Radiation damage study on graphite from multiple cascade events in molecular dynamics simulation

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Abstract. Graphite is one kind of materials commonly used in nuclear reactor as neutron moderator and structural material. During the nuclear reactor operation, graphite is bombarded with high dose of radiation, especially neutron. This condition can cause damage to the graphite’s crystal structure which then leads to the change of the graphite’s physical properties. The alteration of graphite properties is undesirable since it affects the operation and limits the lifetime of nuclear reactor. In this research, radiation-induced damages on graphite were investigated using molecular dynamic simulation. Reactive force-field potential was employed as the interatomic potential in a simulation box containing about five hundred thousand carbon atoms. The first cascade was initiated using primary knock-on atom having kinetic energy of 1 keV. Following the first cascade, multiple cascades were performed in the same simulation box to represent the long-term neutron irradiation on graphite and to observe the effect of this cascade repetition on defect production. The cascade simulation was performed at two variation of temperature. The number of defects was identified using coordination number analysis. It was found that the effect of temperature to the number of defect produced is not significant, while the repetition of cascade does increase the number of defects produced at the end of the cascade simulation.

1. Introduction

The use of graphite in nuclear reactor as neutron moderator, reflector, and structural material has been initiated since the early years of nuclear reactor utilization in 1950s [1]. Until recently, graphite is still considered as a promising candidate material to be used in next-generation reactor design, such as High Temperature Gas-cooled Reactor [2] and Molten Salt Reactor [3]. However, like other materials used in extreme condition inside the nuclear reactor, graphite structure might be damaged due to neutron radiation. This radiation damage, which can lead to the alteration of graphite’s properties, is undesirable since it can affect the safety and limit the lifetime of nuclear reactor operation [4].

Investigation of neutron radiation effect on graphite has been performed extensively so far. There have been sufficient data available showing radiation damage on graphite in microscopic scale obtained from observation and characterization [5-7]. However, a satisfactory understanding about the detailed mechanism of radiation damage in atomic scale is still deficient [8]. Attaining such understanding will complement the radiation damage studies and provide new insights about how to resolve the damage or even to create new material which is more radiation resistant. This research is
aimed to study the atomic-scale mechanism of neutron-induced damage in graphite through collision cascade phenomena using molecular dynamic simulation.

Collision cascade is a phenomenon of atomic displacement in material’s lattice after being knocked by a projectile, which is neutron in this case. The first atom hit and displaced by the neutron is called Primary Knock-on Atom (PKA). H.J Christie et. al. [9] has performed a single cascade simulation on graphite with several variation of PKA’s energy and initial direction using Environment Dependent Interaction Potential (EDIP). To investigate the accumulation of defect, multiple cascade simulation must be performed overlapping previous cascade area. Some research regarding cascade overlap with pre-existing damage has been performed for metals and alloys. For example, cascade overlap on iron and tungsten [10-14], zirconium [15,16] and nickel [17]. However, the study of cascade overlap in graphite is very limited. Hence, in this work, we performed multiple cascades on graphite by means of molecular dynamic simulation using Reactive Force Field (ReaxFF) potential. The number of defects was analysed at the end of each cascade simulation.

2. Method
Cascade simulation was performed using a well-known molecular dynamics program, Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS), developed by Sandia National Laboratories [18-19]. Graphite structure was modelled using rhombohedral unit cell with lattice parameter of 2.461 Å. This unit cell is then used to create a simulation box with size of 147.6 x 149.4 x 201.5 Å containing around 504,000 carbon atoms with periodic boundary condition in three-dimensional direction. The simulation was performed using NVE ensemble with two outer layers of the simulation box used as thermostat. As the representation of interatomic interaction in graphite, Reactive Force Field (Reaxff) potential [20] was employed.

Cascade simulation was performed five times with two variations of temperature, which is low temperature (300 K) and high temperature (1273 K). This high temperature is a typical temperature of HGTR reactor. The first cascade was performed on the simulation box containing perfect crystal structure of graphite, and the next cascade was performed on the same simulation box using the configuration of the previous cascade simulation. One PKA with energy of 1 keV was initiated in (x, y, z) direction of (0.71, 0.68, 0.22). All simulations were performed using the same PKA energy and direction. The data obtained from MD simulation was visualized and analyzed using OVITO software. The number of defects was calculated based on atomic coordination number. The carbon atoms having coordination number other than three are considered as defect. A region with several uncoordinated atoms can be considered as a vacancy-like defect [8].

3. Results and Discussion
Figure 1(a) and (b) displays the snapshot of the simulation box containing configuration of atoms in a perfect graphite crystal and configuration of atoms after performing lattice equilibration at 300 K, respectively. The equilibrium configuration of the high temperature cascade simulation (1273K) had similar view with the low temperature so that it is not displayed here. This figure shows that the graphite layers had some degree of wrinkles due to thermal vibration of the lattice atoms. Despite the wrinkle, the deviation of carbon atoms from their original position was not significant in this state. It was evident from its radial distribution function which was still peaked at a distance around the lattice parameter of perfect graphite crystal (~1.42 Å).

Figure 2 shows the change of the PKA’s kinetic energy both in low and high temperature cascade simulation. The PKA, which has initial recoil energy of 1 keV, had almost come to rest at simulation time of about 1 ps. It is also shown that the PKA’s energy decreased faster in the low temperature cascade compared to the high temperature cascade simulation. Some modulation of the PKA’s energy shown at some points in the graph, especially at the initial stage of the high temperature cascade, can be originated from the dynamic of the interaction between the PKA and the lattice atoms. Since the PKA had almost come to rest at around 1 ps, it was reasonable to perform the next cascade simulation using the simulation box configuration and parameters at this point of time. This consideration is valid
for the short time interval between cascade performed in this work and it is expected to represent the condition of high neutron flux inside nuclear reactor. Other research regarding cascade overlap sometimes used a longer time interval between each cascade simulation in accordance with their respective research objectives.

