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Electrode Temperature Measurement for Carbon Nanomaterial Production by Arc Discharge Method

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Abstract: Carbon electrode temperature in DC arc method was successfully measured by using high-speed camera system with appropriate band-pass filters. Obtained result revealed that arc-anode attachment mode can be controlled by changing arc current, resulting in the different anode temperature and its fluctuation. The temperature of carbon electrode has an important role in the nanomaterial production.

Keywords: Electrode temperature, Carbon electrode, DC arc, Arc-anode attachment.

1. Introduction

The arc discharge method has been applied to prepare lots of carbon nanomaterials such as fullerene, multi-wall carbon nanotube (MWNT), single-wall carbon nanotube (SWNT), polyhedral graphite particle (PGP), and graphene flakes [1-5]. These carbon nanomaterials were formed on different positions according to the growth temperature. Cathode and anode as the main growth regions of these carbon nanomaterials have been investigated [6, 7]. However, in spite of the considerable efforts on the arc discharge method, the formation mechanism of the carbon nanomaterial has not been understood yet.

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. However, the electrode temperature cannot be measured by conventional methods because of the strong radiation and dynamic behavior of the arc.

In the present work, the two-color pyrometry combined with a high-speed camera and the appropriate band-pass filters was applied to solve the above problems in the temperature measurement. The objective of this work is to measure the temperature of carbon electrode during the arc discharge and to investigate the dynamic behavior of the carbon electrode on the millisecond time scale. 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 experimental apparatus 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 at the fixed electrode gap distance of 3 mm.

The anode was evaporated by the high heat flux from the arc, leaving an evaporated hole at the arc attaching region on the anode surface. The evaporated carbon was deposited on the peripheral area of the evaporated hole to form the anode deposit.

2.2 Temperature measurement

Figure 2 shows the schematic illustration of two-color pyrometry for the temperature measurement of carbon electrode during arc discharge. The radiation emitted from the carbon electrode was divided by a splitting mirror, and then it passed through two band-pass filters. In order to minimize the influence of line radiation from the arc, two band-pass filters of 763±3 and 880±5 nm were selected.

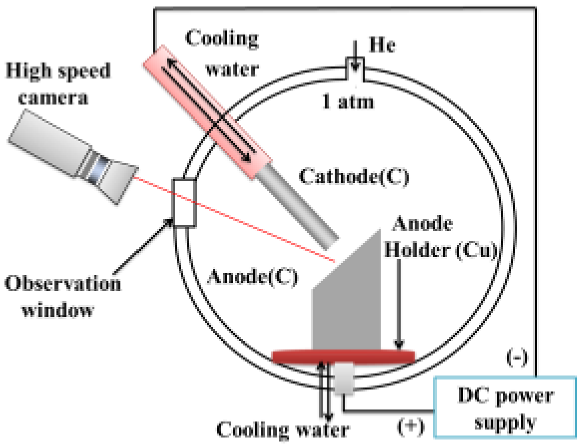

Fig. 1 Schematic diagram of experimental apparatus for the preparation of carbon nanomaterials by arc discharge method

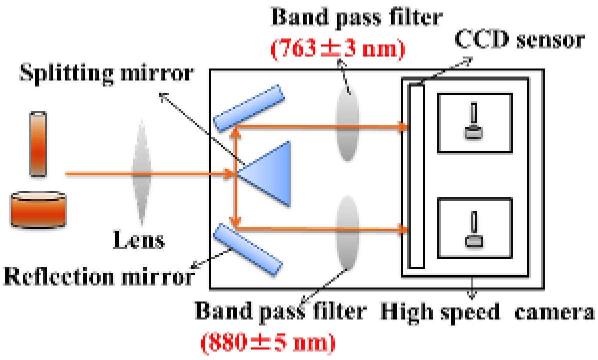

Fig. 2 Schematic illustration of the temperature measurement system by two-color pyrometry

The band-pass filter was selected with no line emission in the bandwidth of these two filters. As shown in **Fig. 3 (a) - (b)**, the images of the electrode at wavelengths of 880±5 and 763±3 nm were simultaneously recorded by the high-speed camera (FASTCAM-SA WTI, Photron). Measurement conditions of the high-speed camera were the frame rate of 10000 fps, shutter speed of 20 µs.

