CVD Synthesis and High Pressure Treatment of Carbon Nanotubes

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Owing to their new functional properties as electrode materials and hydrogen storage materials, carbon nanotubes attract rapidly growing attention. Concerning the preparation of the carbon nanotubes, one of the most important problems is how to prepare nanotubes with desired form. Another significant problem is how to obtain such nanotubes in a large scale. In order to overcome the above mentioned problems, we have tried to synthesize carbon nanotubes with desired forms by two different ways using mesoporous silica: i) template carbonization method, ii) catalytic CVD (CCVD) method.

Mesoporous silica with mesopores of 2-4 nm and 6-30 nm in diameter were synthesized in the presence of cetyltrimethylammmonium bromide surfactant and triblock copolymer, respectively. Trimethylbenzene was used as an organic swelling agent to enlarge the mesopore. The solution mixture was aged for 24 h at 353-383 K. Calcination was carried out by slowly raising the temperature from room temperature to 873 K.

The loading of catalytic metal nanoparticles was carried out by introducing metal nitrate and/or sulfate aqueous solution in vacuum and subsequently drying it at 393 K for 5h. The catalyst particles used in the present study were Fe, Ni, and Co.

CVD experiments were performed as follows. A mesoporous silica sample was evacuated in a SiO_2 tube at 383 K for 30 min. Then the reactor was filled with nitrogen or argon gas and the temperature of the reactor was raised to 1073 K. Hydrocarbon (methane, propylene, acethylene) and carrier (nitrogen, argon) gases were flowed for 1 h.

The synthesized mesoporous silica and the CVD treated samples were characterized by XRD, N_2 adsorption-desorption isotherm experiments and TEM observation.

The relation between the pore sizes of the synthesized silica templates (Fig. 1) and the forms of the obtained multiwalled carbon nanotubes (MWNT) by template carbonization method was written elsewhere.[1]

In the case of CCVD experiments, both MWNT and single walled carbon nanotubes (SWNT) were synthesized (Fig. 2). The yield of the carbon nanotubes obtained by acethylene gas CVD treatments highly depends on the catalyst materials (Table 1). We have also investigated the yield with propylene gas as a carbon source. It was found that the dependencies of the yields on the catalysts are not noticeably different in the cases of acethylene and propylene gas CVD experiments.

I will also discuss the results of high pressure treatments of carbon nanotubes.

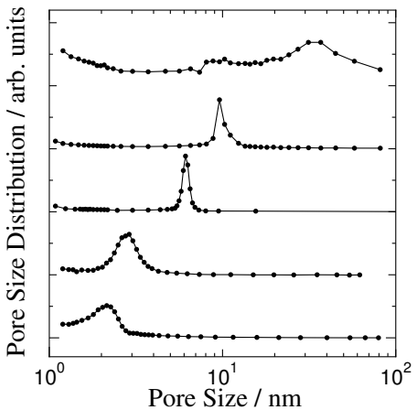

Figure 1: Pore size distribution curves of the synthesized mesoporous silicas.

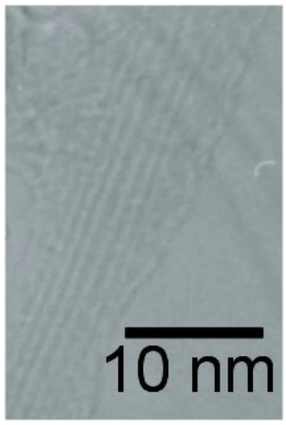

Figure 2: TEM image of the SWNT obtained by the present CCVD method.

Table 1: The relation between the catalyst source materials and the yields of carbon nanotubes obtained by CCVD method using acethylene gas as a carbon source gas.

catalyst source	yield of SWNT	yield of MWNT
$Co(NO_3)_2$	–	low
$CoSO_4$	–	low
$Ni(NO_3)_2$	–	high
$NiSO_4$	–	low
$Fe(NO_3)_2$	–	low
$FeSO_4$	low	very high

REFERENCE

[1] S. Kawasaki, S. Komiyama, S. Ohmori, A. Yao, F. Okino and H. Touhara, MRS Proceedings Volume 675, Nanotubes, Fullerenes, Nanostructured and Disordered Carbon, W3.2.1-W3.2.5, (2001)