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Effects of Gas Pressure on Size of Carbon Nanoparticles Prepared by Methane Plasma Process

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1. Introduction

Carbon nanoparticles have become attractive for an increasing number of their applications such as electronics, catalysts, and fuel cells. Their nanoscale size, structure and components are the keys to enhancing physical, chemical, and biological properties of carbon nanoparticles [1-3].

Plasma process is one of the promising methods to synthesize carbon nanoparticles [4-7]. Conventional size control is realized by pulse discharges, while the throughput is rather low. Therefore, continuous fabrication of size controlled nanoparticles is important. So far, we have developed a multi-hollow discharge plasma chemical vapor deposition (MHDPCVD) method [8]. Using the method, we have realized to continuously synthesize size-controlled Si nanoparticles of 2 nm in size with a narrow size dispersion of 0.5 nm [9]. For the method, particle size is controlled by the short gas residence time. Here we have produced carbon nanoparticles using the MHDPCVD method and studied effects of gas pressure on their size.

2. Experimental

Experiments were carried out using the MHDPCVD having a multi-hollow electrode consist in 8 hollows of 5 mm in diameters in the electrode. CH_4 and Ar gas were supplied at the total gas flow rate of100 sccm. The flow rate ratio of Ar to CH_4 was 6:1. The total pressure was set in a range of 2 to 5 Torr. Plasma discharges were generated in hollows with 60MHz radio frequency power source at 40 W. The discharge duration was 90 min.

Nanoparticles were sampled by mesh grids for TEM, and Si substrates set on the sample holder at 50 mm downstream from the electrode. The size of nanoparticles were measured by TEM. Their structure was characterized by Raman spectroscopy using 532 nm laser light excitation.

3. Results and discussion

Figure 1 shows size distribution of nanoparticles as a parameter of gas pressure. For 2 Torr, mean size of nanoparticles is 37.7 nm. The size range is from 17.5 nm and 70 nm. The maximum area density is 5.9×10^{13} m^{-2} at 35 nm. The size monotonically increases with increasing pressure from 37.7 nm for 2 Torr to 46.3 nm for 5 Torr. The size range for 5 Torr is from 17.5 nm to 110 nm. While the minimum size for 5 Torr is same as that for 2 Torr, the maximum size for 5 Torr is larger than that for 2 Torr. The maximum area density for 5 Torr is 4.3×10^{13} m^{-2} which is lower than that for 2 Torr. These results suggest that collision frequency between two nanoparticles increases with increasing gas pressure. Nanoparticles are grown by coagulation between two nanoparticles.

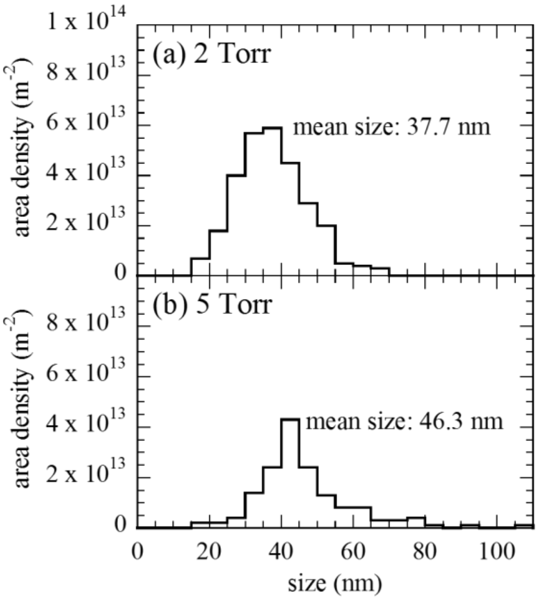

Fig. 1. Size distribution of carbon nanoparticles as a parameter of gas pressure.

We have measured Raman spectra of the nanoparticle deposited on Si substrates. G- and D-peaks were detected in the spectra both for 2 and 5 Torr. The background which corresponds to polymer components increases with increasing the gas pressure. It suggests that structure of carbon nanoparticles is changed by the gas pressure.

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