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High yield formation of carbon nanotubes using a rotating cathode in open air

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Abstract

Carbon nanotubes (CNTs) have been synthesized using the arc discharge method with a rotating graphite disc as the cathode. Arcing was carried out in open air and without the use of catalysts. The current density was maintained constant through out the experiment, while, the rate of rotation of the cathode and atmosphere under which arcing was carried out were changed during experimentation. Characterization of the samples produced indicates that rotation of the cathode has a significant impact on the quality and yield of the process. It is proposed that rotation of the cathode drags plasma formed between two electrodes away from high temperature region. This results in a sudden quenching of the reactive plasma. The time available for nucleation and growth phenomena is significantly reduced and thus leads to the formation of highly graphitic multi walled CNTs (yield 60%) and traces of double walled CNTs.

Keywords: Nanotubes; DC arc discharge; High resolution electron microscopy

1. Introduction

Since the landmark paper by Iijima in 1991 [1], carbon nanotubes (CNTs) have been studied for their potential application in various fields viz. in electronic devices [2], as an activated fiber and a probe for atomic force microscopy [3], as a strong reinforcement for composite materials and for hydrogen storage [4]. One of the issues that still have to be resolved to exploit the capabilities of CNTs, is the need to synthesize CNTs in high yields in straightforward process. The Arc discharge though has this capability, due to the general ease of formation by this method. However, certain experimental improvements are still necessary to further improve the ease of operation and the yields of CNTs in this process. Crucial requirements in producing CNTs by the arc discharge method include close control of experimental parameters such as plasma temperature, electrode gap and electrode feed rate. Almost all of the work reported in the literature using arc discharge technique for CNT synthesis, involves use of controlled inert atmosphere. Typically helium gas has been used, to create an inert atmosphere. Moreover, arcing has been carried out between a stationary cathode electrode and an anode that has been continuously fed forward. Recently the effect of a high speed rotating anode (anode speed of 10,000 rpm, He atmosphere) called “Plasma rotating electrode process” (PREP) was studied recently [5]. According to their studies this technique leads to a proper mixing of carbon vapor due to a uniform temperature distribution across the embryonic growth of carbon clusters and resulting in significant higher yields compared to classical arcing process without rotating anode. Nevertheless in the PREP technique, as well as in the classical plasma process, arcing is still maintained over the same position of the cathode which affects uniform evaporation of the graphitic anode massively and further arrests the arcing process. Furthermore it would be an improvement towards ease of operation, if the arcing process could be maintained under ambient (air) conditions.

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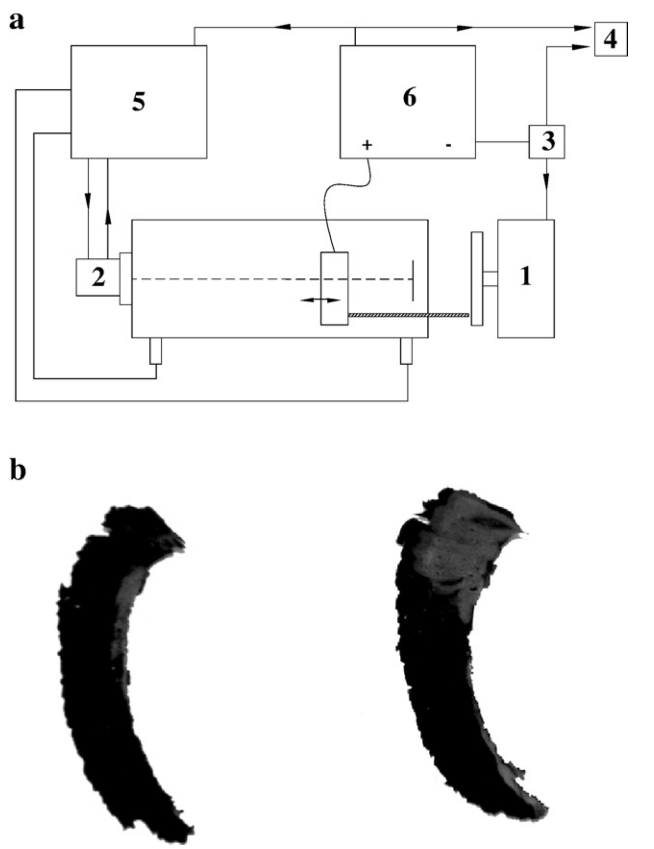

Fig. 1. (a) Schematic diagram for experimental setup; 1: rotating cathode, 2: servo motor, 3: shunt resistance, 4: to computer and 5: control panel, 6: power supply and (b) photograph of soot produced with a rotating cathode in open air.

