

Functional Nano-Particles Simulation

Project Representative

Takashi YOSHIDA
TOSHIBA Corporation

Authors

Takashi YOSHIDA^{*1}, Satoshi ITOH^{*1}, Fumihiko AIGA^{*1}, Misako IWASAWA^{*2}

* 1 TOSHIBA Corporation

* 2 Japan Agency for Marine-Earth Science and Technology

Abstract

It has been known that Carbon-alloy materials have activity of catalysis for oxygen reduction reaction (ORR). This material calls "Carbon-alloy Catalyst (CAC)". The CACs are obtained from pyrolysis of metal-porphyrin and/or metal-phtalocianin with phenol-resin. Ordinary, CACs are known to have only ORR activity. We have explored possibility of that CAC-materials are able to have oxidation activity for anode of Fuel-cell by using first-principles simulation method. In this work by using Earth Simulator, we attempt to examine a possibility of the ability of having oxidation-catalysis at nanographite surface . This study aimed at an understanding of oxidation process for methanol at zigzag-edge regions of nanographite.

The electronic structure calculation was carried out based on density functional theory with PBE96 formulation for the exchange-correlation energy functional. The simulation uses a model system, which consists of single-layered nanographite-ribbon and a methanol molecule under the three-dimensional periodic boundary condition. And we constructed a situation of methanol adsorption in the zigzag-edge of nanographite. The atomic positions of nanographite-ribbon and reactants (methanol) were determined by the first-principles molecular dynamics method. Actual performance of the present version of PHASE (Ver. 8.00) showed 0.770 TFLOPS using 4 nodes of the Earth Simulator.

In this work using the Earth Simulator, we obtained the following results of understanding a reduction of the Methanol deprotonation reaction:

- 1) In the deprotonation of OH, zigzag-edge of nanographite shows high reaction barrier. This value is 1.68 eV, while its value is 0.9 eV lower on the Pt(111) surface.
- 2) For the substitution of Boron in the place of nanographite-edge, a valence-bond is formed between MeO and nanographite's carbon. Consequently, the final-state is stabilized. However, this state does not contribute in reduction of reaction-barrier for deprotonation.
- 3) For N-substituted nanographite, an increment of O-H bond stretching energy is suppressed.

Keywords: Nano-particle, Fuel Cell, Carbon-alloy catalyst, Nanographite, Oxidation, First-principles, Density functional theory calculation