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Synthesis of Single-Walled Carbon Nanotubes by Low-Frequency Bipolar Pulsed Arc Discharge Method

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Abstract: The effective synthesis of single-walled carbon nanotubes (SWNTs) by low-frequency bipolar pulsed arc discharge method using a newly developed bipolar pulsed current circuit with constant current and constant pulse duration is reported. In this method, there is no cathode deposit, which means that all the sublimated carbon becomes soot containing single-walled nanotubes. The soot production rate of the bipolar pulsed arc discharge method is compared with those of the DC, the AC, and the unipolar pulsed arc discharge methods. It is found that the soot production rate increases with increasing frequency whereas the quality of the produced SWNTs remains almost the same. The nanostructures of the synthesized SWNTs are characterized by a TEM and Raman spectroscopy.

Keywords: Single-walled carbon nanotubes, bipolar pulsed arc discharge, synthesis, input power, Raman spectroscopy

1. Introduction

Single-walled carbon nanotubes (SWNTs) have been expected to have various industrial applications since they were discovered [1-3]. Because of their desirable properties such as high mechanical strength, chemical stability, and high aspect ratio, SWNTs have many nanometer-scale applications, which include electronic devices, optical devices, nanoscale sensors, nanocomposite materials [4-6]. However, the progress of applications has been held back by the lack of a method for producing high-yield, high-quality SWNTs. Therefore it is necessary to find high-performance routes for the desired production of SWNTs. Different techniques have been used for the synthesis of SWNTs such as electric arc discharge [7-10], chemical vapor deposition [11, 12], laser ablation [13], the pyrolysis of hydrocarbons [14], and high temperature single pulsed arc discharge [15]. The arc discharge method has attracted attention owing to its simple operation, smaller number of defects in the produced SWNTs and less content of amorphous carbon. However, there are a number of parameters in the process such as the gas pressure, the discharge current, the distance between the electrodes, the electrode size, and the content and components of the metal catalyst, which should be controlled and adjusted to improve the efficiency and selectivity of the SWNT production [16]. A reaction model for the production of SWNTs by the arc discharge method has been proposed previously from our laboratory [17]. The growth of SWNTs is strongly affected by the gas temperature, diffusion speed, cooling rate of the particles, carbon density and catalyst particle density [18, 19].

Generally in the DC arc discharge method, SWNTs are produced by the arc discharge between a graphite cathode and a graphite anode containing a metal catalyst (Ni+Y). During discharge sublimated carbon from the anode tends to deposit on the cathode surface by diffusion in an electric field, this carbon deposit does not contain SWNTs [20, 21]. In this case, the amount of cathode deposit is about 40% of the source material indicating a major loss of the source material. The deposit changes the shape of the cathode and shifts the arc position, which disturbs the control of the discharge. The AC arc discharge is also employed to produce SWNTs where the two electrodes have same diameter [9, 10]. In this case there is no deposition on the electrode. However, as the discharge current is sinusoidal, the input power is timely modulated, which causes the cooling effect of the electrodes [22]. As the carbon sublimates above the threshold temperature, the carbon sublimation rate considerably decreases, and the production efficiency of SWNTs becomes much lower.

To suppress the carbon deposit and increase the production efficiency of SWNTs, a bipolar pulsed arc discharge method has been developed in our laboratory, where two graphite electrodes of the same diameter containing a metal catalyst (Ni+Y) are used, and constant plus current and minus current flow alternately. The reason for employing a bipolar pulsed arc discharge is that both electrodes will sublime carbon alternately and there are no carbon deposits on both electrodes. As the electrodes are kept clean, the discharge control is easy. The electric power is effectively input to the arc region like the DC arc discharge. There are a few reports on the use of

unipolar pulsed arc discharges [23] and high frequency arc discharges for the preparation of CNTs [24]. To the best of our knowledge, no systematic study has been performed on the utilization of the bipolar pulsed arc discharge. Therefore, we have studied the low-frequency bipolar pulsed arc discharge for the large-scale synthesis of SWNTs by controlling the bipolar-pulse current.

