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Zinc oxide catalyzed growth of single-walled carbon nanotubes

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ABSTRACT

We demonstrate that zinc oxide can catalyze the growth of single-walled carbon nanotubes (SWNTs) with high efficiency by a chemical vapor deposition process. The zinc oxide nanocatalysts, prepared using a diblock copolymer templating method and characterized by atomic force microscopy (AFM), were uniformly spaced over a large deposition area with an average diameter of 1.7 nm and narrow size distribution. Dense and uniform SWNTs films with high quality were obtained by using a zinc oxide catalyst, as characterized by scanning electron microscopy (SEM), Raman spectroscopy, AFM, and high-resolution transmission electron microscopy (HRTEM).

1. Introduction

It is well known that metal nanoparticles (NPs) are indispensable for the growth of single-walled nanotubes (SWNTs) by chemical vapor deposition (CVD). Metal NPs act as the initiating centers and the SWNTs grow out from them via a vapor–liquid–solid (VLS) mechanism. The Fe-family of elements is known to be the most effective group of catalysts. However, in the past three years many other metal NPs such as Au, Ag, Cu, Pd, Rh [1–5], semiconductors NPs such as Si and Ge [6], carbides such as SiC_3 and Fe_3C [7], and more recently Mg, Mn, Cr, Sn, Al, Pb and SiO_2 [8–10] have been reported to be active for the growth of SWNTs. These materials were hitherto regarded as inactive catalysts for the growth of SWNTs in the past. Therefore, these findings challenge the traditional thinking about the growth of SWNTs and the role of the catalysts. Furthermore, different types of catalysts will provide a greater understanding of the relationship between the catalysts and the structures of the SWNTs and thus may find out the approach for selective growth of semiconducting SWNTs (s-SWNTs) or metallic SWNTs (m-SWNTs). Pure s-SWNTs with uniform diameter and chirality are required for SWNTs-based devices. A narrow (n, m) distribution of SWNTs grown from Fe-Ru [11] and Co-Mo [12] catalyst has been reported, indicating the dependence of the chirality of the SWNTs on the type of catalyst. Moreover, it was found that the types of catalysts could affect the diameter distribution of SWNTs [13]. Therefore, the exploration of new catalysts for CVD preparation of SWNTs will be helpful for both controllable growth of SWNTs with specific diameters or chirality and in-depth studies of growth mechanism of SWNTs.

In this paper, we demonstrate that ZnO is an effective catalyst for the synthesis of high-quality SWNTs on the surfaces of silicon wafers. By using ethanol as carbon source and catalyst solution deposited along a strip near the edges of silicon substrates, horizontally aligned SWNT array can be generated. Dense and uniform SWNTs film were characterized using scanning electron microscopy (SEM), Raman spectroscopy, atomic force microscopy (AFM), and high-resolution transmission electron microscopy (HRTEM).

2. Experimental

2.1. Growth of SWNTs on SiO_2/Si wafer using PVP-capped Zn as catalyst precursor and ethanol as carbon source by CVD

For growth of random SWNT networks, the ethanol solution of $ZnCl_2$ (0.5 mM) and PVP (250 mM counted by vinylpyrrolidone monomer) was dropped onto the full silicon wafer. For growth of horizontally aligned arrays of SWNTs, a drop of catalyst solution was deposited along a strip near the edges of silicon substrates. Then the wafer was placed into the quartz tube reactor and calcined in air at 700 °C for 5 min [14]. SWNTs were grown by ethanol CVD method. After the furnace was heated to 900 °C in H_2, then 300 sccm of H_2 was bubbled through ethanol containing 2 wt% deionized (DI) water in a bubbler at a temperature of 20 °C for 10 min at atmospheric pressure to synthesize the SWNTs.

2.2. Characterization

The morphology and microstructure of the SWNTs were characterized using AFM, SEM, HRTEM and Raman. AFM char-

