Molecular Dynamics Simulations of Catalytic CVD Process of Single-Walled Carbon Nanotubes from Alcohol and from Fullerene

Shigeo Maruyama
Department of Mechanical Engineering, The University of Tokyo

Investigation of the formation mechanism is crucial in order to develop a large scale, high-purity, and chirality-controlled generation of single-walled carbon nanotubes (SWNTs). Recently, we have developed a high-quality synthesis technique of SWNTs at relatively low-temperature by catalytic chemical vapor deposition (CCVD) technique using alcohol as the carbon source [1]. In addition, we have demonstrated the macroscopic CCVD generation of diameter-controlled SWNTs from fullerene C_{60} and C_{70} [2]. Here, the nucleation and growth of SWNTs in CCVD process were studied with molecular dynamics (MD) simulations. The simulation techniques were similar to our previous MD simulations of formation processes of empty fullerene [3, 4], endohedral metallofullerene [5], and SWNTs [6] in laser-oven or arc-discharge conditions.

In experimental CCVD processes, supported or non-supported metal clusters are prepared before the assembly of carbon atoms. In MD simulations, various initial metal clusters between about 0.8 nm to about 1.6 nm were prepared as the floated catalyst. Assuming that the carbon source molecule such as methane and methanol decomposed to solid carbon only at the catalytic metal surface, specially designed van der Waals potential in the simulation prohibited clustering of carbon-carbon atoms, even though hydrogen or oxygen atoms were not explicitly included. Here, the chemical reaction process such as decomposition of hydrocarbon was completely neglected in these simple simulations.

For simulations of CCVD from alcohol molecules, in addition to the metal cluster, completely random vapor mixture of carbon-containing molecules was allocated in 20 nm cubic periodic cell. At earlier stages, all carbon atoms were absorbed in the metal cluster. When saturated, hexagonal carbon networks were observed inside the metal-carbon binary cluster. Depending on the initial Ni cluster size and on temperature, the formation of various nanotube cap structures was demonstrated with the continuous supply of carbon.

The similar simulation starting from a metal cluster and C_{60} molecules resulted the indication of the formation of the cap structure made of a C_{60} molecule after the saturation of metal-carbon binary cluster. The diameter of expected SWNTs from the cap structure was about 0.7 to 1 nm in very good agreement with our experimental results [2].

REFERENCES

[1] S. Maruyama, R. Kojima, Y. Miyauchi, S. Chiashi and M. Kohno, Chem. Phys. Lett, 360 (2002), 229.
[2] S. Maruyama, Y. Miyauchi, T. Edamura, S. Chiashi and Y. Murakami, Chem. Phys. Lett., submitted.
[3] Y. Yamaguchi and S. Maruyama, Chem. Phys. Lett., 286 (1998), 336.
[4] S. Maruyama and Y. Yamaguchi, Chem. Phys. Lett., 286 (1998), 343.
[5] Y. Yamaguchi and S. Maruyama, Euro. Phys. J. D, 9 (1999), 385.
[6] S. Maruyama and Y. Shibuta, Mol. Cryst. Liq. Cryst., 387 (2002), 87.