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Author(s)	Nguyen, Thanh Cuong
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Theoretical study of Pt clusters adsorbed on single wall carbon nanotubes

Nguyen Thanh Cuong (430201)

School of Materials Science,
Japan Advanced Institute of Science and Technology

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Highly dispersed Pt nanoparticles on carbon supports have been received considerable attention in electrocatalysts for fuel cell. We succeeded in establishing a new concept based on fundamental bottom-up (Single-Atom-to-Cluster) approach to synthesize highly dispersed and size-controlled Pt clusters on carbon nanotube supports. The catalytic activity of Pt cluster depends strongly on the peculiar electron states, which are localized near surfaces of the Pt cluster and carbon nanotubes and are related to the geometric and electronic structures of Pt cluster adsorbed on carbon nanotubes. In this thesis, we have studied the adsorption of single Pt atom and Pt_n (n=3, 5, 7) clusters on graphene surface and single wall carbon nanotubes (SWNTs) using theoretical calculations.

Our theoretical calculations demonstrate that bridge-type sites are the most preferred binding sites for the adsorption of Pt atoms on graphene surface and both metallic and semiconducting single wall carbon nanotubes. Comparing the binding energies on the small radius tubes with those on large tube and on graphene surface, we could conclude that the curvature of the carbon nanotube has on obvious effect on the interaction between Pt atom and carbon nanotube. We have observed a substantial hybridization

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in the electron orbitals of the Pt atom and the Pt-adjacent C atoms in the SWNT. The energy calculations also show that the Pt cluster state is found to be more stable than the single Pt atom dispersed state.

The interaction between Pt_n (n=3, 5, 7) clusters and metallic (5, 5) single wall carbon nanotube has been studied detail. The binding energy of Pt clusters varies strongly on the cluster size. The deformation of geometric structure of Pt clusters is driven by $sp^2 \rightarrow sp^3$ transition of Pt-adjacent C-atoms. The density of state (DOS) and Mulliken charge analysis show that the charge transfers from Pt cluster toward the (5, 5) SWNT. The charge transfer amount (per Pt atom) is found to decrease with the size of the adsorbed cluster, qualitatively and quantitatively in agreement with our experimental observation. The theoretical investigation of the adsorption of O_2 on the Pt cluster adsorbed on the SWNT confirms the influence of the charge transfer behavior and the transition in the electronic structure of both Pt clusters and carbon nanotubes on the performance of the catalytic activities of the system.

The results have demonstrated that the geometric and electronic properties of systems consisting of nanotubes and Pt clusters can be controlled by a mixing between the wave functions of the nanotubes and those of Pt clusters. Therefore, we can control over the performance of the catalytic of the system.