Finite Temperature Effect on Structural Changes of Single Crystalline Graphite under Plasma Irradiation

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1 Introduction

Carbon materials are promising candidates for divertor plates in nuclear fusion reactors. Divertor plates of a nuclear fusion reactor are bombarded with hydrogen plasmas. The hydrogen plasmas erode the carbon divertor plates, yielding H_2 and other hydrocarbon molecules such as CH_x and C_2H_x, which are undesirable impurities in plasma confinement experiments. To understand the nature of chemical and physical interactions between hydrogen plasmas and the divertor plates, it is important to clarify the elementary processes of reaction.

Under bombardment of plasmas whose thermal energies are higher than several eV, it is expected that the structures of target materials dynamically changes. For example, when single crystalline graphite is bombarded by hydrogen plasmas, the surface of the graphite becomes hydrogenated amorphous carbon by the chemical and physical reaction of hydrogen atoms. This structural change affects the processes of retention, reflection and sputtering.

In our previous work [1], hydrogen injection into single crystalline graphite was performed by binary-collision-approximation-based (BCA) simulation and a hybrid simulation of BCA and molecular dynamics (MD) simulation. As a result, an interesting phenomenon called "channeling" was observed in the case of crystalline structure. Because of the channeling effect, incident atoms with specific incident angle corresponding to crystalline structure move straightly without causing large angle scattering in the target material. This effect is not negligible in retention process because it makes incident atoms to reach deeper position in the target material. From the simulation, it was also found that the channeling effect gradually disappears because of the amorphousization of the target material. The mean depth of incident atoms in the case where the channeling effect exists may strongly depend on the temperature of the target materials because the vibration of atoms of target material causes dechanneling by hitting and kicking the projectile away from the channeling path.

The time evolution of the bonding states of the target material under plasma irradiation was also investigated in our previous work. In that result, almost 30% of sp content ratio was obtained at the maximum. In our other previous work [2], we formed amorphous carbon in a manner of deposition on a diamond substrate by MD simulation. In the simulation of deposition, sp^2 rich amorphous carbon was obtained. In that case, the percentage of sp carbons is less than 10%. We infer the reason of the difference of sp content ratio in the amorphous carbon materials formed by plasma irradiation and deposition process is that the simulation of plasma irradiation was executed under the condition where the temperature of target material was set to 0 K. It is expected that more sp^2 or sp^3 carbons are generated if the simulation is executed in the case where the thermal vibration exists, because the sp carbons may immediately make bonds with other carbons by annealing process.

In this paper, therefore, to investigate the effect of the finite temperature of the target material, we perform hydrogen injection into a single crystalline graphite which has finite temperature by taking the thermal vibration of atoms on Debye model in BCA simulation and attaching Langevin thermostat to the MD simulation box in BCA-MD hybrid simulation.

2 BCA-MD Hybrid Simulation [3]

To explain the hybrid simulation of BCA and MD more concretely, let's consider a hydrogen atom injection into a graphite material (Fig. 1). An injection kinetic energy of the hydrogen atom is set to 1 keV, which is higher than $E_{\rm th}$. While the kinetic energy of the hydrogen atom is higher than $E_{\rm th}$, trajectories of the hydrogen atom and the surrounding carbon atoms are calculated by BCA simulation. The hydrogen atom dissipates its kinetic energy by interacting with carbon atoms in graphite and then the kinetic energy becomes lower than $E_{\rm th}$. At that moment, MD simulation starts to calculate the motions of the hydrogen and the carbon atoms instead of BCA simulation. Some of carbon atoms which collide with the incident hydrogen are kicked out on the way of BCA simulation. The motions of the kicked carbons in the cascade process are solved by the same procedure as the incident hydrogen.

Unlike BCA simulation, MD simulation cannot treat a large scale material. To save the computation time, cubic simulation boxes (MD simulation boxes) whose side is 34 Å long are picked up for MD simulation. The motion of equation for all atoms in the MD simulation boxes is solved. After each injection, the structural relaxation of all atoms in the MD simulation boxes are executed to minimize their potential energies.

3 Method for Finite Temperature

3.1 Thermal Vibration in BCA Simulation

To investigate the temperature dependence of the channeling effect, the thermal vibration of target atoms in BCA

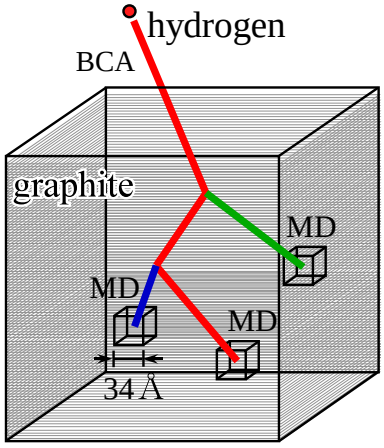

Fig. 1. Schematic diagram of the BCA-MD hybrid simulation of hydrogen injection into a graphite.[3]

simulation is taken into account.