2. Experimental Setup and Method

The experiment is carried out using the system schematically shown in Fig. 1. The system consists of a stainless-steel vacuum chamber of 150 mm diameter and 200 mm height, which is pumped by a rotary pump. Two rectangular type C-Ni-Y electrodes ($6 \times 6 \times 50 \text{ mm}^3$, Toyo Carbon Cluster Mate, including 4.2 wt% Ni and 0.9 wt% Y) are set horizontally inside the chamber, and the gap between them can be controlled by a motor drive. The chamber has two large optical windows (100 mm in lengths and 40 mm in widths) to observe the discharge process. First, the chamber is evacuated to less than 5 Pa. Then He gas with a pressure of 50 kPa is introduced into the chamber through a regulator. Then a regulated DC power supply with constant-current control (Daihen ARGO-300P, 0-300 A) is switched on, and a DC bipolar pulse converter (which was developed in our laboratory) is activated by a pulse generator (HP-3312A). This converter has 6 high-current electric relays, and produces bipolar square-wave pulse current, where the current is 0-70 A and the frequency is 0-5 Hz. The applied pulse frequency is set to 0.05, 0.1, 0.2, 0.5, 1 or 2 Hz and the applied current is set to 55 A. The duty ratio is 50 % and the absolute values of the positive and

negative currents are the same. The interchange time from -55 A to +55 A is about ~ 10 ms. The arc discharge is started by contact ignition. The gap distance is kept at approximately 5 mm and the voltage between the electrodes is 23-25 V. The discharge time is 20 min. All the parameters are carefully controlled since a slight change in the parameters would change the discharge condition. The behavior of the applied voltage and the current are monitored by using a digital oscilloscope (Aglient U1604B). A video camera is set in front of one of the optical windows to observe the discharge process. After the discharge, the chamber is opened, the produced soot is separately collected from the top, side, and bottom parts of the chamber, and their weights are measured. To prove the reproducibility of our experiment, the arc productions are carried out several times for each frequency. To compare the bipolar pulsed arc discharge results with those of other arc discharge methods, a number of samples are produced by a DC arc discharge, an AC arc discharge and unipolar pulsed arc discharges under the same experimental conditions.

The morphology of the nanotubes is observed by a transmission electron microscope (TEM) (Hitachi H-7500, acceleration voltage of 100 kV). Their Raman spectra are measured by a Raman spectrometer (JASCO NR-1800, $\lambda = 532$ nm).

3. Results and Discussion

At first, time variation of the discharge parameter is measured. Figure 2 shows the waveform of the discharge current I_d , voltage between the electrodes V_E , and power P , during the discharge

when the pulse frequency $f = 0.11$ Hz. The ratio of the plus current time to the minus current time is 50/50. It is shown that the positive and negative values of discharge voltage and current are symmetrical, and the waveforms are rectangular. The symmetrical nature of the discharge current as indicated in Fig. 2(a) confirms that the bipolar pulsed current is well controlled. The absolute discharge voltage is gradually decreasing with time during one pulse shown in Fig. 2(b), most likely because the carbon sublimation from the electrodes would slightly change the discharge condition. From Fig. 2(c), it is shown that waveform of the input power exhibits the rectangular behavior. The quasi-constant power makes the constant sublimation of carbon atoms from the electrodes for each half cycle.

The production rate of the carbon soot and the sublimation rate of the electrodes in case of the bipolar pulsed arc discharge are examined by changing the pulse frequency. Production rates of the soot for the six frequencies are shown in Fig. 3(a). The production rate gradually increases with increasing the pulse frequency. It is conjectured that the number of current change in a unit discharge time slightly affects the sublimation, though the discharge power is kept constant. In the bipolar pulsed arc discharge, the two electrodes alternately behave as the cathode and anode with short interchange time, and both receive the same amount of electron heating. In this situation, both the electrodes contribute to the sublimation of carbon atoms above a threshold temperature, and the deposition on the cathode is suppressed. Therefore, both the electrodes have nearly the same sublimation rates. Sublimation rates of the two electrodes for the six frequencies are shown

in Fig. 3(b). The measured values are summarized in Table 1, where summation of the sublimation rate from the two electrodes is listed up.

