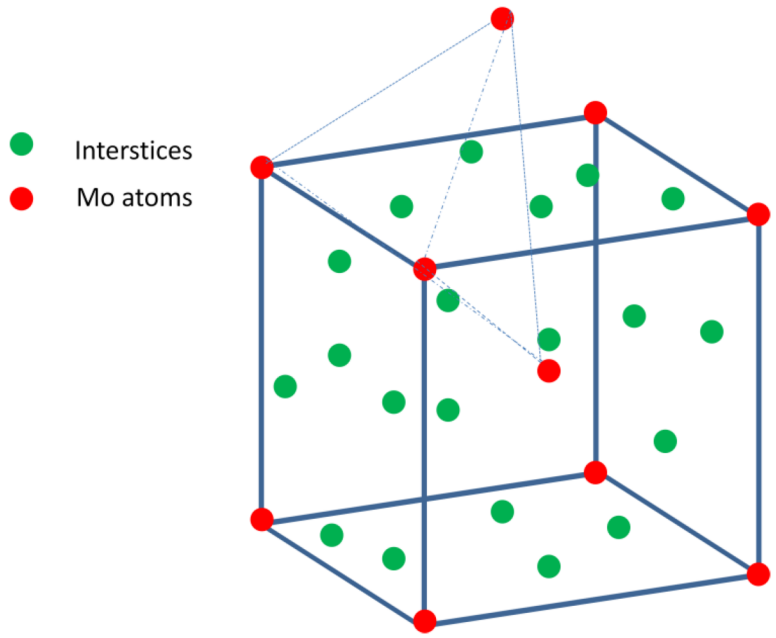

Figure 1. Sketch of a unit bcc Mo cell with 32 interstitial sites.

We assumed [0, 0, 0] as the initial positions of the interstitials and then moved at a distance of 0.245Å for [110] dumbbell along the x and y axes, 0.291Å in 3D for [111] dumbbell, and 0.249Å in 3D for [111] crowdian and in each case at the final positions, we calculated their formation energies. The interstitial energy for the creation of an interstitials can be empirically determined by using the following relation:

$$E_I^f = E_{tot}(N+1) - \frac{N+1}{N} E_{tot}(N), \tag{2}$$

where E_I^f represents the interstitial formation energy and $E_{tot}(N+1)$ and $E_{tot}(N)$ represent the energy of the system with and without interstitial, respectively [19, 20]. The amount of energy needed to create vacancy in a system is given by

$$E_v^f = E_{tot}(N-1) - \frac{N-1}{N} E_{tot}(N), \tag{3}$$

where E_v^f is representing the final energy for the creation of a vacancy [19, 20]. Table 1 tabulates the values of formation energies obtained using different interatomic potentials developed for Mo.

Table 1. Formation energies of different possible interstitial configurations of Mo

Potentials	Authors	Dumbbell [111]	Dumbbell [110]	Crowdian [111]	Vacancy
EAM	Zhou et al. [14]	7.63	6.85	7.63	2.97
MEAM	Park et al. [17]	7.93	7.96	7.91	2.89
SNAP	Chen Chi et al. [18]	8.21	8.35	8.20	2.55
EAM2	Ackland Thetford [16]	6.78	6.59	6.7	2.58
ADP	Starikov et al. [15]	7.67	7.68	7.60	3.12

We used SRIM [21] based on Kinchin Pease model to study the damage effects and implanted ions concentration at a specific energy of 300keV energy with a given dose of $5.29\times10^{16}\text{ions/cm}^2$ of He ions. In order to take into account the local defects, we calculated the displacement rate as a function of depth. The empirical formula for dpa estimation is given by

$$dpa = \text{Fluence} \times \frac{N_{\text{vac}}^{\max}}{a_t}, \tag{4}$$

where $N_{\text{vac}}^{\max}$ represent the vacancy concentration obtained from the sum of vacancies due ions and recoils generated in the vacancy.txt file [22], while a_t is the atomic density which in this case is $6.45\text{E}+22\,\text{atoms/cm}^3$. The threshold displacement energy used in this calculation was 31.7eV [23].

