Ar/CH₄マルチホロー放電プラズマ中にカーボンナノ粒子の成長機構 Growth Kinetics of Carbon Nano Particles Generated

in an Ar/CH₄ Multi-Hollow Discharge Plasmas

黃 成和, 鎌滝 晋礼, 板垣 奈穂, 古閑 一憲, 白谷 正治 Sung Hwa Hwang, Kunihiro Kamataki, Naho Itagaki, Kazunori Koga, and Masaharu Shiratani

> 九州大学 Kyushu University

1. Introduction

Research interest of carbon nanoparticles (CNPs) has been diversified due to their excellent properties. Among many methods to synthesize CNPs, low pressure plasma processes are one of the promising methods, which can produce them in gas phase and transport toward the substrate [1]. So far, we have successively produced Si nanoparticles and controlled their size and structure using the multi-hollow discharged plasma chemical vacuum deposition (MHDPCVD) method [2]. Here, we examined optical emission spectroscopy (OES) measurements to understand the formation mechanisms of CNPs.

2. Experiments

Experiments were carried out using the MHDPCVD method, for which plasmas were sustained in 8 small hollows of 5 mm in diameter. Discharge frequency and power were 60MHz and 40W, respectively. CNPs were produced in hollows where Ar and CH₄ gases pass through. Working pressure and Ar/CH₄ ratio were kept at 2 Torr and 6 to 1 respectively. CNPs were sampled by grounded grid meshes for a transparent electron microscope (TEM) which set at 50 mm downstream from the electrode. Size distribution were observed by using TEM (JEOL-JEM2010). To obtain the information of radical generation and electron energy distribution, optical emission from the plasmas was measured by using a spectroscope (Ocean Optics-USB2000+).

3. Results and discussion

Fig.1 shows the TEM images of CNPs as a



Fig. 1. TEM images of CNPs. Scale bar in each image shows 500 nm.



Fig. 1 Normalized intensity of Ar and CH4 plasma.

parameter of gas flow rate from 10 to 125 sccm. With increasing the gas flow, the mean particle size decreases from 223 nm for 10 sccm to 31.6 nm for 120 sccm. The area density also increases. For above 125 sccm, no particles deposit on the TEM mesh. Fig.2 indicates the normalized intensity of ArI lines at 425.9, 750.4 and 811.5 nm and CH* line at 432.6 nm. They gradually decrease in a gas flow range from 10 sccm to 100 sccm. However, it becomes almost constant above 100 sccm as 60% of that for 10 sccm. The intensity ratio is irrelevant to the gas flow rate. These results suggest that the generation rate of carbon related radicals decreases with increasing gas flow velocity until 100 sccm, while the electron energy distribution has less sensitivity against the gas flow rate. This tendency is related to the electron collision rate of electron density to gas molecules. For the gas flow rates less than 100 sccm, the growth rate of CNPs decreases with increasing the gas flow rate, while the growth rate of CNPs is irrelevant to the gas flow rate.

References

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