In case of the AC arc discharge, both electrodes undergo sublimation. The AC arc discharge is also carried out in our laboratory by varying the discharge current. The effective current is 50-60 A. It is found that both, the soot production rate and the electrode sublimation rate increase with increasing the discharge current, but the production rate is considerably smaller compared with the bipolar pulsed arc discharge. The measured values are shown in Table 1. In case of the AC arc discharge, the input power changes sinusoidally with time. The sinusoidal input power produces heating and cooling, and the sublimation takes place above the threshold temperature. Therefore the sublimation rate seriously decreases.

The unipolar pulsed arc discharge is also examined by varying the frequency. The discharge current is 50 A in a half period and 30 A in the other half period. In case of the unipolar arc discharge, only the anode sublimates carbon like the case of DC discharge, and there is also a large cathode deposit. The numeric results are shown in Table 1. Though the production rate and the electrode sublimation rate of the unipolar pulsed arc discharge increase by increasing the frequency, they are smaller compared with the cases of the DC arc discharge and the bipolar pulsed arc discharge.

From Table 1, we can compare the differences among the bipolar pulsed arc discharge, the DC arc discharge, the AC arc discharge and the unipolar pulsed arc discharge. For the DC arc

discharge, about 40 % of the sublimated carbon is deposited on the cathode. For the unipolar pulsed arc discharge, 20-30% of the sublimated carbon is deposited. There are no deposit in case of the bipolar pulsed arc discharge and the AC arc discharge. During the discharge, the electrode deposit is carefully observed from the viewing port.

In the bipolar pulsed arc discharge, the discharge is very stable and there are no fluctuations of the arc flame. Both the electrodes remain in good condition. When the pulse frequency is increased to more than 5 Hz, the interchange time (10 ms) becomes no longer negligible, and the production rate of the soot monotonously decreases with pulse frequency.

Figure 4 shows TEM images of the soot from the bipolar pulsed arc discharges and the DC arc discharge, where right side of Fig. 4 shows the higher magnification images. Left side of Fig. 4 shows the lower magnification images, where there are considerable numbers of SWNTs present in the both cases. Usually the SWNTs tend to aggregate into bundles [25]. Higher magnification of the TEM images shows that there are bundles of SWNTs, metal catalyst particles and amorphous carbon layers covering the particles in the both cases. No double walled nanotubes (DWNTs) and multiwalled nanotubes (MWNTs) are seen in the both samples. Bundles of SWNTs usually have three dimensional structures so that all the SWNTs in the bundles would not be observed. It is observed that the bundles of SWNTs consist of 1-12 SWNTs. The number of SWNTs in a bundle is counted in the TEM images and it is found that the average number of SWNTs in a bundle in case of the DC arc discharge is 3.1, whereas they are 3 to 4 in case of the bipolar pulsed arc

