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Field Emission Behaviour of the Single Wall Carbon Nanotubes Grown by Plasma Enhanced Chemical Vapour Deposition (PECVD) System

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Single wall carbon nanotubes have been grown on Fe using Plasma Enhanced Chemical Vapour Deposition (PECVD) system. The thickness of the Fe film prepared by RF sputtering system was about 10 nm. The field emission characteristic was measured which showed good enhancement factor. The grown CNTs were characterized by various techniques such as SEM, Raman study etc.

Keywords: Carbon nanotubes, PECVD, RF sputtering, Field emission, Scanning electron microscopy, Raman.

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1. INTRODUCTION

The unique electrical, thermal and mechanical properties of carbon nanotubes (CNTs) are expected to find many applications not only for academic interest in physics and chemistry but also for industrial applications such as field effect transistors[1-3], field emission displays [4], sensors [5-7], nano-probes and various types of electronic devices [8, 9]. In such applications, the standard method for single wall carbon nanotube (SWCNT) production rely on co-vaporization of graphite and transition metal catalysts in an inert gas atmosphere, either by an electrical arc [10] or laser vaporization [11]. Dai and co-workers [12, 13] have reported the production of individual SWNTs. Lieber and co-workers [14] produced individual SWNTs for the application in the scanning probe microscope.

However, during the last few years, chemical vapor deposition (CVD) using hydrocarbon as feedstock has emerged as a promising alternative for SWCNT synthesis. The advantages of CVD are lower preparation temperatures, simple equipment, better prospective for large-scale production, and the possibility to grow long and impurity free carbon nanotubes in specified locations on a substrate for incorporation into electronic devices.

However, the synthesis of CNTs requires temperature of 700-1000 °C using thermal chemical vapour deposition (TCVD) method. This temperature requirement far exceeds the temperature limit of microelectronic, which is typically 400-500 °C. For this purpose, Plasma enhanced chemical vapour deposition (PECVD) method has been proposed as an alternative method to further reducing of the synthesis temperature. Kato et al. [15] successfully grew the SWCNTs at temperature of 550 °C using PECVD. Li et al. [16] grew the SWCNTs at temperature of 600 °C. The plasmatic energy in PECVD efficiently dissociates gas molecules at lower temperatures and the synthesis of carbon nanotubes might occur at lower temperatures. The presence of built-in electric field in a plasma sheath aligns the growing CNTs along the field lines. Thus, PECVD method favours low temperature synthesis of VA-CNTs.

2. EXPERIMENTAL

Iron (Fe) catalyst film onto ultrasonically cleaned Silicon (Si) substrate was prepared by RF sputtering. The catalyst film was then placed on a graphite heater which was heated up to 550 °C at a rate of 100 °C/min in a 750 sccm hydrogen flow and staying there for 10 min for the pre-treatment of the catalyst. Acetylene (C_2H_2) at the rate of 20 sccm was then added to start the CNT growth while the heater temperature was quickly raised to 600 °C. The growth time was kept for 15 min. The pressure inside the belljar reactor of the PECVD system was kept at around 15 mbar during pre-treatment and growth processes. During growth process, dc plasma at a power of 40 W, aligns the CNTs vertically from the substrate. The growth is terminated by turning off the power supply of the heater and C_2H_2/H_2 flow. The samples were then cooled down to room temperature.

The CNTs grown sample was characterized using different techniques. SEM (FEI Nova Nano) was used to study the morphology of the as-grown sample. The structure of the grown CNTs was studied by Raman Spectrometer. The sample was irradiated with a wavelength of 633 nm. The field emission measurement was carried out at room temperature in a high vacuum chamber using cathode anode arrangement. Data obtained from the JE and FN plots was used to calculate the field enhancement factor.

3. RESULTS AND DISCUSSION

3.1 SEM Study

Fig. 1 shows the morphology of the uniformly as-grown vertically aligned single wall carbon nanotubes (VA-SWCNTs) characterized by Scanning Electron Microscope (SEM). The diameter of the VA-SWCNTs is found to be in the range of 2 to 3 nm.

3.2 Raman Spectroscopic Study

To evaluate the quality of nanotubes, we performed Raman characterisation of as-grown CNTs using a laser with an excitation wavelength of 633 nm. Fig. 2 shows

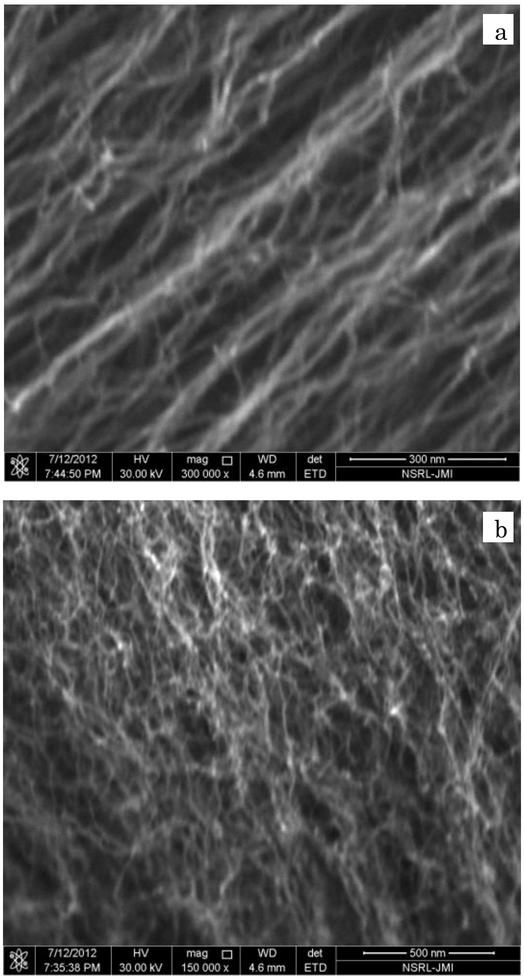

Fig. 1 – Micrograph of VA-SWCNT (a) and micrograph of VA-SWCNT (b)

