§4. First-Principles Molecular Dynamics
Studies of Plasma-Surface Interactions

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Over the last several decades, carbon or carbon-based materials have been extensively used for plasma-facing components in magnetic confinement fusion devices[1]. It is therefore of great importance to have a thorough understanding of processes involved in the interactions (chemical or physical sputterings, etc.) between carbon and the plasma species (mainly hydrogen or isotopes)[1]. However, it is difficult to investigate such phenomena by the experiments because these phenomena are of the atomic level. So we are trying to investigate them by computer simulation.

We are currently studying the hydrogen storage phenomena in graphite by using the first-principles molecular dynamics simulation in the framework of the local density functional theory [2]. We adopt the tight-binding method and the norm-conserving pseudopotentials by Troullier and Martins [3]. The self-consistent electron density is calculated with the Kohn-Sham equation, while atoms are moved by the classical molecular dynamics.

Through adsorption, the bonded hydrogen atom changes the local electronic states of graphite from $sp^2$-like to $sp^3$-like orbitals[4]. Thus, this may break the C-C bonds formed between the hydrogen-adsorbed carbon and other carbon. However, an energy maximum (barrier) occurs at the CH$_3$ molecule when the number of adsorbed hydrogen atoms on one carbon atom is increased. Thus, some special mechanism including physical sputtering is expected to overcome the barrier and help to create the hydrocarbon molecules like CH$_3$ and CH$_4$.

Then, we have simulated the phenomenon of graphite that happens after the saturation state. In these calculations, the graphite in the unit cell consists of 5 layers with each layer containing 24 carbon atoms ($7.41 \times 8.56 \times 31.19 \text{Å}^3$). We do both geometry optimization and molecular dynamics calculations by adding hydrogen atoms successively. The hydrogen atoms penetrate through the graphite surface, modify the internal graphite layers while reducing velocities, and are finally adsorbed to graphite. After having reached the saturated condition, we keep adding the hydrogen atom.

Figure 1 shows the collapse of graphite by the hydrogen adsorption. It is said that the hydrogen adsorption becomes saturated at about 40% [5]. We have shown that after the saturation of hydrogen atoms the hydrocarbon molecules are generated, and that at the same time graphite layers are destroyed at the hydrogen adsorbing sites.

![Graphite Collapse](image)

(a) Initial condition
(b) Relaxed condition

Fig. 1. Appearance of collapse of graphite. (a) initial condition and (b) relaxed condition by the first-principle molecular dynamics. White and black balls denote hydrogen and carbon atoms, respectively.

References