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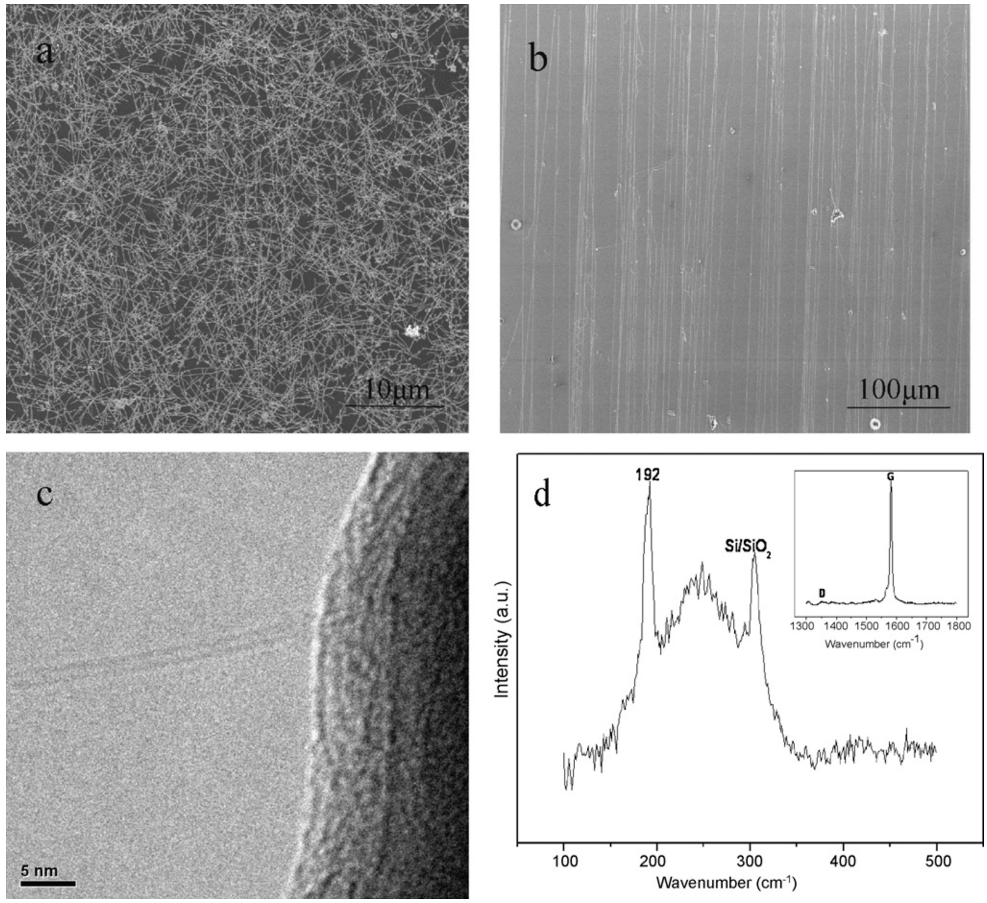

Fig. 1. SEM image of (a) high densely random networks of SWNTs, (b) horizontally aligned array of SWNTs grown on silicon wafer, (c) HRTEM image of individual SWNTs, (d) Raman spectrum of the SWNTs, the inset is typical G band and D band feature of the SWNT.

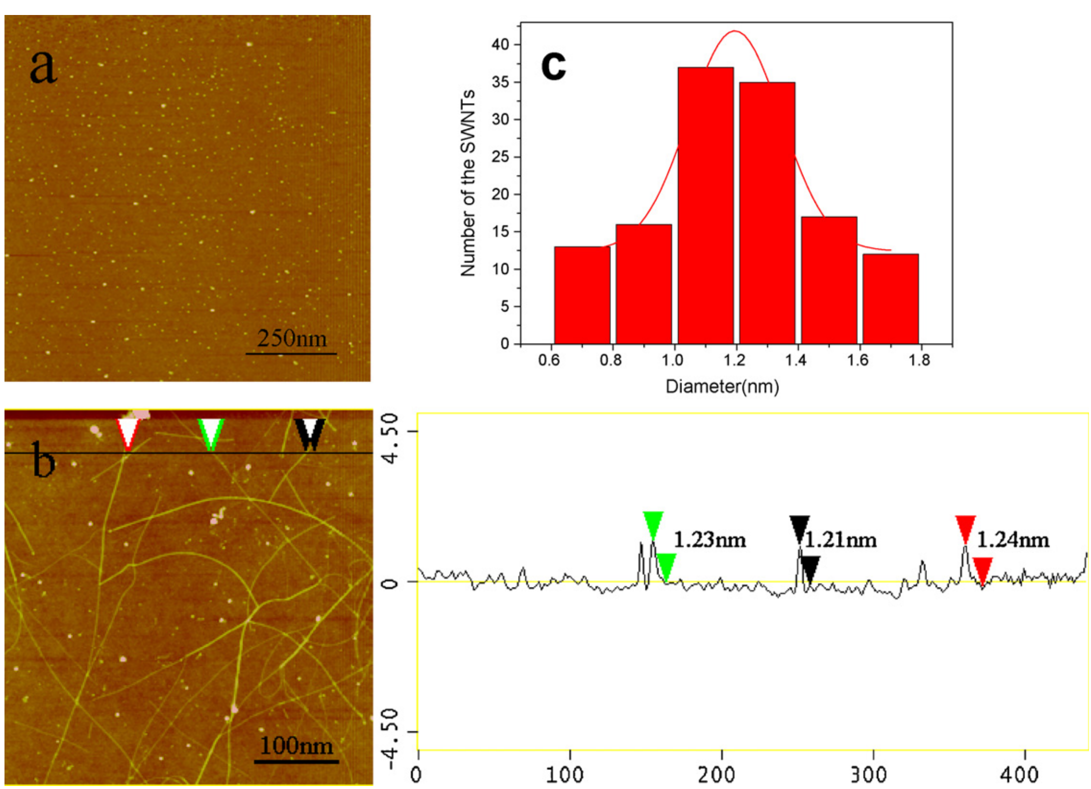

Fig. 2. (a) AFM image of well-separated ZnO nanoparicles when the catalysts were heated to 900 °C in an H_2 atmosphere, (b) AFM image of SWNTs and height measurement, (c) diameter distribution of the SWNTs.