Plank's law was employed in two-color pyrometry to evaluate 2-dimensional temperature distribution of the carbon electrode. With the assumption of the same radiation emissivity at different wavelengths, electrode temperature can be expressed by the following equation.

$$T = \frac{C_b(\lambda_1 - \lambda_2)}{\lambda_1 \lambda_2} \frac{1}{\ln(I_1 \lambda_1^5 / I_2 \lambda_2^5)} \quad (1)$$

where, λ_1 and λ_2 are selected wavelengths through band-pass filters; C_b equals to 0.014388 mK; I_1 and I_2 correspond to radiation intensities ($W \cdot m^{-3} \cdot sr^{-1}$) at the wavelength of λ_1 and λ_2, respectively. Considering the bandwidth of band-pass filters and the sensitivity of high-speed camera, the two-color pyrometry system was calibrated by employing a tungsten lamp which was used as a standard light source. As shown in **Fig. 3 (c)**, temperature distribution map was obtained by the above equation with the measured intensity ratio of selected radiations.

Figure 3 (c) indicates two representative regions of the anode surface, which were taken into account. The central of anode surface was focused to investigate the effect of arc-anode attachment mode on the temperature fluctuation. In addition, the anode deposit region locating at the peripheral area of the evaporated hole was measured to investigate the temperature fluctuation effect on the carbon nanomaterial production. In the present work, the position at 0.5 mm from the evaporated hole was selected as the representative anode deposit.

2.3 Voltage and arc behavior measurement

The voltage of each electrode was recorded at 10 MHz by an oscilloscope (Scope Corder DL 850, Yokogawa).

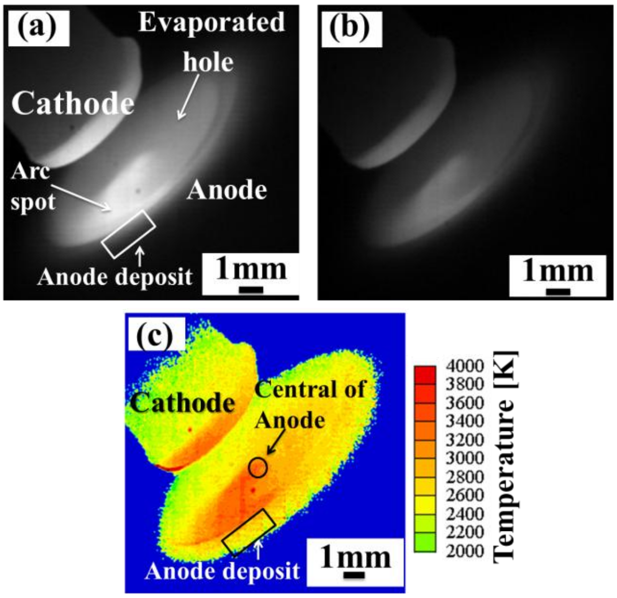

Fig. 3 Images of the electrode at the wavelength of (a) 880±5 nm and (b) 763±3 nm recorded by the high-speed camera, and (c) a corresponding temperature distribution map calculated by Plank's law based on radiation intensity from images (a) and (b).

For the arc behavior observation, the same high-speed camera was used at the frame rate of 10000 fps.

3. Results and discussion

The voltage trace as an easy way to provide basic characteristics of arc discharge was employed. **Figure 4 (a) - (c)** shows the waveforms of the arc voltage at different arc currents. These voltage waveforms exhibit similar shapes which are derived from the current ripple of the power supply at 100Hz of frequency. Moreover, the arc voltage fluctuated with high frequency about 10kHz at the arc current of 100 and 150 A.

Figure 4 (d) shows a Fast Fourier Transform (FFT) spectrum of the voltage waveform at 200 A of the arc current. In addition to the peak at 100Hz and its harmonics derived from the power supply, the peak at 125Hz was found. This characteristic frequency corresponds to the arc behavior. The detail will be explained at the following paragraph.

Figure 5 shows the snapshots of high-speed camera at the wavelength of 763±3 nm and the corresponding temperature distribution maps. The high temperature regions clearly correspond to the bright area in the high-speed snapshots. This result indicates the electrode temperature was successfully measured during arc discharge, because the high-speed camera with suitable band-pass filters can cut the arc emission effectively.

