Effect of arc behaviour on the temperature fluctuation of carbon electrode in DC arc discharge

F Liang¹, M Tanaka¹, S Choi¹ and T Watanabe¹, ²
¹Dept. Environmental Chemistry and Engineering, Tokyo Institute of Technology, Yokohama, Japan
²Dept. Chemical Engineering, Kyushu University, Fukuoka, Japan

watanabe@chem-eng.kyushu-u.ac.jp

Abstract. Diffuse and multiple arc-anode attachment modes were observed in a DC arc discharge with a carbon electrode. During the arc discharge, the surface temperature of the electrode was successfully measured by two-colour pyrometry combined with a high-speed camera which employs appropriate band-pass filters. The relationship between the arc-anode attachment mode and the temperature fluctuation of electrode surface was investigated. The diffuse arc-anode attachment mode leads to relatively large temperature fluctuation on anode surface due to the rotation of the arc spot. In the case of diffuse mode, the purity of synthesized multi-wall carbon nanotube was deteriorated with temperature fluctuation.

1. Introduction

The arc discharge is an attractive method to prepare nanomaterials with several advantages [1]. Firstly, it is an environmentally benign technique without generating any toxic by-products and hazardous gases. In addition, the high temperature of arc plasma provides the products with fewer defects. Finally, high productivity in the arc discharge method reduces the manufacturing cost.

The arc discharge method has also been employed to prepare lots of carbon nanomaterials successfully. Fullerene was scraped from the chamber wall in helium arc discharge at low pressure [2]; multi-wall carbon nanotube (MWNT) was collected from cathode deposit at low pressure argon arc [3]; single-wall carbon nanotube was found in a “collaret” around the cathode deposit, the web-like structure suspended between cathode and chamber wall, and cloth-like soot suspended in the chamber wall by using the catalyst in helium arc [4]; and graphene flakes was produced in the inner wall of the chamber under hydrogen atmosphere [5].

In spite of the considerable efforts on the arc discharge method, however, the formation mechanism of carbon nanomaterial has not been understood yet. Understanding the characteristics of thermal plasma is necessary to explain the formation mechanism of carbon nanomaterial. The anode provides the carbon species for the production of carbon nanomaterial, and the anode ablation rate determines the productivity. Therefore, further analysis of the anode region and anode phenomena is required to improve the production of carbon nanomaterial by arc discharge method. For the anode boundary, there are three typical arc-anode attachment modes, diffuse attachment, multiple attachments, and constricted attachment [6]. Heberlein et al investigated the formation mechanism and the characteristics of arc-anode attachment modes with a metal anode [7-9]. However, the arc-anode...
attachment in a carbon electrode and the effect of arc-anode attachment mode on the electrode temperature fluctuation have not been reported.

The temperature of carbon electrode is an important parameter in determining the characteristics of carbon nanomaterial. Therefore, an accurate temperature measurement is required to understand the growth of carbon nanomaterial in dynamic interaction between the electrode and arc plasma. The measurement of electrode temperature, however, is difficult with conventional methods because of the strong radiation and dynamic behavior of the arc column.

In the present work, the two-colour pyrometry combined with a high-speed camera which employed appropriate two band-pass filters was applied to measure the surface temperature of carbon electrode during the arc discharge. The objective of this work is to investigate the effect of arc-anode attachment mode on the electrode temperature fluctuation. In order to achieve the purpose, the effect of arc current on the arc behavior and the electrode temperature was investigated.

2. Experimental

2.1. Experimental setup

Figure 1 indicates the schematic illustration of an arc discharge system for the preparation of carbon nanomaterials and a high-speed camera system for temperature measurement. A graphite anode rod of 30 mm in diameter (99.99%, Toyo Tanso Co., Ltd.) with an inclined top plane was put on a water-cooling copper plate. A graphite rod of 6 mm in diameter (99.99%, Toyo Tanso Co., Ltd.) was placed at an oblique angle from the anode. The arc was generated for 5 min in helium environment at atmospheric pressure. The arc current was controlled at 100, 150, and 200 A and the electrode gap

Figure 1. Schematic diagram of arc generation system.

Figure 2. Schematic illustration of temperature measurement system by two-color pyrometry.

Figure 3. Images of the electrode at the wavelength of (a) 880±5 nm and (b) 763±3 nm recorded by the high-speed camera, and (c) a corresponding temperature distribution map calculated by Plank’s law based on radiation intensity from images (a) and (b).
The evaporated carbon species were deposited on the peripheral of the evaporated hole of the anode to form the anode deposit, where grows the carbon nanomaterial.