According to the Debye model, the internal energy of a solid in a thermal equilibrium state at the temperature T is written as

$$U = \int_{-\infty}^{\infty} D(\epsilon) \frac{\epsilon}{e^{\epsilon/k_{\mathrm{B}}T} - 1} \mathrm{d}\epsilon, \tag{1}$$

where k_{B} is the Boltzmann constant, ϵ is the energy of phonon, $D(\epsilon)$ is the phonon density of states described as follows:

$$D(\epsilon) = \begin{cases} \frac{3V}{2\pi^2(\hbar c)^3}\epsilon^2, & 0 \le \epsilon \le k_{\mathrm{B}}T_{\mathrm{D}} \\ 0, & \epsilon < 0, k_{\mathrm{B}}T_{\mathrm{D}} < \epsilon \end{cases} \tag{2}$$

where c is the sound velocity in the solid, V is the volume, $\hbar$ is the Planck constant devided by 2π. T_{D} is the Debye temperature described as

$$T_D = \frac{\hbar c}{k_{\mathrm{B}}} \left(\frac{6\pi^2 N}{V} \right)^{1/3}, \tag{3}$$

where N is the number of atoms which constitute the solid. The probability $F(\epsilon)\mathrm{d}\epsilon$ that the average energy per atom $u \equiv U/N$ in a range from ϵ to $\epsilon + \mathrm{d}\epsilon$ is written as

$$F(\epsilon)\mathrm{d}\epsilon = \begin{cases} \frac{3l^3}{2\pi^2(\hbar c)^3} \frac{\epsilon^3}{e^{\epsilon/k_{\mathrm{B}}T}-1}\mathrm{d}\epsilon, & 0 \le \epsilon \le k_{\mathrm{B}}T_{\mathrm{D}} \\ 0, & \epsilon < 0, k_{\mathrm{B}}T_{\mathrm{D}} < \epsilon \end{cases} \tag{4}$$

where $l \equiv (V/N)^{1/3}$ is the mean interatomic distance.

Harmonic oscillation is assumed for the thermal vibration of atoms. In the case of harmonic oscillation with the energy ϵ in classical mechanics, the probability $W(\epsilon, r)\mathrm{d}r$ that an atom is observed in a range from r to $r + \mathrm{d}r$ is described as

$$W(\epsilon, r)\mathrm{d}r = \frac{1}{\pi} \frac{1}{\sqrt{(2\epsilon/k)^2 - r^2}} \mathrm{d}r, \tag{5}$$

where k is the spring constant. The probability distribution function $W(r)$ which is integrated for the energy range from 0 to ∞ is described as follows:

$$W(r) = A \int_0^{k_{\mathrm{B}}T_{\mathrm{D}}} \frac{\epsilon^3}{e^{\epsilon/k_{\mathrm{B}}T} - 1} \frac{1}{\sqrt{(2\epsilon/k)^2 - r^2}} \mathrm{d}\epsilon, \tag{6}$$

where A is the normalization constant.

The spring constant k is obtained by MD simulation. The position of the target atom is shifted by $W(r)$ at each binary collision.

3.2 Thermostat in MD simulation

Langevin thermostat is attached to the MD simulation box to keep the material temperature the preset temperature. Langevin thermostat controls the temperature of atoms by applying the random external force. The equation of motion for the atoms are described as follows:

$$\dot{\boldsymbol{r}}_i = \frac{\partial \mathcal{H}}{\partial \boldsymbol{p}_i}, \quad \dot{\boldsymbol{p}}_i = -\frac{\partial \mathcal{H}}{\partial \boldsymbol{r}_i} - [\gamma \boldsymbol{p}_i + \boldsymbol{R}_i(t)] \tag{7}$$

where, $\boldsymbol{r}_i$ and $\boldsymbol{p}_i$ are the position and the momentum of the i-th atom, respectively. $\mathcal{H}$ is the Hamiltonian of the system. γ is the viscous damping coefficient. The random force $\boldsymbol{R}_i$ is described as the white noise as follows:

$$\langle \boldsymbol{R}_i(t) \rangle = 0, \quad \langle R_{i\alpha}(t_1) R_{i\beta}(t_2) \rangle = 2k_{\mathrm{B}}T\delta_{\alpha\beta}\delta(t_1 - t_2) \tag{8}$$

where, T is the preset temperature, δ is the Dirac delta function, α and β denote the x-, y-, or z-component. The bracket $\langle\ \rangle$ denotes a long-time average.

4 Research Approach

Hydrogen atom injection into single a crystalline graphite material, which has finite temperature by taking the thermal vibration of atoms on Debye model in BCA simulation and attaching Langevin thermostat to the MD simulation box, is performed by BCA-MD hybrid simulation. Hydrogen atoms are injected in succession, so that the structure of the target material gradually changes in time evolution. The temperature dependence of the time evolution of the density of incident atoms in target material is extracted from the simulation results. The temperature dependence of the time evolution of the bonding states of target material is also investigated.

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