SYNTHESIS OF DWNTS BY CATALYTIC CHEMICAL VAPOR DEPOSITION METHOD

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Introduction

Much attention has been focused on synthesis of double-walled carbon nanotubes (DWNTs) because their unique coaxial structure is very interesting for studying wall-wall interaction, and for applying this tubes as field emitter due to their high structural stability as compared with that of single wall carbon nanotubes (SWNTs). Furthermore, selective functionalization of only outer shell will make them useful in the fabrication of multi-functional nanocomposite.

DWNTs have been synthesized by several methods such as arc discharge, the catalytic chemical vapor deposition (CCVD), thermal treatment of peapod. Among them, CCVD method is considered to be very promising for large-scale production of DWNTs. In this study, we synthesized DWNT through the catalytic decomposition of methane over iron particles on magnesium oxide by changing reaction temperatures, and evaluated the quality and also purity of tubes by using high-resolution transmission electron microscope (HRTEM), field-emission scanning electron microscope (FESEM) and Raman techniques in detail.

Experimental

In this synthetic system, we utilized Fe/MgO as nanotube catalyst and Mo/Al_2O_3 as condition catalyst [1]. The preparation of catalyst on magnesium oxide and/or aluminum oxide was carried out through a simple excess solution impregnation as following: (1) dispersion of support materials in distilled water using an ultrasonic process, (2) adding the appropriate concentration (5 wt. %) of ferric ammonium citrate to the solution, and heating at 90^{o}C using the hotplate in order to evaporate the solvent completely during stirring, (3) drying in vacuum for 12 hrs, (4) pulverizing into a fine powder using the mortar.

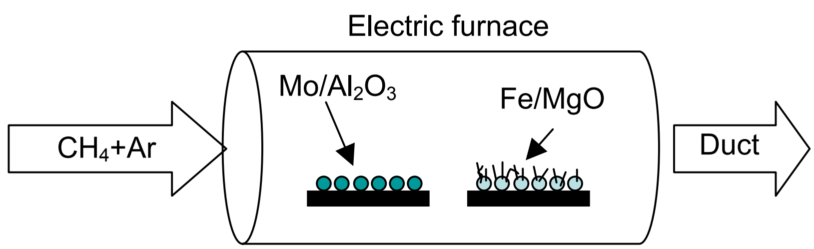

Figure 1. Schematic diagram of catalytic CVD apparatus for the synthesis of DWNTs

The synthesis of DWNTs was carried out using a horizontal quartz reactor. Fe/MgO catalyst was placed at the center of reactor while Mo/Al_2O_3 (conditioning catalyst) was settled in front of main catalyst as shown in Fig. 1. The reactor was heated up to specific reaction temperature (875 – 950^oC) in argon atmosphere. And then carbon feedstock (methane + argon mixture, 1:1) was fed into reactor for 10 minutes.

Results and Discussion

Fig. 2 exhibits HR-TEM images of samples synthesized at different conditions. With increasing temperatures, distinctively tube diameter increases, possibly due to sintering of metal nanoparticles. When utilizing conditional catalyst (Fig. 2 (b)), relatively high portion of DWNTs was obtained. As described in ref. 1, increased portion of active species (benzene) is closely related with the increased yield of DWNTs. We measured the diameter distribution DWNT and SWNT, purity of CNT for samples by using Gatan program. Reaction temperature of 875^oC and utilization of conditional catalyst resulted in narrow distribution, and also high purity of DWNTs. We think this phenomenon is closely related with growth mechanism of nanotubes because basic different point is activity of carbon feedstock for nanotube growth. This study confirmed that active carbon feedstock is important for obtaining high quality carbon nanotubes.

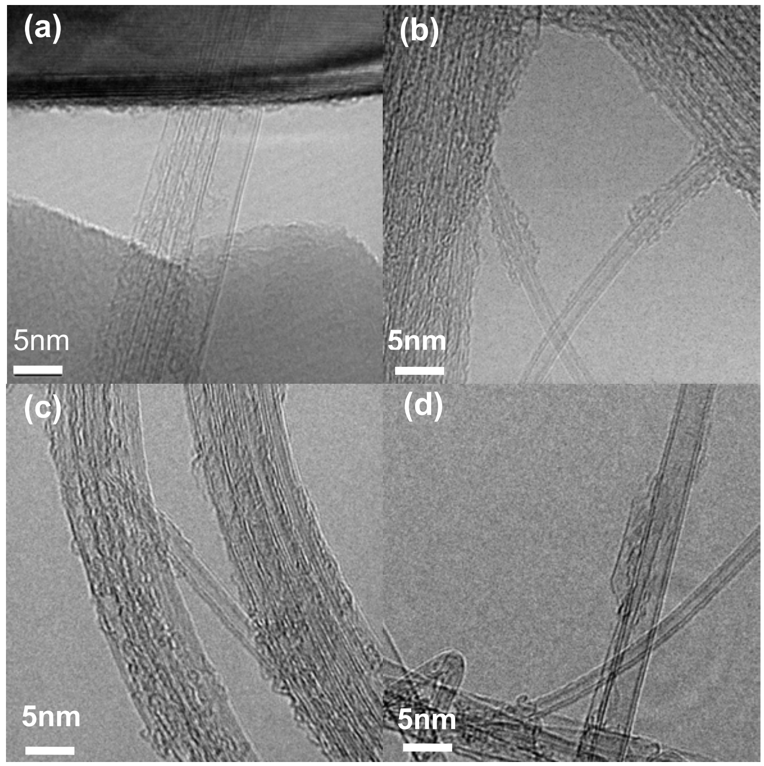

Figure 2. High resolution TEM images of DWNTs synthesized at 875^oC without conditional catalyst (a), at 875^oC with conditional catalyst (b), at 900^oC with conditional catalyst (c) and at 950^oC with conditioning catalyst.

References

[1] N. R. Franklin and H. Dai, Adv. Mater., 12, 890 (2000).

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