2.2. Temperature measurement

Figure 2 shows the schematic illustration of two-colour pyrometry for the temperature measurement of electrode surface during the arc discharge. The radiation emitted from the electrode surface was divided by a splitting mirror, and then it passed through two band-pass filters. In order to eliminate the influence of line emission from the arc, two band-pass filters of 763±3 and 880±5 nm were employed. According to optical emission spectrum, band-pass filters were selected with no line emission in the bandwidth of them. As shown in Figures 3 (a) and (b), images of the electrode in different wavelengths at 880±5 and 763±3 nm were simultaneously recorded by the high-speed camera (FASTCAM-SA WTI, Photron). The frame rate was fixed at 10,000 fps (frame per second) and the shutter speed was fixed at 20 μs.

Plank’s law was used to evaluate 2-dimensional temperature distribution of the electrode surface in two-colour pyrometry. Since the emissivity $\varepsilon$ is unknown for many applications, the grey body assumption $\varepsilon(\lambda_1) = \varepsilon(\lambda_2)$ is used where it is hypothesized that the surface emissivity is independent of wavelength. Therefore, surface temperature is expressed as follows:

\[
T = \frac{C_b(\lambda_1 - \lambda_2)}{\lambda_1^2 \lambda_2^2 \ln(I_1 \lambda_1^2 / I_2 \lambda_2^2)}
\]

where, $\lambda_1$ and $\lambda_2$ are selected wavelengths (nm) of two band-pass filters; $C_b$ equals to 0.014388 mK; and $I_1$ and $I_2$ correspond to radiation intensities (W·m⁻³·sr⁻¹) at the wavelengths of $\lambda_1$ and $\lambda_2$, respectively. Considering bandwidths of band-pass filters and the sensitivity of high-speed camera, the two-colour pyrometry system was calibrated by a tungsten lamp which was used as a standard light source. As shown in Figure 3 (c), the temperature distribution map was obtained by the above equation with the measured intensity ratio of selected radiations. Since the sensitivity of the high-speed camera was poor at a low temperature region, only high temperature region where the temperature is over than 2,000 K was calculated in the temperature distribution map. In Figure 3, the anode deposit region locating at the peripheral area of the evaporated hole was measured to investigate the temperature fluctuation effect on the characteristics of synthesized carbon nonmaterial.

2.3. Voltage and arc behavior measurement

The voltage of arc discharge was recorded by an oscilloscope (Scope Corder DL 850, Yokogawa) at 10 MHz. For the observation of dynamic arc behavior, the same high-speed camera was used at the frame rate of 10,000 fps.

3. Results and discussion

Figure 4 shows the snapshots of high-speed camera at different arc currents of 100, 150 and 200 A. A band-pass filter which has the wavelength of 763±3 nm was used. Several small bright regions appear on the anode surface randomly without periodicity at the lowest arc current of 100 A. These bright regions represent high temperature areas on the anode surface, which correspond to the anode spots. It means that the multiple arc-anode attachment mode was formed [7], which was confirmed by high-speed camera observation at a shorter shutter speed of 0.37 μs. The multiple arc-anode attachment mode was also observed at 150 A. In contrast, a bright region with a relatively large size appears on the anode surface when the arc current reaches to 200 A. In addition, the bright region rotates on the anode surface periodically. A single arc spot is explained by the formation of diffuse attachment mode [7]. However, one bright region is separated into several small regions at 4 and 5 ms in Figure 4 like as the multiple attachment mode. Therefore, the arc-anode attachment mode is the mixture of diffuse attachment and multiple attachment modes and is dominated by diffuse attachment mode in the case of 200 A.
As shown in Figure 4, the rotation frequency of bright area is about 125 Hz when the arc current is 200 A. Although the reason of this periodical arc rotation is unclear, the Lorenz force induced by the arc itself is related to the periodical arc behavior [10]. The formation of different arc-anode attachment modes is explained by the competition of the cathode jet and the anode jet. The strong cathode jet leads to the transition of arc-anode attachment mode from diffuse attachment mode into multiple attachment mode with increasing the arc current.