discharge. It is also observed that the bipolar pulsed arc discharge has a tendency to produce thicker bundles of SWNTs. The tube diameters are approximately 1.0- 1.5 nm. This result is in good agreement with the results obtained from the Raman spectra as shown in Fig. 6. It is difficult to measure the exact length of the SWNTs from the images because they are long and winding. Fig 5(a) shows the lower magnification image, and the length of the bundles from the image is measured to be about 3 μm . From the several TEM images, the estimated length is in the range of 1-10 μm . The number of carbon particles (N_{cp}) and the number of SWNT bundles (N_{ntb}) are counted from the TEM images with the same magnification ($\times 50000$) over an equal area of $1.44 \times 10^6 \text{ nm}^2$ and their ratio is calculated. The result is shown in Table 2. Generally shapes of the carbon particles are spheroid where both metal catalyst particles and simple amorphous-like carbon particles are considered. The densely stacked region of carbon particles and SWNTs are excluded from the images due to the difficulty of counting. It is found that the number ratio of SWNT bundles to carbon particles gradually increases with the pulse frequency, and it is slightly higher than the result of DC arc discharge. These ratios can be used to compare the quality of the SWNT production. The size distributions of the metal catalyst particles are calculated by measuring the diameters of more than 100 catalyst particles from the TEM images over the same equal area ($1.44 \times 10^6 \text{ nm}^2$). Diameter of metal catalyst particles is listed in Table 2 and the standard deviations of each case are also calculated. The diameter of the catalyst particles, D_M , increases slightly with pulse frequency. The size of metal catalyst particles may influence on the production of SWNTs.

Fig 5(b) shows that nanotubes are attached to the catalyst particles as they were grown from the catalyst particles. It is observed that nanotubes and catalyst particles are embedded in an amorphous carbon flakes.

Raman spectroscopy is used to measure diameters of the SWNT, and the intensities of the G band and D band. The left side of Fig. 6 shows the radial breathing mode (RBM) of the produced soot for the DC arc discharge and the bipolar pulsed arc discharges. The wave number at each RBM peak is directly dependent on the nanotube diameter through the relation $\omega_{\text{RBM}} = (A/d) + B$, where A and B constants vary between single SWNT and SWNTs bundles [26]. To characterize the diameter distribution of SWNT bundles, $\omega_{\text{RBM}} = A/d + B$ are used; where $A = 234 \text{ nm cm}^{-1}$ and $B = 10 \text{ cm}^{-1}$ [27, 28]. For the bipolar pulsed arc discharge, there are two dominant RBM peaks at approximately 170 and 187 cm^{-1} in most of the cases, from which the diameters are calculated to be 1.46 and 1.32 nm , respectively. In the DC arc discharge, the RBM peak is at approximately 174 cm^{-1} , which corresponds to a diameter of 1.42 nm .

The right side of Fig. 6 shows the D (1345 cm^{-1}) and G (1590 cm^{-1}) bands of the Raman spectra of the produced soot. They provide information about the presence of disordered and ordered phases in graphite materials [29-31]. From the graph, it is observed that the amount of ordered carbon is much larger than the amount of disordered carbon in both the DC and the bipolar pulsed arc discharge methods. Figure 7 shows the intensity ratios I_D/I_G for the six frequencies. In case of the bipolar pulsed arc discharges, I_D/I_G values are from 0.021 to 0.026 , which is similar to

the previous results for the DC arc discharge obtained at our laboratory [6, 21]. In this case, the DC arc discharge has a slightly larger I_D/I_G value. The variation in these spectra might be the result of slight change of the experimental condition during the production. The variations of I_D/I_G values are small among the frequencies. The production process occurs in the cooling region above the arc plasma between the electrodes and there is a convective flow of He gas.

In case of the bipolar pulsed arc discharge method, source material is effectively used without cathode deposition. Moreover, the control of the arc discharge is easier as the shape of the electrodes does not change significantly because both the electrodes sublime almost the same amount of carbon. In this method, the effective input of electric power and the effective sublimation of carbon material comparable with the DC arc discharge method are obtained. The bipolar pulsed arc discharge will make a new way for high efficiency production of SWNTs.

