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SELECTED TOPICS in APPLIED PHYSICS I
New Advances in Carbon Nanotube:
From New Growth Processes to Nanodevices

Parametric Study on Growth of Carbon Nanocoil by Catalytic Chemical Vapor Deposition

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Carbon nanocoils (CNCs) were synthesized by the catalytic pyrolysis of acetylene over a mixture of Fe and SnO_2 particles in a chemical vapor deposition (CVD) apparatus. The effects of reaction temperature, the composition of Fe and SnO_2 in the catalyst, and the ratio of acetylene (C_2H_2) to helium (He) gas were examined. An optimum output was obtained when the Fe/SnO_2 ratio was 2/1, the flow rates of C_2H_2 and He gases were 150 and 700 sccm, respectively, and the reaction temperature was 700°C. Under these conditions, the CNCs were of 80% purity, with average coil diameters of 750 nm, fiber diameters of 300 nm and pitches of approximately 750 nm. The CNC yield weighed up to 60 times that of the catalyst in 10 min.
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KEYWORDS: carbon nanocoil, catalytic CVD, iron–tin–oxide catalyst, helical carbon nanofiber, acetylene

1. Introduction

Carbon coils used to be hindrances that emerged during the catalytic conversion of petroleum and were occasionally found to be by-products of carbon fibers in the catalytic pyrolysis of carbon containing gases.[1–3] In 1991, Motojima *et al.* added a small amount of thiophene into a chemical vapor deposition (CVD) reactor during the pyrolysis of acetylene over Ni catalysts and discovered that sulfur in the thiophene dramatically promoted the growth of helical carbon fibers with coil diameters of several micrometers. They named the helical carbon fiber the carbon microcoil.[4] In 2000, Zhang *et al.* deposited an iron film of 15 nm on indium–tin–oxide (ITO)-coated glass and used it as a catalyst in a CVD process for the direct synthesis of carbon nanotubes (CNTs). Instead of CNTs, carbon nanocoils (CNCs) were formed with high yield. After a detailed analysis, they discovered the following: with iron-on-glass or silicon substrates, aligned CNTs grew perpendicular to the substrate surface; without iron on ITO, indium oxide, tin oxide, glass, or silicon surfaces, only thin amorphous carbon films were formed; on an iron-coated tin oxide surface, only CNTs were observed; and on the indium-oxide coated surface, CNCs were formed with low yield. They concluded that iron was necessary for the growth of tubular carbon, indium oxide contributed to the coil formation, and tin oxide increased the deposition velocity.[5]

Although disputes still exist on the formation mechanism involved in the catalytic synthesis of CNCs,[4,6] it has been established that a catalyst is composed of an iron-group metal which can dissolve carbon or form metal carbide and a second element in the form of either sulfur, phosphorus or another metal.[7–13]

In efforts to find a highly efficient catalyst for the deposition of carbon nanomaterials, we investigated the synergies of metals and metal oxides by simply mixing commercially available powders and found that mixtures of iron and tin oxide were the most efficient combination for CNC synthesis.[14]

In this study, we focus on the optimization of process parameters for CNC synthesis. These parameters include the catalyst composition, the reaction temperature, and the gas composition. The optimum process conditions are elucidated after intensive experimentation.

2. Experimental

The catalytic pyrolysis of acetylene was investigated using a CVD apparatus of a cylindrical quartz tube reactor (300 mm long, 40 mm in inner diameter) described in a previous paper.[10] The temperature was initially raised to the reaction temperature under the flow of helium (He) gas at a rate of 420 sccm. Then C_2H_2 gas flowing of 180 sccm was added to the He flow for 10 min. Finally, the reactor was cooled to room temperature under the flux of He gas.

After the reaction, the carbon deposit was weighed using an analytical balance. For the as-grown carbon deposit mixed with the catalyst, the carbon deposition rate was calculated as follows:

$$\text{Deposition rate of carbon} = \{(W_{tot} - W_{cat})/W_{cat}\}/\text{time},$$

where W_{tot} is the total weight of the catalyst and the carbon deposit, W_{cat} is the weight of the catalyst, and time is the duration of acetylene introduction. Therefore, the deposition rate reveals the carbon yield per unit weight of the catalyst per unit time.

The carbon deposit was observed using a field emission scanning electron microscope (FE-SEM) (S-4500II, Hitachi, Japan) and was evaluated based on its CNC percentage estimated from several visual fields in the FE-SEM micrographs. The coil diameters, the fiber thicknesses and the coil pitches of the CNCs were also measured on the FE-SEM micrographs.

C_2H_2 and He gases flowed through a mixer to the reactor. The partial pressure of the C_2H_2 gas was calculated from its flow rate divided by the total gas flow rate. All reactions were carried out under atmospheric pressure.

The catalyst was prepared by mixing micrometer-order powders of iron and tin oxide; the substrate was simply

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