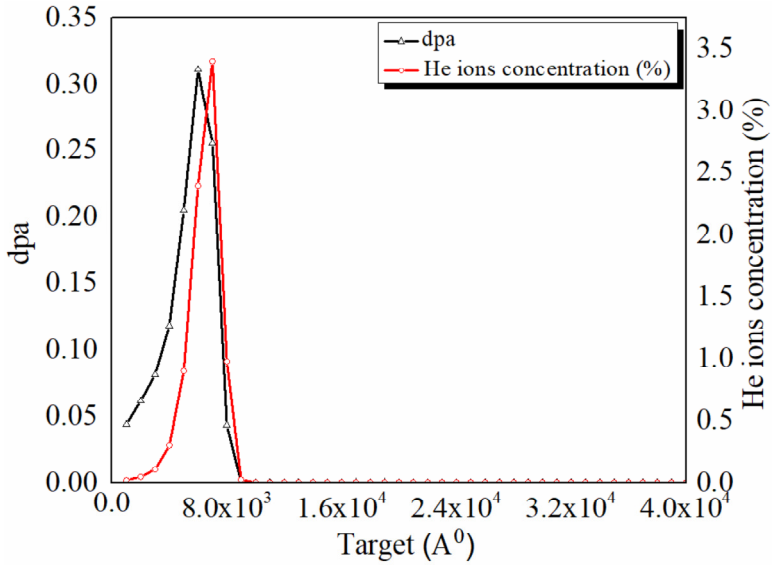

Figure 2. Dpa and implanted ions concentration against penetration depth of ions.

The number of displacements per atom (dpa) is actually the accumulated density of Frenkel pairs. A maximum of 0.31 dpa is attained for Mo which concurred that 31% atoms have been displaced with this fluence when the incident energy of He ions is 300keV. If the value reaches to exactly one for dpa then it will imply that almost all the atoms in the target targeted by ions are displaced [24]. The ions concentration embedded within the matrix of the Mo is 31% too while the rest of ions are either reflected back or having no capability to inoculate into the matter. From the Figure 2, it can be seen that the mean projected range of ions deposition and local damages is close to 10000Å which is the thickness of the specimens set during simulation and imply that most of the ions have penetrated and moved out at the opposite side. However, the local displacement induced by the incident ions is reasonable therefore estimation of the Frenkel's pair is necessary.

A Frenkel defect is a pair of vacancy and interstitial defect. This is caused when an ion moves into an interstitial site and creates a vacancy. Mathematically, the number of Frenkel pairs (N_F) can be obtained by using the following relation:

$$N_F(\mathrm{cm}^{-3})(D) = [IONV(D) + RECV(D)] \times \frac{10^8 \times \text{fluence}}{D_{\max}}, \tag{5}$$

where $IONV(D)$ and $RECV(D)$ are obtained from the VACANCY.TXT file and $D_{\max}$ is the maximum depth. Another important event during damage with ions bombardment on metals is NIEL. The NIEL takes into account the energy which has been lost in non ionizing events. The NIEL is a function of threshold energy (T_d) and atomic density (ρ) [25] is given by mathematically