Raman spectra of CNTs grown on Si substrate. The radial breathing mode (RBM) peak of SWCNTs at 110 cm^{-1} can be seen clearly which confirms the existence of SWCNTs. The diameter of SWCNTs shown in the inset of the Fig. 2 was estimated using the correlation, $d = 248/\text{v}$, where d is the diameter of SWCNT in nm and v is the Raman shift in cm^{-1}.

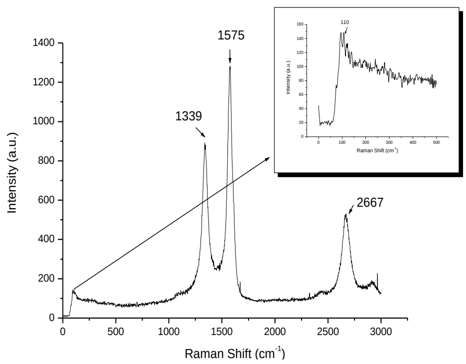

Fig. 2 – Raman Spectra of the grown SWCNTs

3.3 Field Emission Measurements

Field emission measurements are usually analysed in the framework of the Fowler-Nordheim theory, which explains the emission of the electrons from emitter surface as a quantum mechanical tunnelling enhanced by a high electric field. The FN equation can be written as

$$J = AE^2 \exp\left(-\frac{B\varphi^{3/2}}{E}\right) \quad (1)$$

where $A = 1.54 \times 10^{-6}$ AevV^{-2}, $B = 6.83 \times 10^7$ ev$^{3/2}$ V cm^{-1} are constant, E is applied Electric field, Vcm^{-1} and φ is the work function of emitting material.

The enhanced electric field at the CNTs tip E_h is related to the applied electric field via $E_h = \beta E_{applied} = \beta V / d$, β being the field enhancement factor at the sharp tip of the CNTs and d is the CNT cathode and anode separation. Typical FE measurements for as-grown CNTs are shown in Fig. 3 and 4. Electric field versus current density (JE) plot for as-grown CNTs is shown in Fig. 3. As clear from JE plot, as-grown CNTs has the emission current density 2.12mA/cm^2 with turn-on field 3.4 V/μm.

Fig. 4 depicts the FN plot of as-grown CNT cathodes. Almost straight line of FN plot confirms that field emission phenomenon is taking place in our sample. According to the FN model, a plot of $\ln(I / V^2)$ vs. $1 / V$ (known as F-N plot) has a linear behaviour with a slope that is also used to evaluate the field enhancement factor by using following simplified equation derived from FN equation (1)

$$\beta = \frac{B\varphi^{3/2} d}{m} \quad (2)$$

where m = slope of FN plot, d = distance between cathode and anode and $\varphi = 5$ eV as for carbon. The calculated β value from the slope of the FN plot is 1.75×10^3 for the as-grown MWCNTs. Aligned growth of CNTs shows the improvement in the field enhancement factor by way of increased aspect ratio. The increased geometrical field enhancement factor ultimately results in lowering of threshold field and improved field emission current. The measurement results of emission current data obtained by CNT cathode samples in diode configuration show great potential for CNT cold cathode applications in display devices.

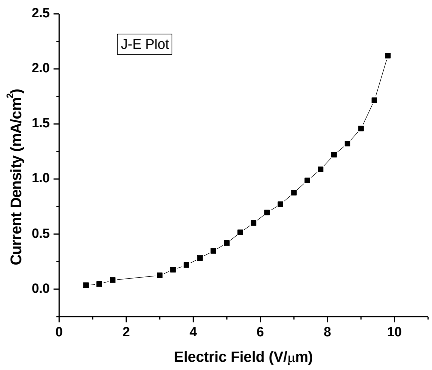

Fig. 3 – J-E Plot of as-grown CNTs cathode

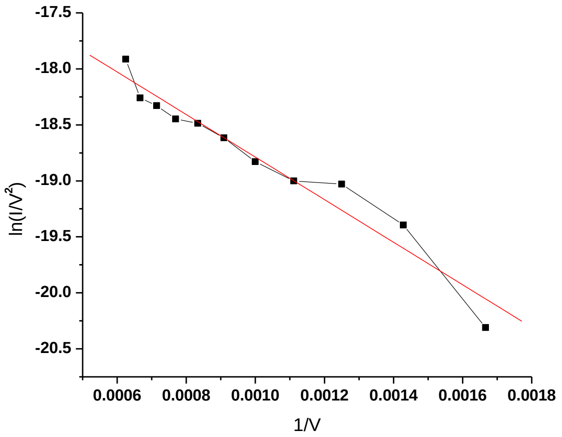

Fig. 4 – F-N Plot of as-grown CNTs cathode

4. CONCLUSION

In conclusion, vertically-aligned SWCNTs up to hundreds of microns in length have been grown on silicon substrate. Radial breathing mode in raman spectra shows the diameter of the SWCNTs 2 to 3 nm. Aligned growth of SWCNTs shows that there is an improvement in the field enhancement factor.

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