Optimizing growth conditions for coaxial multi-walled carbon nanotubes

[Optimizing coaxial CNT growth](brief running head)

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Abstract

Coaxial multi-walled carbon nanotubes have a multi-layered structure in which a core multi-walled carbon nanotube is grown inside a sheath multi-walled nanotube from a metal-catalyst nanoparticle. In this paper we report the optimum conditions to grow coaxial multi-walled carbon nanotubes by chemical vapor deposition. The coaxial nanotubes are studied by means of transmission electron microscopy to reveal their crystallinity and morphology. Our results show that chemical vapor deposition growth at around 1000 °C with a 0.04-0.07 mg/cm^3 dose of palmitic acid yields the best result.

Keywords: carbon nanotube, coaxial nanotube, growth condition

Introduction

Carbon offers novel structures in the nano scale such as nanotubes, fullerenes, and graphene. The physical properties and technological applications of these structures have been extensively studied in many reports (1-3). A strong motivation remains to discover new novel forms of carbon, and utilize their properties. The possibilities seem limitless even in the last decade. Carbon nanofoam exhibits interesting physical properties such as ferromagnetic behavior up to 90 K (4). Carbon nanoribbons have technological potential as waveguides for sub-wavelength photonics (5), field-effector transistors (6, 7), and as infrared photodetectors (8). The new carbon structures have motivated computational and experimental studies of the transport properties (9-12), as well as the optical response in the broad THz-visible spectrum (13-16). Potential technological applications also motivate the study of the formation mechanism of these nanostructures (17-21). In summary, the discovery of new carbon structures opens up new avenues in technological applications and brings a wealth of new scientific and engineering knowledge; therefore it remains important to search for new novel nanostructures and to study their physical properties and formation mechanisms.

We have recently reported the growth of a multi-walled carbon nanotube (MWCNT) inside a larger MWCNT where one Fe nanoparticle acts as the growth catalyst for both the outer MWCNT (sheath) and inner MWCNT (core) (22). Fig. 1 shows a representative coaxial MWCNT. The coaxial MWCNTs were grown via a simplified chemical vapor deposition (CVD) process. The possibility to manipulate the radius and/or number of walls of both inner and outer MWCNTs strongly motivates a careful study of different growth conditions for this nano structure. In this work, we investigated two growth parameters: the growth temperature and the density of the carbon source (palmitic acid). The optimal growth parameters for the CVD growth of the coaxial nanotubes were a 0.04-0.07 mg/cm^3 dose of palmitic acid and a growth temperature of around 1000 °C.

Experimental

A 20 nm thick Fe film was deposited on a SiO_2 plate, and then the substrate was sealed in an evacuated silica tube with a variable amount of palmitic acid. The silica tube had a 6 mm inner diameter and a 25 cm length. The mass of the carbon source varied from 0.3-1.5 mg and corresponds to a carbon source gas density of about 0.04-0.21 mg/cm^3 inside the silica tube. A 0.3 mg amount of palmitic acid was the smallest amount that could be accurately measured. The tube was then annealed for 30 minutes. Different annealing temperatures were investigated in the 900-1100 °C range. The samples were then cooled to room temperature outside of the furnace. The synthesized CNTs were mounted on a carbon microgrid by rubbing the substrate surface with the microgrid, and then observed on a JEOL JEM2010 transmission electron microscope (TEM).

Results and Discussion

First, we studied how the difference in the amount of carbon source affected the quality of the structure of the grown coaxial nanotubes. We synthesized coaxial MWCNTs using 0.3, 0.5, 0.7, 1.0 and 1.5 mg of palmitic acid at a temperature of 1000 °C. Fig. 2 shows that nanotubes with clear sheath-core structure were grown at the lower amounts of palmitic acid: 0.3 mg and 0.5 mg. The results show that the higher amount of palmitic acid, the more graphite walls were formed. Although the nanotubes that were grown using more than 0.7 mg of palmitic acid had a visible bi-layered structure, the nanotubes had so many graphitic walls that their lattice fringes could not be observed. Furthermore, the cores of those nanotubes were filled with graphitic carbon, as shown in (e) and (f), presumably due to a very high degree of supersaturation of the carbon source gas during the growth.

Next, we examined how the growth temperature influenced the CVD growth of the structures. Nanotubes were grown at temperatures of 900, 950, 1000, 1050, and 1100 °C, with a 0.5

mg dose of palmitic acid. As shown in Fig. 3, the structure of the nanotubes grown at 900 or 1100 °C was considerably distorted, while we could find well-organized coaxial nanotubes when grown at 950-1050 °C. Therefore, the results show that the optimal growth temperature is around 1000 °C.

We note that we did not observe a clear dependence of the nanotube diameter on either the growth temperature or the amount of palmitic acid: the growth temperature only affected the crystallinity of the nanotubes, and a high degree of supersaturation resulted exclusively in filling of the core. Therefore, the nanotube diameter, which is the outer diameter of the sheath nanotubes, is considered to be simply determined by the initial size of the metal catalyst nanoparticles.

Conclusion

It has been elucidated that crystallinity and morphology of the coaxial MWCNTs strongly depend on the growth conditions such as the temperature and the amount of carbon source. Growth at a temperature around 1000 °C with a 0.04-0.07 mg/cm^3 dose of palmitic acid yielded the best result in terms of crystallinity and morphology. The growth of the coaxial MWCNTs could be further investigated with different growth parameters such as choice of materials for carbon source and metal catalyst. Future investigations should focus on new growth parameters or techniques that could limit the number of walls. The ongoing effort to control the amount of walls, and improve the homogeneity and structure of MWCNTs, will allow elucidation of the optical and transport properties of coaxial nanotubes grown by chemical vapor deposition.

