§22. Destructions of Layered Structure of Graphite by First-principle Calculations

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The sputtering and erosion of atoms from surfaces by particle bombardment is significant process in a wide range of materials physics and applications, such as microfabrications of semiconductor processes or plasma-wall interaction in a magnetic confinement fusion device. Especially chemical erosion/sputtering processes are important to determine etching characteristics, such as species of desorbed clusters.

Previously, the first-principle calculations of hydrogen adsorption in graphite was performed determining the energetically most stable configuration of the system consisted of graphite and hydrogen atoms with the conjugate gradient (CG) method. They employed SIESTA code developed by Spanish atomic physicists.

We have continued our work which extend the previous calculation so as to include the spin polarization effect of atoms. We employ the graphite consisting of five graphene sheets with 108 hydrogen atoms as the initial state. Hydrogen atoms placed at the 1st, 2nd, and 3rd inter-layer space and there are 36 hydrogen atoms for each inter-layer. All of the other conditions are same with those of Ref.4). Figure 1 shows the hydrogenated graphite after energetically stabilization calculation (however, it is not most stable state). It can be easily seen that 2 dimensional structure is collapsed into 3 dimensional, i.e., the bond structure between carbon atoms has been changed from sp2 like to sp3 like.

Figures 2 and 3 show the first and third layer of the graphite shown in Fig.1. First of all, the equivalent existence of hydrogen atoms on both sides of a graphene sheet is important for modifications of the sheet, because the 3rd layer in Fig.3 is clearly well modified rather than the 1st layer in Fig.2. Moreover, we can not find clear evidence that each CH$_2$ site tends to break and leave from a graphene sheet. In Fig.3, one bond between carbon atoms is broken, but all CH$_2$ sites still connecting each other. Then, we speculate that main desorbed products are consisted of not only CH$_2$ type but many of C$_y$H$_x$ (y, x ≠ 0 or 1) type. Indeed, by classical molecular dynamics simulations, it has been obtained that most frequently observed etching product species has the form C$_2$H$_x$ and next one is CH$_x$. Then, we can also speculate that adjacent two CH$_2$ sites is one of the most in actual stage of chemically etched byproducts.

5) A. Gracia et al., The Siesta (Spanish initiative for electronic simulations with thousands of atoms) code, http://www.uam.es/departamentos/ciencias/fisnaturiac/siesta/