High temperature regions appear on the anode surface randomly without constant periodicity at the arc current of 100 A. Several small regions with high temperature were observed simultaneously. The reason for the several high

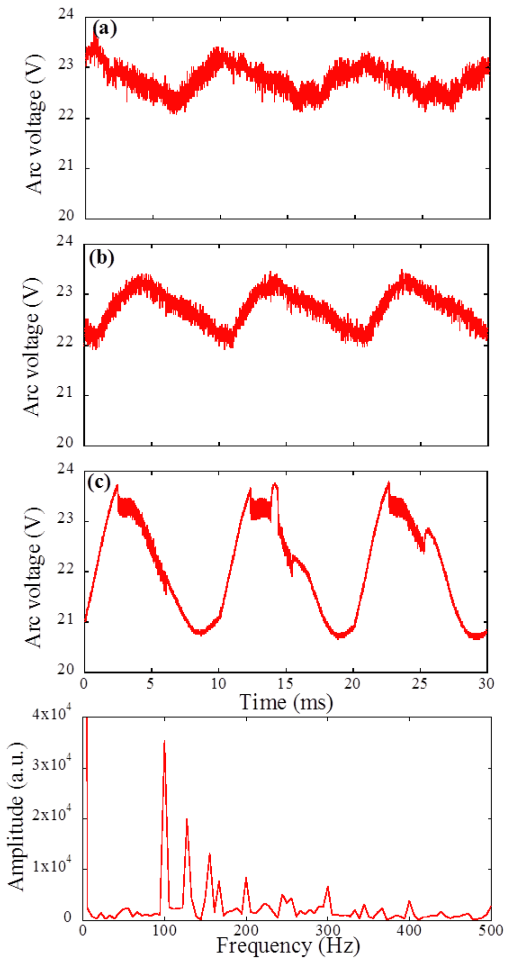

Fig. 4 Arc voltage fluctuation at different arc currents: (a) 100 A, (b) 150 A, and (c) 200 A. (d): corresponding Fast Fourier Transform (FFT) spectra of voltage at 200 A

temperature regions were the multiple arc-anode attachments, which were confirmed by high-speed observation at shorter shutter speed of 0.37 μs.

High temperature region with a relatively large size appears on the anode surface when the arc current reaches to 200 A. The temperature behavior follows the arc behavior as shown in the corresponding snapshots. It can be explained by the formation of diffuse attachment [8], due to the strong cathode jet at the arc current of 200 A. It was evaluated that the frequency about the rotation of high temperature region is about 125Hz, which is the same with the frequency of arc spot rotation at the arc current of 200 A. Although, the reason of this periodical arc rotation is not clear, the Lorenz force induced by the arc itself should relate to this periodical arc behavior.

In order to understand the dynamic behavior of the carbon electrode during the arc discharge, temperature fluctuation was investigated at two kinds of representative regions of the anode, the central region and the deposit region. The temperature of the central region should be related to the productivity of the carbon nanomaterial because the carbon anode is mostly evaporated from the central region. In addition, the temperature of the deposit region has an important role in the controllability of carbon products.

Figure 6 shows the time variation of the anode temperature in central region during 30 ms. The temperature has higher fluctuation frequency when the arc current is 100 A. The reason for the high frequency of the anode temperature fluctuation at the arc current of 100A is the formation of the multiple arc-anode attachment. Contrastingly, four peaks about temperature fluctuation were clearly found during 30 ms at the arc current of 200 A. This characteristic frequency of about

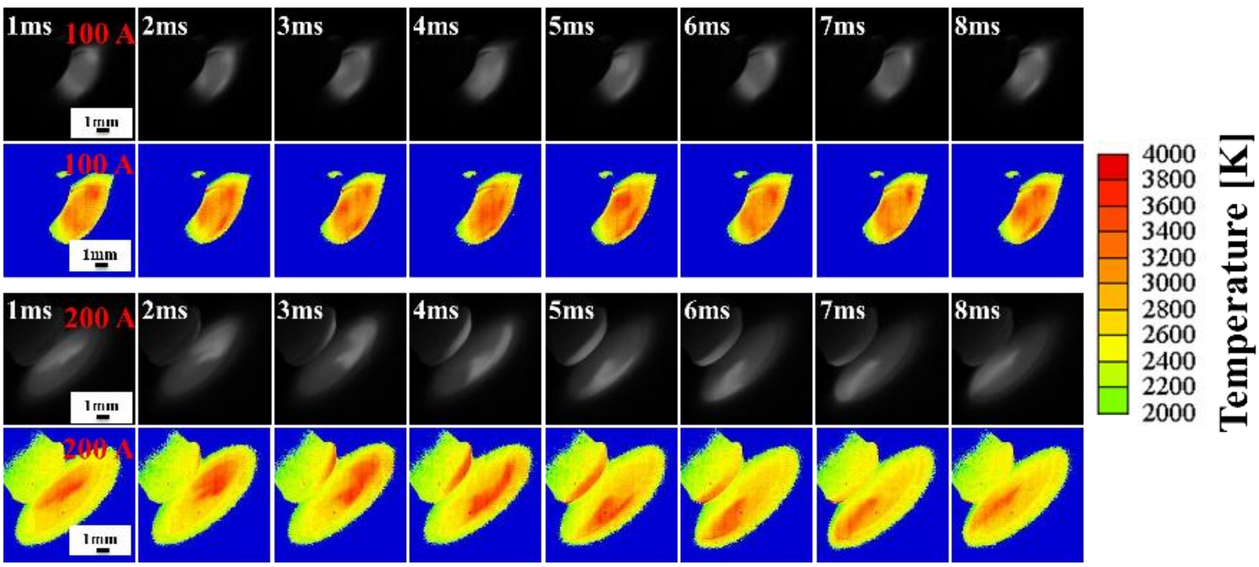

Fig. 5 Representative snapshots and the corresponding temperature distribution maps at different arc currents.