$$NIEL = \frac{M[IONV(D) + RECV(D)]10^5}{\rho}, \tag{6}$$

where M is given by

$$M = \frac{1}{1000}\left(\frac{T_d}{0.4} + 2\right),$$

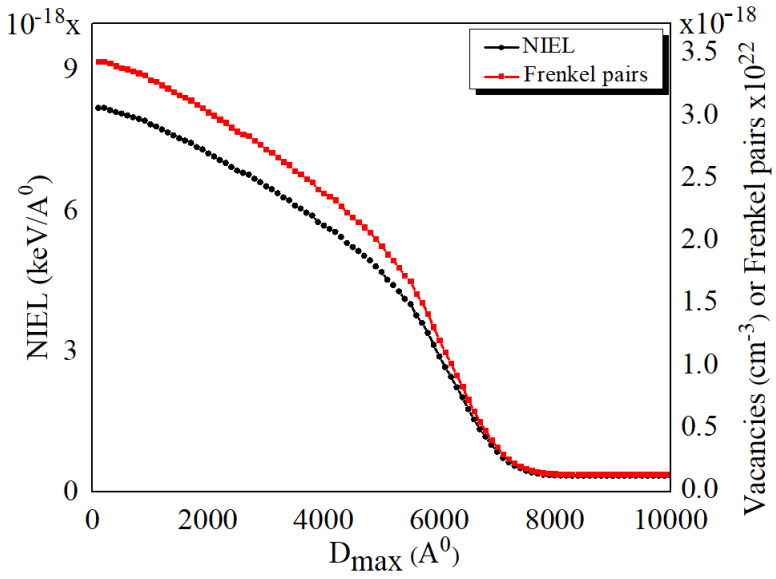

Figure 3. NF and NIEL vs depth for single crystalline Mo.

The plot shows that both the NIEL and NF are following the same trend and showing decrease in their values as the ions are penetrating deep into the matrix. The reason for this decrease is the loss in the impinging energy.

As the ions traversed through the material it ionized the atoms by imparting them kinetic energy and remove their outermost electrons or well shielded electrons as well if received enough energy. Using SRIM, we calculated the ionization of single crystalline Mo using fluence of 5.29×10^{16} ions/cm^2 of He ions with 300keV of energy.

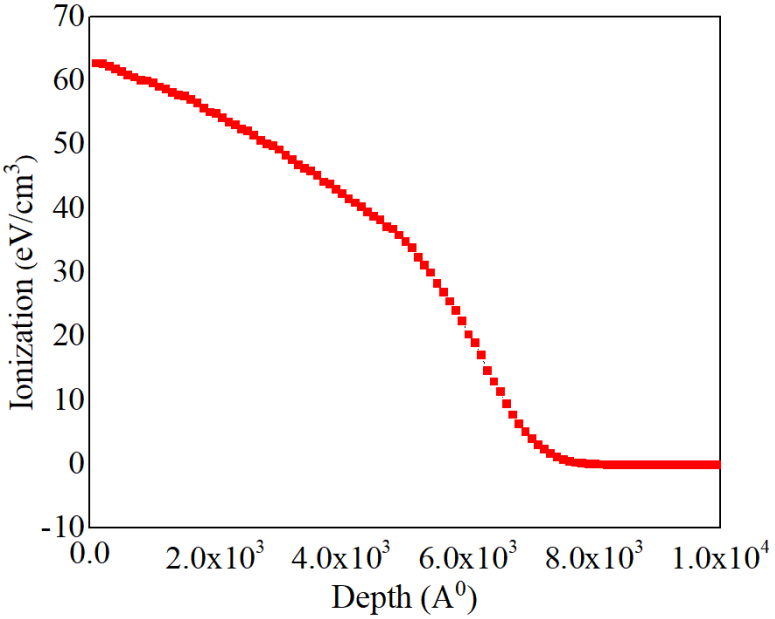

Figure 4. SRIM estimations for ionization in $\mathrm{eV/cm^3}$.

Figure 4 concluded that ionization of the specimen is decreasing with increasing depth. It means that the beam of ions along its trajectory is imparting its energy to the matrix, atom and lattice of the material and losing its potential to knock out atoms from their respective positions.

4. Conclusion

In this work, we performed MS and SRIM simulations in order to model defects behaviours for single crystalline Mo. Different interatomic potentials were utilized previously developed for Mo-Mo interaction. We reported the updated values of vacancy and interstitial formation energies at 300K. Using a dose of $5.29\times10^{16}\mathrm{ions/cm^2}$ of He ions with 300keV of energy caused almost 31% of dpa and 31% deposition of ions into the matrix took place. NIEL, ionization and N_F were calculated against the depth of the sample and found a fall in these parameters against depth.

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