§7. First-principle Calculations for Destructions of Graphite by Hydrogen Adsorption

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The sputtering and erosion of atoms from surfaces by particle bombardment is a significant process in a wide range of material physics and applications, such as micro-fabrications of semiconductors [1] and plasma-wall interaction in magnetic confinement fusion devices [2].

Especially chemical erosion and sputtering processes [3] are important to determine the etching characteristics, including the species of desorbed clusters.

Previously, Koga and Tanaka performed [4] the first-principle calculations of hydrogen adsorption in graphite to determine the energetically most stable configuration of the system that consists of graphite and hydrogen atoms by using the conjugate gradient (CG) method. They employed SIESTA code developed by Spanish atomic physicists [5] where the maximum atomic displacement was limited to 0.1 Å for each step to realize stability of calculations.

In the previous investigation, adsorbed hydrogen atoms broke carbon network in a graphene sheet forming the CH_2 site in the network. The carbon atom in this CH_2 site was lifted up from the initial graphene sheet and its bond structure changed from sp2-like bond to sp3-like bond [6]. Additionally, they reported that two hydrogen atoms were added closely to the hydrogenated graphene sheet easily form a free hydrogen molecule.

In this study, we extend the previous calculation so as to include the spin polarization effect of atoms. Moreover, the central purpose of this study is to find the conditions for desorption of a carbon atom from the carbon network in graphite. Thus, we employ the graphite consisting of five graphene sheets with 84 hydrogen atoms as the initial state (Fig.1). Hydrogen atoms placed in the 1st, 2nd, and 3rd inter-layer spaces are 28 hydrogen atoms for each inter-layer (Fig.1). All of the other conditions are same with those of Ref. [4].

We have observed also the changing of bond structure, from the sp2-like bond to sp3-like bond. Adsorbed hydrogen atom on a carbon atom breaks a π bond between carbon atoms. Then, this carbon atom forms the sp3-bond with 3 other carbon atoms and a hydrogen atom. Also, a big hole in a graphene sheet has emerged in many CH_2 sites. However, free hydrogen atoms between graphene sheets hardly show

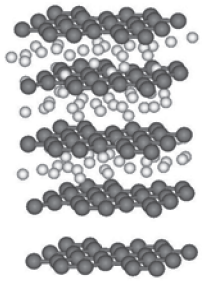

FIG.1 The initial configuretion of the graphite with 84 hydrogen atoms.

the tendency of forming hydrogen molecules (Fig.2) due to the spin polarization effect. Also depending on the spin effect, the evolution of bonding between hydrogen and carbon atoms becomes slower than those shown in Ref.[4].

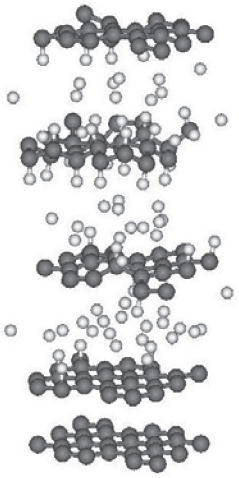

FIG.2 The energetically most stable configuration of the graphite with 84 hydrogen atoms.

By classical molecular dynamics simulations [7], it has been obtained that most frequently observed etching product species has the form C_2H_x and next one is CH_x. Then, we can speculate that adjacent two CH_2 sites is one of the most influential seed of chemically etched byproducts.

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