In order to examine the characteristics of dynamic arc discharge behavior, the change of arc voltage

![Figure 4. Representative snapshots of high-speed camera at different arc currents.](image-url)

![Figure 5. Voltage waveform at different arc currents: 100, 150, and 200 A. The corresponding FFT spectra of voltage waveform at different frequency ranges.](image-url)
according to time was traced. Figure 5 shows the waveforms of the arc voltage and the corresponding Fast Fourier Transform (FFT) spectra at different arc currents. These voltage waveforms exhibit similar shapes derived from the current ripple of the power supply. As a result the main frequency is 100 Hz, which comes from the power supply. According to the FFT spectrum of the voltage waveform in a wide range, the arc voltage fluctuated with high frequency about 10 kHz in multiple attachment mode. In contrast, the relatively small amplitude of peak is observed in the FFT spectrum of voltage waveform with the highest arc current of 200 A due to the diffuse attachment mode. In addition to the common peak at 100 Hz and its harmonics, the peak at 125 Hz is found in the case of 200 A. This characteristic frequency corresponds to the arc rotation which was measured by the high-speed camera in Figure 4.

Figure 6 shows the snapshots of electrode temperature distribution according to time at different arc currents. The high temperature regions on anode surface correspond to the bright area in the snapshots of high-speed camera in Figure 4. In the arc current of 200 A, it was evaluated that the rotation frequency of high temperature region is about 125 Hz which is the same frequency with the arc spot rotation.

Figure 7 indicates the FFT spectra of anode temperature at different arc currents. No significant peak can be observed in the range of 0 to 500 Hz in multiple attachment mode when the arc current are 100 and 150 A. However, a dominant peak around 125 Hz is appeared in the diffuse attachment mode when the arc current reaches to 200 A. This fluctuation frequency of anode temperature comes from the rotation of arc spot.

Figure 8 shows the time variation of the temperature of anode deposit during 30 ms. The anode deposit temperature fluctuated with the frequency of 125 Hz in the diffuse attachment along with the arc rotation. Although the average temperatures of anode surface in different attachment modes were similar as 2,700 K for the multiple mode and 2,690 K for the diffuse mode, the standard deviations of
measured temperature were quite different as 181 K for the multiple mode and 260 K for the diffuse attachment mode, respectively. According to temperature distribution of the anode deposit, the MWNT with higher purity was obtained in multiple attachment mode than diffuse attachment mode, due to the large temperature fluctuation in diffuse attachment mode. Since different carbon nanomaterials can be obtained in different temperature ranges, the temperature with large fluctuation amplitude and small fluctuation frequency results into the low purity of MWNT on the anode deposit.

4. Conclusion
The multiple arc-anode attachment mode and the diffuse arc-anode attachment mode were observed by changing the arc current in the DC arc discharge with carbon electrode. The arc-anode attachment mode was transformed from the multiple mode into the diffuse mode when the arc current changed from 100 to 200 A. The FFT spectrum of voltage waveform indicates the frequency around 10 kHz can be observed in the multiple attachment mode. In contrast, the arc spot moved with a constant frequency of 125 Hz in the diffuse attachment mode. The temperature of carbon electrode was successfully measured by using two-colour pyrometry combined with a high-speed camera during the arc discharge. The anode deposit temperature fluctuated at the frequency of 125 Hz in diffuse attachment at 200 A due to the rotation of arc spot. Enhanced temperature fluctuation results in the degradation of purity for the final product from the anode deposit.

References
[1] Keidar M, Shashurin A, Li J, Volotskova O, Kundrapu M and Zhuan T 2011 J. Phys. D: Appl. Phys. 44 174006
[2] Krätschmer W, Lamb L, Fostiropoulos K and Huffman D R 1990 Nature 347 354
[3] Iijima S 1991 Nature 354 56
[4] Keidar M, Levchenko I, Arbel T, Alexander M, Waas A M and Ostrikov K 2008 Appl Phys Lett. 92 043129
[5] Subrahmanyam K S, Panchakarla L S, Govindaraj A and Rao C N R 2009 J. Phys. Chem. C. 113 4257
[6] Sanders N, Etemadi, Hsu K C and Pfender E 1982 J. Appl. Phys. 53 4136
[7] Heberlein J V, Mental J and Pfender E 2010 J. Phys. D: Appl. Phys. 43 023001
[8] Yang G, Heberlein J V and Pfender E 2006 J. Phys. D: Appl. Phys. 39 2764
[9] Yang G and Heberlein J V 2007 Plasma Sources Sci. Technol. 16 529
[10] Collares M P and Pfender E 1997 IEEE Trans. Plasma Sci. 25 864