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Controllable growth of individual, uniform carbon nanotubes by thermal chemical vapor deposition

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Abstract

We report on the controllable growth of individual, uniform carbon nanotubes using thermal chemical vapor deposition (CVD). We performed a detailed study of the various factors influencing the growth of single nanotubes. In particular, we investigated the role played by catalyst layer thickness, catalyst dot size, deposition temperature, and gas source pressure on the growth process of straight, single nanotubes. Straight, individual nanotubes with uniform diameter can be obtained by decomposition of 0.1 mbar of acetylene at a temperature of 800 °C over a 5 nm thick nickel film that is patterned into square dots with dimensions below 500 nm. We compare the performance of thermal CVD and of plasma enhanced CVD for growing individual nanotubes.

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Keywords: Chemical vapor deposition; Individual carbon nanotubes; Synthesis

1. Introduction

The control of the growth of carbon nanotubes (CNTs) and their selective positioning is very important for developing CNT-based devices, e.g., sensitive gas detectors or biosensors [1,2], and for integration with conventional microelectronics. The idea of integrating nanotubes with silicon-based electronics to obtain new types of nanoscale devices requires, however, the ability to synthesize individual nanotubes. Individual single-walled CNTs were synthesized on silicon wafers covered with a pattern of micrometer-sized islands of a

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catalyst material [3]. Some of the synthesized nanotubes bridged the gap between two metallic islands in a random and uncontrollable way, and offered the prospect to develop electrical interconnects. Afterwards, directed lateral growth [4–6] of suspended single-walled CNT networks was achieved for elevated tower structures on silicon substrates using liquid-phase catalyst precursor materials [7] and modified catalyst material [8]. Applying an electrical field considerably enhances the possibilities to align nanotubes on a substrate during the lateral growth process [9,10].

Vertical growth of individual aligned nanotubes also received the necessary attention because of the technological significance for applications such as scanning probe microscopy [11] and field emission-based flat panel displays [12]. Ever since the vertical growth of individual CNTs was successfully achieved by plasma-enhanced chemical vapor deposition (CVD) [13], extensive and sustained research activities were developed in this field. Most of the experimental efforts focused on fabrication with the aid of a catalyst material in order to obtain a periodic and controllable positioning of individual CNTs. The controlled positioning relied on electron beam lithography (EBL) [14,15], optical lithography [16], pulse-current electrochemical deposition [17], and polystyrene nanosphere lithography [18,19]. It is worth pointing out that all of the above methods for growing individual nanotubes were based on the plasma CVD technique, enabling the achievement of the required vertical alignment of individual CNTs. However, the fabricated CNTs were cone-shaped structures with a varying diameter along their length, which are less suitable for practical applications. Consequently, conventional thermal CVD recently started to attract the attention of researchers as an alternative method to for convenient, low cost synthesis of individual CNTs [20].

Here, we explore the thermal CVD growth of CNTs, all the way from the mass, random production of CNTs to the growth of individual uniform CNTs. The growth of the individual CNTs relies on the decomposition of acetylene (C_2H_2) over patterns of nickel catalyst material on a silicon substrate covered with a silicon dioxide layer. We performed a detailed study of the various factors influencing the growth of single nanotubes.

2. Experimental

Ni catalyst square patterns are fabricated on a SiO_2/Si substrate by combining EBL with metal evaporation. A barrier layer of SiO_2 is required to prevent silicide formation when using Ni as a catalyst material [21]. We use a bilayer electron beam resist (950 K MW polymethyl methacrylate (PMMA) spun on the top of the copolymer poly(methyl methacrylate-methacrylic acid) (P(MMA-co-MAA))) that is patterned with a modified JEOL JSM-5600 scanning electron microscope (SEM) equipped with a RAITH ELPHY pattern generator. The resist is developed in a solution of methyl isobutyl ketone (MIBK) and isopropyl alcohol (IPA) (MIBK : IPA = 1:1), resulting in a sufficiently large undercut. In this process MIBK develops the PMMA and IPA develops the P(MMA-co-MAA). Next, 2–40 nm of Ni is deposited in a molecular beam epitaxy (MBE) system. Finally, the required Ni pattern is obtained after resist/metal liftoff in acetone.

The nanotubes are synthesized by decomposition of acetylene (C_2H_2) in an electrical furnace. First, a SiO_2/Si substrate containing the Ni patterns is put into a quartz ampoule with a length of 25 cm and an inner diameter of 1 cm, which is then pumped down to a pressure between 10^{-2} and 10^{-3} mbar with a mechanical pump. Next, C_2H_2 gas is introduced into the ampoule up to a pressure of 0.05–0.5 mbar. The growth of nanotubes is carried out in an electrical furnace at a temperature ranging between 700 and 900 °C for 5 min.

3. Experimental results

3.1. Influence of the catalyst

It is believed that the nanotubes grow by decomposition of the carbon containing gas on the surface of catalyst particles (usually Fe, Co, or