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Figure captions

Fig. 1: (a) Representative TEM image of a coaxial MWCNT. High-resolution TEM image (b) shows the contrast between the outer and inner MWCNTs.

Fig. 2: (a)-(f) TEM images of the coaxial MWCNTs grown at 1000 °C with different amounts of palmitic acid: (a)(b) 0.3 mg, (c)(d) 0.5 mg, and (e)(f) 0.7 mg.

Fig. 3: TEM images of the coaxial MWCNTs grown with 0.5 mg of palmitic acid at: (a) 900 °C, (b) °C, (c) 1000 °C, (d) 1050 °C, (e) 1100 °C.

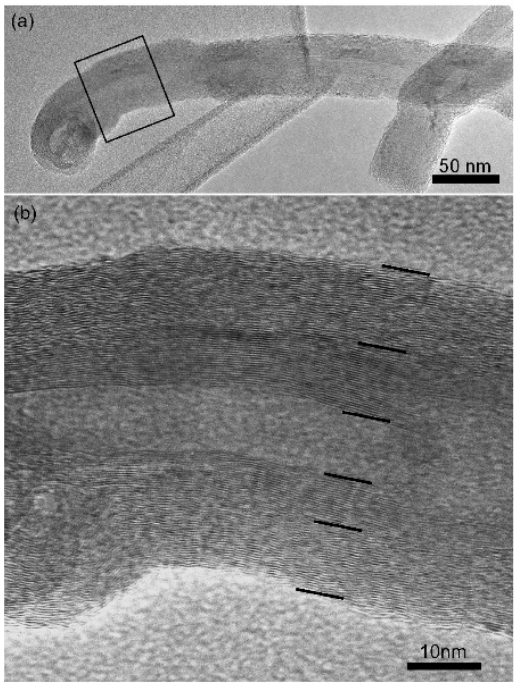

Fig. 1

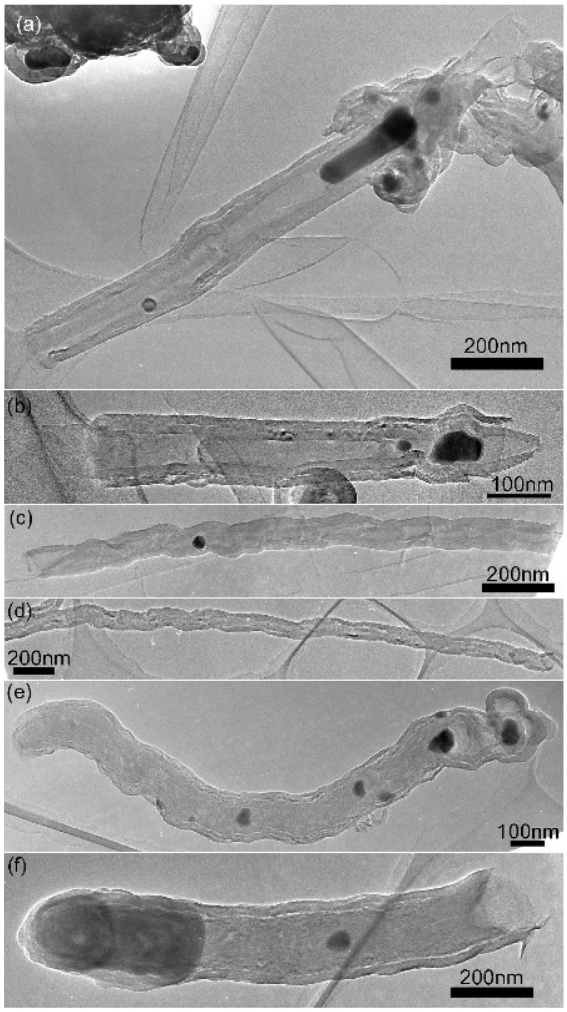

Fig. 2

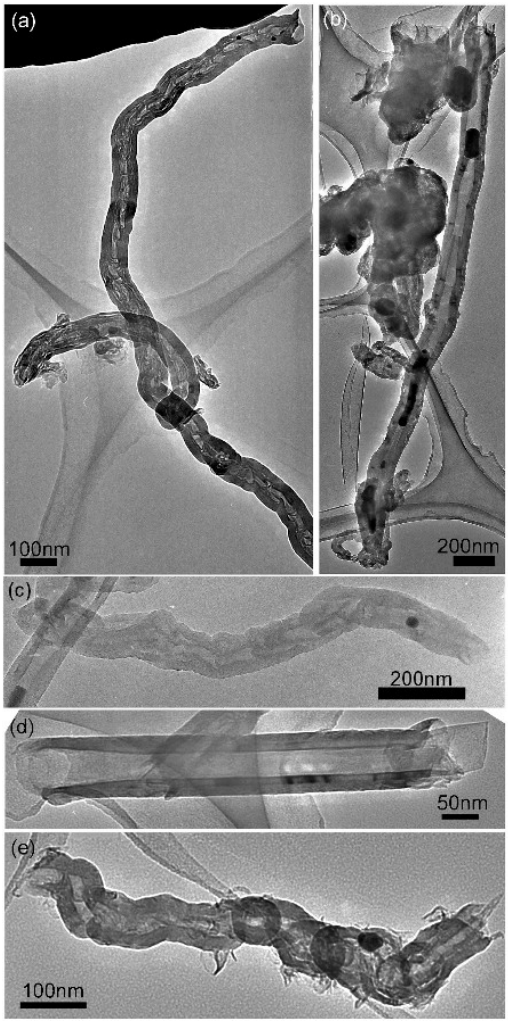

Fig. 3