4. Conclusions

The synthesis of SWNTs by the bipolar pulsed arc discharge with graphite electrodes of the same diameter containing Ni/Y catalyst is carried out. This method can realize the effective input of electric power to the electrodes and the effective sublimation of carbon material from both electrodes with no carbon redeposition. As the electrodes are kept clean during the discharge, the discharge is well controlled. A number of arc discharge methods have been examined in this article. Compared with the other arc discharge methods, the bipolar pulsed arc discharge is found to be

very stable. The bipolar pulsed arc discharge has a good production rate of SWNTs when the pulse frequency is 0.05-5 Hz. The Raman analysis shows that the diameters of the SWNTs are mostly range from 1.32 to 1.46 nm which is in good agreement with the TEM results. The TEM images also show that the nanotubes are attached with catalyst particles. The small I_D/I_G ratio in the Raman measurement indicates that the quality of SWNTs is as good as that from the DC arc discharge method.

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Figure Captions

Fig. 1. Schematic of experimental setup of the bipolar pulsed arc discharge reactor.

Fig. 2. Waveforms of (a) discharge current I_d , (b) electrode voltage V_E and (c) input power P with the pulse frequency, $f = 0.11$ Hz.

Fig. 3. (a) Variation of production rate of the soot, W_{soot} , for six pulse frequencies. (b) Variation of sublimation rates, $W_{electrode}$, from the two electrodes.

Fig. 4. TEM images of the soot synthesized by (a) the DC arc discharge, and by the bipolar pulsed arc discharges of (b) $f = 0.05$ Hz, (c) $f = 0.5$ Hz, and (d) $f = 2$ Hz. The left side images and the right side images have magnification of ($\times 50000$) and ($\times 100000$), respectively.

Fig.5. (a) A lower magnification ($\times 15000$) image to measure the nanotube length. (b) A higher magnification ($\times 300000$) TEM image of nanotubes which are attached to the catalyst particles.

Fig. 6. Raman spectra for the DC arc discharge and the bipolar pulsed arc discharges with frequencies, $f = 0.05$, 0.5 and 2 Hz. The RBM modes are shown on the left side, and the G and D bands are shown on the right side.

Fig. 7. Variation of I_D/I_G ratios of soot from the bipolar pulsed arc discharges for $f = 0.05$ - 2 Hz, which are calculated from the data of Fig. 5.

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Table 1. Soot production rates, total electrode sublimation rates, and cathode deposition rates for the bipolar pulsed arc discharge, the DC arc discharge, the AC arc discharge and the unipolar pulsed arc discharge.

Discharge method	Discharge current I_d (A)	Frequency f (Hz)	Soot production rate W_{soot} (mg/min)	Total electrode sublimation rate $\Sigma W_{electrode}$ (mg/min)	Cathode deposition rate W_{C-DEP} (mg/min)
Bipolar pulsed arc	55/-55	0.05	12.46	13.35	No deposit
		0.1	12.55	13.66	No deposit
		0.2	12.71	13.95	No deposit
		0.5	13.39	15.64	No deposit
		1	13.86	16.85	No deposit
		2	14.82	18.08	No deposit
DC arc	55	0	13.02	25.40	8.92
AC arc	50 (effective current)	60	2.56	4.19	No deposit
	55 (effective current)	60	2.64	4.26	No deposit
	60 (effective current)	60	3.32	5.95	No deposit
Unipolar pulsed arc	50/30	0.5	5.09	9.63	2.78
		1	5.39	8.42	1.77
		2	5.59	10.50	2.07
		5	5.87	10.95	1.85
		10	6.11	10.63	1.77
		15	6.42	11.19	2.56
		20	8.02	12.09	2.42

Table 2. The ratio of number of SWNT bundles to the number of carbon particles, $N_{\text{ntb}}/N_{\text{cp}}$, diameter of metal catalyst particles, D_M for the DC arc discharge and the bipolar pulsed arc discharge.

Discharge method	Frequency f (Hz)	Ratio of number of SWNT bundles to the number of carbon particles, $N_{\text{ntb}}/N_{\text{cp}}$	Diameter of catalyst particles D_M (nm)
DC arc discharge	0	0.54	11.98 ± 0.13
Bipolar pulsed arc discharge	0.05	0.58	12.21 ± 0.10
	0.5	0.59	12.33 ± 0.11
	2	0.62	12.64 ± 0.11