![Simulation box containing perfect graphite crystal and equilibrium configuration at 300 K](image1)

**Figure 1** (a) The simulation box containing perfect graphite crystal and (b) equilibrium configuration at 300 K

![PKA’s kinetic energy during the first picosecond of collision cascade event (Cascade 1)](image2)

**Figure 2.** PKA’s kinetic energy during the first picosecond of collision cascade event (Cascade 1)

The analysis of atomic coordination number was performed at the end of each cascade simulation. Table 1 and Table 2 show the coordination number of atoms in low and high temperature cascade simulation, respectively. The atoms having coordination number less than 3 are called under-coordinated and ones having coordination number more than 3 are called over-coordinated atoms. It can be observed that the number of under- and over-coordinated atoms in low and high temperature cascade simulation was similar, except in Cascade 1 of high temperature simulation which had only three under-coordinated atoms. These results showed that the effect of temperature in the number of defects produced was not significant. Moreover, these results showed that the repetition of cascade simulation on the same area does increase the number of under- and over-coordinated atoms. Since the
coordination number is associated with defect, it can be concluded that multiple cascades lead to defects accumulation in the crystal. This conclusion is valid for the short time cascade simulation which excludes the possibility of defect recovery through annealing process or defect evolution during longer period of simulation time [21].

| Table 1. Coordination number of atoms at the end of each low temperature cascade simulation |
|-----------------------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Coordination Number | Cascade 1 | Cascade 2 | Cascade 3 | Cascade 4 | Cascade 5 |
|----------------------|-----------|-----------|-----------|-----------|-----------|
| 0                    | 0         | 0         | 0         | 0         | 0         |
| 1                    | 0         | 0         | 0         | 1         | 1         |
| 2                    | 5         | 8         | 12        | 17        | 23        |
| 4                    | 5         | 10        | 12        | 12        | 14        |
| Total                | 10        | 18        | 24        | 30        | 38        |

| Table 2. Coordination number of atoms at the end of each high temperature cascade simulation |
|-----------------------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Coordination Number | Cascade 1 | Cascade 2 | Cascade 3 | Cascade 4 | Cascade 5 |
|----------------------|-----------|-----------|-----------|-----------|-----------|
| 0                    | 0         | 0         | 0         | 0         | 0         |
| 1                    | 0         | 1         | 0         | 0         | 1         |
| 2                    | 3         | 14        | 15        | 20        | 18        |
| 4                    | 0         | 3         | 12        | 9         | 17        |
| Total                | 3         | 18        | 27        | 29        | 36        |

The locations of these under- and over-coordinated atoms were mainly along the trajectory of the PKA. After interacting with the PKA, some over- or under-coordinated atoms remained in that state for a longer time and some of which can be restored and regain a coordination number of 3. To illustrate the location of those atoms, two snapshots of the simulation box from Cascade 5 for both low and high temperature simulation are displayed in Figure 3. Blue and red colour are assigned for under- and over-coordinated atoms, respectively. From the top-view of the simulation box, it can be observed that some under-coordinated atoms gathered at a certain area and this can be considered as a cluster of vacancy-like defect. Using the same way, the collection of some over-coordinated atoms at a certain area can be considered as a cluster of an interstitial-like defects.

Figure 4 shows the curve of radial distribution function (RDF) of graphite calculated at the end of the last cascade simulation (Cascade 5) for both low and high temperature simulation. The cut-off radius used in calculating the RDF was 1.85 Å [9]. From the RDF curve we can see that the radial distribution function was still peaked around 1.42 Å, which is the in-plane atomic spacing of carbon atom in perfect crystal configuration. At low temperature cascade simulation, the peak of the RDF curve was sharper than the peak of the RDF at high temperature cascade simulation. This means that at high temperature the atomic vibration is more intense than at low temperature. However, at both low and high temperature simulation, we can see that the graphite structure was still in crystalline state even after being irradiated by neutron. This can also be implied that the graphite does not show amorphization process after being irradiated by neutron imparting 1 keV recoiling energy to the PKA.
Figure 3. The location of under-coordinated (blue sphere) and over-coordinated (red sphere) atoms at the end of Cascade 5 in (a) low temperature, and (b) high temperature cascade simulation.
4. Conclusion
Molecular dynamics simulation of multiple cascade event on graphite due to neutron irradiation has been performed using reactive force field (ReaxFF) potential to investigate the effect of the overlapping cascades to the number of defects produced. The cascades were initiated using one PKA having kinetic energy of 1 keV at two variation of temperature condition, which is low temperature (300 K) and high temperature (1273 K) condition. From the results obtained in this simulation, it can be concluded that the effect of temperature to the number of defects produced at the end of cascade simulation is not significant. On the other hand, the repetition of cascade using the same energy and direction of PKA and overlapping the same area in the simulation box does increase the number of defects produced. This implies that the multiple cascades lead to defects accumulation. From the coordination number analysis performed at the end of the last cascade, it was found that several under- and over-coordinated atoms were gathered in a nearby location and this was associated with the cluster of a vacancy-like defect and interstitial-like defects, respectively. The radial distribution function of the graphite calculated at the end of the simulation showed that the graphite was still in crystalline state after being irradiated by neutron with low recoiling energy. The conclusions obtained from this work are limited to the scenario and parameters used in the simulation. To obtain more accurate results with better statistics, it requires simulation with more variations of the cascade scenario, and this might become the topic for further research in the future.

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