High-Purity Catalytic CVD Generation of Single-Walled Carbon Nanotubes from Alcohol

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By using ethanol or methanol as the carbon source, we have developed a new simple catalytic chemical vapor deposition (CCVD) technique to generate high-purity single-walled carbon nanotubes (SWNTs) at low temperature [1]. Fe/Co catalysts supported with Y-type zeolite were prepared from the metal-acetate in ethanol following the technique developed at Shinohara group [2]. The low-pressure vapor of ethanol or methanol was introduced to catalysts on a quartz boat inside an electric heater. The blackened sample was analyzed with Raman spectroscopy, transmission electron microscopy (TEM), scanning electron microscopy (SEM), and thermo-gravimetric analyzer (TGA).

High purity SWNTs with negligible amount of amorphous carbon, multi-walled carbon nanotubes, carbon particles or metal particles were obtained at 700 - 800 °C reaction temperature using ethanol as shown in Fig. 1. The Raman spectra showed a sharp G-band, a negligible D-band, and a clear radial breathing mode signal. In comparison with the Ni/Co loaded graphite in the laser-oven technique, the tube diameter was smaller and more widely distributed. With decreasing reaction temperature, the diameter distribution shifted to the thinner side. By using methanol, generation of SWNTs even at 550 °C is demonstrated.

The reason why alcohols are much better carbon sources for SWNTs than hydrocarbons is explained by the role of decomposed OH radicals as follows. Since an OH radical is decomposed on the catalyst surface from an alcohol molecule, it will attack nearby carbon

atoms with a dangling bond to form CO. Then, seeds of amorphous carbon are efficiently removed in its very early stage. Because the preference of SWNTs against other amorphous carbon is driven by this reaction, the high-purity generation at low temperature is possible.

[1] S. Maruyama et al, *Chem. Phys. Lett.* (2002), in press.

[2] K. Mukhopadhyay et al., Jpn. J. Appl. Phys. **37** (1998), L1257.

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Fig. 1 TEM image of as-grown SWNTs from ethanol over Fe/Co supported with zeolite at 800 °C.