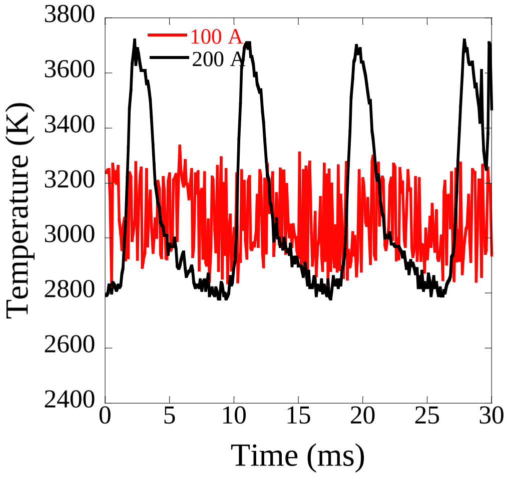

Fig. 6 Temperature variation of anode surface with the time at different arc currents

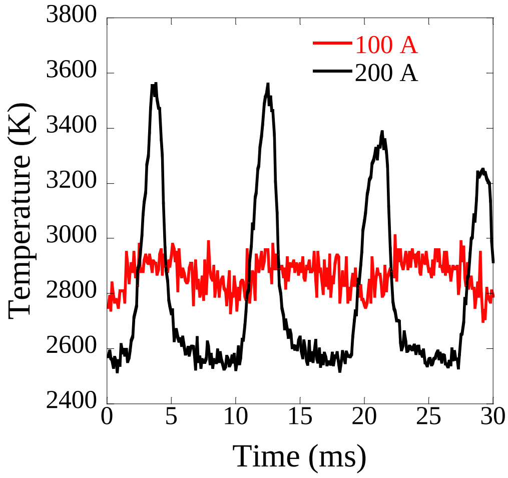

Fig. 7 Temperature variation of anode deposit with the time at different arc currents

125Hz is same with the frequency of the arc rotation evaluated by both the voltage analysis and the high-speed image analysis. Thus, the temperature fluctuation at 200 A of arc current is strongly influenced by the arc rotation, which can be observed only in the diffuse attachment mode.

Figure 7 shows the time variation of the anode temperature in anode deposit region during 30 ms. Both of the anode deposit regions are 0.5 mm away from the evaporated hole on the anode. The frequency of the anode deposit temperature is 100 Hz at the arc current of 100 A, while the anode deposit temperature fluctuated with the frequency of 125Hz as well as the anode temperature in the central region at 200 A. The calculated standard deviation of the temperature was 60 and 300 K for the arc current of 100 and 200 A, respectively. In addition, the average temperature of 2750 and 2870 K correspond to the arc current of 100 and 200 A, respectively**.**

The main product of MWNTs with high purity was obtained at 100 A, whereas the mixture of MWNTs and PGPs were found when the arc current reached to 200 A. These different products can be explained as follows. Thermodynamically unstable material can be formed at high temperature region, thus PGPs can be formed in the region with higher temperature, MWNTs can be formed in the region with lower temperature [9]. High purity of MWNTs were obtained at 100 A, since the low average temperature with small temperature fluctuation. The high average temperature with large temperature fluctuation resulted in the mixture of MWNTs and PGPs at the arc current of 200 A.

4. Conclusion

The temperature of carbon electrode was successfully measured by using the high-speed camera system with the appropriate band-pass filters during DC arc discharge. The anode temperature, including the central region and the deposit, has a relationship with the arc-anode attachment mode. The temperature fluctuation of the central region has higher frequency at the arc current of 100 A due to the formation of multiple arc-anode attachments, while the frequency of 125Hz was obtained in diffuse attachment mode when the arc current reaches to 200 A. In order to obtain the carbon nanomaterial with high purity, to control the fluctuation of anode deposit temperature is required. The information of the electrode temperature and its fluctuation enables us to understand the electrode phenomena and to improve the productivity and the controllability of the carbon nanomaterial.

5. References

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