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Stack growth of aligned multiwalled carbon nanotubes using floating catalyst chemical vapor deposition technique

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ABSTRACT

The Letter reports another approach to grow vertically aligned millimeter length multiwalled carbon nanotubes (MWCNT) using chemical vapor deposition technique. In this stack growth, the first grown MWCNT layer is observe to have been lift-off from the substrate surface by the newly grown underneath layer as a result of the diffusion of iron catalyst and carbon source through the first layer. The first grown layer acts as a permeable membrane allowing the catalyst vapor and carbon to reach the bottom layer and the top surface of the substrate, resulting in the growth of another layer of MWCNT underneath it.

1. Introduction

Carbon nanotubes (CNT) are one of the allotropes of carbon with a long and cylindrical hollow structures first discovered in 1991. Its unique and excellent characteristics such as high mechanical strength (1 TPa) due to its sp^2 bonding, electronic properties that can be alter by changing the chirality of the tube (metallic or semiconducting) and high aspect ratio (nanometer diameter to millimeter long nanotubes) attracts great interest in various research fields [1–7]. These excellent characteristics of CNT has led to various potential applications such as gas sensing for health and environment monitoring, field emission sources, composite reinforcements, transistor and logic circuits [8–12]. The well-known synthesis techniques of CNT are arc discharge, laser ablation and chemical vapor deposition (CVD) [13]. CVD techniques are suitable for electronic device applications since the CNT can be selectively deposited on any substrate, aligned growth of CNT and large area deposition [14].

Successful growth of CNT using various CVD techniques has been reported [15,16]. Majority of the CNTs grown used fixed catalyst on substrate where the catalyst is pre-deposited on the substrate before the growth of nanotubes. Another method is the floating catalyst, of which the CNT is grown by supplying both the catalyst and carbon source in vapor form during the reaction [16]. Both techniques show different growth mechanism, thus the formation of nanotubes need to be understood to fully optimize the production of highly aligned, good quality, millimeter long CNT. The well-known growth mechanisms are the base and tip growth as described by Sinnott et al. [17] and Baker [18]. For the base growth, the decomposed carbon diffuses into the nanoparticle catalyst and precipitates out in a graphitic form when the solubility limit within the metal is reached. The metal catalyst is firmly fixed on the substrate. However for the tip growth, the precipitating graphitic structure lifts the catalyst particles from the substrate. The detached particles encapsulated in the grown nanotubes. The size of catalyst particles influence the type of graphitic structures grown such as MWCNT, SWCNT, carbon fibers and graphite sheets.

The objective of the present work is to investigate the method of controlling the vapor-phase growth which can lead to the formation of CNT in a structure with a distinct layered pattern. It is expected that the grown layers will consist of well-aligned CNT with high density and clear boundaries between the layers.

2. Methodology

Silicon (1 0 0) wafer of p-type, used as the substrate was cut (size 10 mm × 10 mm) and cleaned by using Radio Corporation of America (RCA) method. A thin layer of silicon dioxide was grown thermally in oxygen environment at 1100 °C for 4 h. Subsequently, a thin aluminum film was deposited using electron beam evaporation and immediately oxidized to obtain alumina by oxidizing for 1 h in oxygen environment at 600 °C. As shown in Figure 1, the prepared samples were placed in the second zone whilst the ferrocene sample in the first zone of the tube furnace. The growth of MWCNT was carried out for pre-determined reaction time by purging ethylene and hydrogen at 100 sccm and 80 sccm, respectively at 850 °C. At the end of the growth process, the CVD system was cooled down to room temperature in argon ambient. This process was

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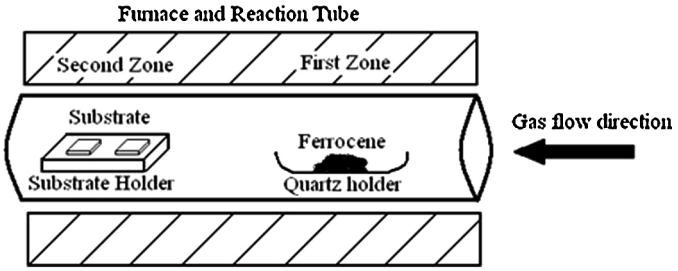

Figure 1. Schematic diagram of the floating catalyst CVD system for the growth of MWCNT.

repeated several times for the growth of several stacked layers of MWCNT.

All samples were analyzed using field emission scanning electron microscope (FESEM) Zeiss Supra55 VP to determine the length, diameter and the surface morphology of MWCNT and structure of metal particles with the possibility of energy dispersive X-ray (EDX) analysis for chemical composition. High resolution transmission electron microscopy (HRTEM-Zeiss Libra) was employed to probe the internal structure of MWCNT and Raman spectroscopy (Horiba JobinYvon) to determine the crystallinity of MWCNT. Ar laser light of 514.53 nm was used for excitation source under air ambient conditions.