Herein we present our work on successful synthesis of CNTs using a rotating cathode electrode. In addition we have set up the process in such a way that it can be run under air avoiding conditions of any inert gas atmosphere operation. Therefore our work combines positive aspects of a rotating electrode with the ease of synthesis in open air to maximize yields of CNT formation to as high as 60%.

2. Experimental details

The synthesis of carbon nanotubes was carried out using arc discharge process, with a DC power supply providing 150 Acm^{-2} current density and 36 V. The power supply unit was an AC/DC inverter TIG power source. Graphite rod with diameter 11 mm (99.7% purity) were used as anodes for arcing. Arcing was carried out with a rotating graphite disc as counter electrode. A thick copper metal plate holds the graphite cathode disc. This plate also helped in passive cooling of cathode carbon disc during arcing process. The cathode disc was slowly rotated and speed was maintained constant during each experiment and varied from a speed of 5 rpm to 20 rpm during each experiment. A schematic of the experimental setup is shown in Fig. 1a.

A servo motor fed the anode electrode forward as and when the anode got consumed in order to maintain the electrode gap constant. Soot deposited on the graphite disc cathode was continuously removed using a thin blade that scrapped the cathode as it is rotated. By this technique the cathode was always maintained free of deposited material which is constantly removed from the high temperature reaction region. All experiments were carried out in open air and without the use of any additional metal catalyst.

The raw deposit formed on the cathode had a curved and elongated shape due to rotation of the electrode. Fig. 1b shows the soot produced using this experimental setup. Soot produced was crushed using mortar and pestle to obtain fine powder. Part of this finely crushed powder was then washed in toluene for 5 h and finally heated in open air in a closed furnace at 600 °C for 2 h [6,8].

After purification, samples were characterized using X-ray Diffraction by a Shimadzu XD-D1 X-ray Diffractometer with Cu Kα radiation, Raman Spectroscopy with excitation energy of 10 mW and a wavelength of 514 nm, a Phillips XL30 FEG scanning electron microscope, and a Phillips CM 20 FEG transmission electron microscope used with an acceleration voltage of 200 KV.

3. Results and discussion

Visual examination of raw soot produced indicated that there were some similarities between the soot produced using rotating cathode and the soot produced using a stationary cathode, but there were also some distinct differences. As in the case of regular stationary arc discharge deposit [7], soot produced by rotating cathode method (Fig. 1b) also had central core soft smooth black region. However outer mushroom morphology, typically observed when a stationary cathode is used, was absent. Instead, fine fibrous structures originating from central core region were present. These fibrous structures were aligned in direction of disc rotation. The elongated and curved raw soot deposit was several centimetres long while its thickness was 1 to 2 mm.

The net yield in this study was observed to be dependent upon speed of cathode disc rotation and atmosphere under which the

Table 1
Table showing yield calculations

Atm.	Cathode disc speed	W_1	W_2	W_3	S_1	S_2	Yield of the process		
							Y_A	Y_O	Y_N
Helium	5	50.05	26.76	15.29	5.00	4.58	65.65	91.61	60.14
(500	10	49.96	33.52	5.79	5.02	4.47	35.20	89.19	31.40
Torr)	15	33.53	17.76	9.39	5.00	3.35	59.19	66.98	39.40
	20	26.76	15.19	4.74	3.81	2.48	40.93	65.11	26.65
Open Air	5	49.44	35.59	8.38	5.00	4.41	60.51	88.19	53.36
	10	49.84	32.19	10.34	5.00	4.60	58.58	91.92	53.85
	15	35.59	20.65	7.45	5.00	4.59	49.85	91.73	45.73
	20	32.19	12.50	9.85	5.00	4.39	50.02	87.73	43.88

(W_1 and W_2 are weight of anode rod before and after arcing, W_3 weight of soot collected from cathode end, S_1 and S_2 Weight of soot before and after oxidation. $Y_{A(\text{yield after arcing})}=W_3/(W_1-W_2)*100$, $Y_{O(\text{yield after oxidation})}=(S_1-S_2)*100/S_1$, and $Y_{N(\text{total yield})}=Y_A^*Y_O$.) (all weights are calculated in grams).