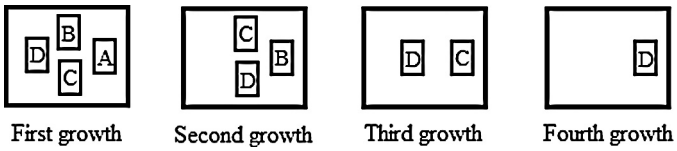

Figure 4. The position of the samples for each growth of MWCNT.

3. Results and discussion

Catalyst thickness and density play an important role for the growth of vertically aligned MWCNT. As reported earlier, successful growth of vertically aligned MWCNT was obtained for iron deposited using ferrocene vaporization [19]. The vaporized iron is in gaseous form and it has an advantage of multidirectional deposition. This creates rougher, thicker and uniform iron distribution on the substrate surface as shown in Figure 2. The average thickness obtained for the coated iron catalyst is around 55 nm. This thick layer of catalyst acts as the nucleation site and provides support for the dense growth of MWCNT. Extended duration of the synthesis (e.g. 60 min) and the position of the substrate with respect to the catalyst source produced millimeter long MWCNT such as shown in Figure 3(a).

Figure 3 shows the grown MWCNT with up to four layers and reaching up to millimeter length. The reaction time for the synthesis of MWCNT for every layer was fixed at 60 min. The stacked layer of MWCNT was sequentially grown from the free moving iron

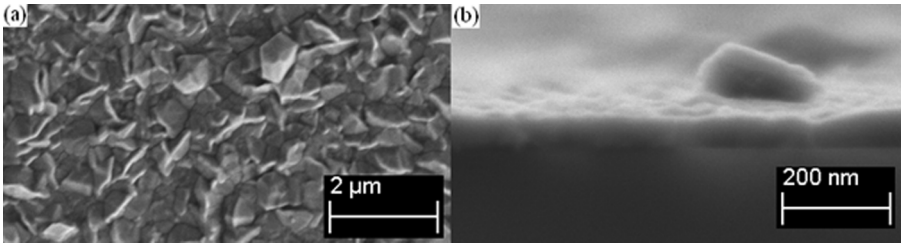

Figure 2. (a) The surface morphology of the coated iron using ferrocene vaporization and (b) the cross-section of the coated iron.

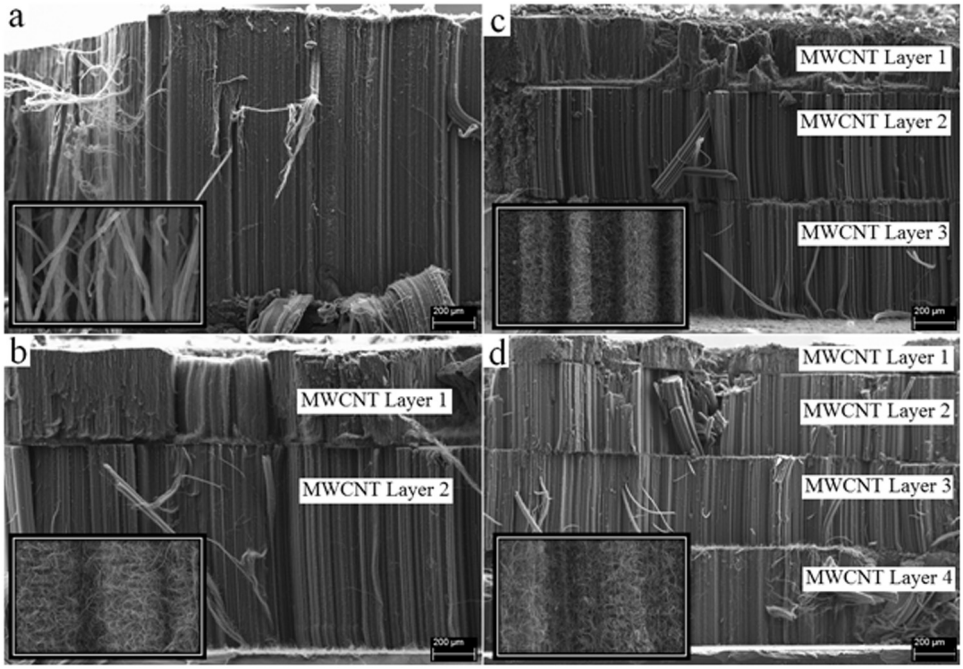

Figure 3. SEM images of the grown MWCNT with (a) one layer, (b) two layers, (c) three layers and (d) four layers.