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Pulse Frequency Effects on Carbon Nitride Synthesized Using Pulsed Microwave Plasma CVD

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Abstract

Carbon nitride has fascinating properties that include high hardness and high current density of field emissions. In addition, if c-C_3N_4 or β-C_3N_4 structure can be synthesized, then it is possible to obtain high hardness exceeding that of diamond. Pulsed plasma CVD can be used with low substrate temperature and can improve the film quality and growth rate. For this study, an investigation was conducted of pulse frequency effects on crystalline carbon nitride synthesized using pulsed microwave plasma CVD. The pulse frequency was varied from 30 to 3000 Hz, maintaining a constant duty cycle of 20%.

Results of SEM observation show that crystalline deposits resembling hexagonal rods were obtained from 300 Hz of microwave frequency. Deposits resembling spheres were observed for CW, 30 Hz, and 3000 Hz. From Raman spectra, only the silicon peak was observed at 300 Hz of microwave frequency. However, a DLC peak was observed at CW, 30 Hz, and 3000 Hz. From XPS measurement, SiC, C–C, C–N, C=N, and Si_3N_4 bonds were observed in all samples. Results show that the effect of pulse frequency on crystalline carbon nitride synthesized using pulsed microwave plasma CVD, crystalline carbon nitride was obtained using a 300 Hz pulse frequency.

Keywords: pulse plasma, carbon nitride, plasma CVD

1. Introduction

Carbon nitride has fascinating properties that include high hardness and high current density of field emissions. Furthermore, if c-C_3N_4 or β-C_3N_4 structure can be synthesized, then it is possible to obtain high hardness exceeding that of diamond [1,2].

Therefore, many studies of carbon nitride synthesis from vapor phase have been attempted using various chemical vapor deposition (CVD) and physical vapor deposition (PVD) methods. Films prepared using PVD are generally amorphous. A small amount of nitrogen was included in the film [3]. Some reports describe that crystalline deposits were obtained using plasma CVD [4].

The authors tried to obtain crystalline carbon nitride. Crystalline deposits were obtained from a mixture of CH_4-N_2 and C_2H_4-H_2-N_2 reaction gas systems using microwave plasma CVD [5,6]. Based on results obtained using the C_2H_4-H_2-N_2 reaction gas, control for optimal plasma is necessary to obtain crystalline carbon nitride. In addition, results indicate that the growth rate is low and that the substrate temperature is high, e.g., 1473 K.

Pulsed plasma CVD can be used with a low substrate temperature, high film quality, and a high growth rate. In addition, pulsed plasma can change the plasma state and control radicals in plasma. A wide range of pulse frequencies can be used for the pulsed plasma. For example, diamond synthesis can be conducted at 50–1000 Hz [7,8,9]. Pulse plasma is regarded as useful for carbon nitride synthesis.

However, the effects of pulse frequency on carbon nitride synthesized using pulse plasma remain unclear. Investigations were conducted of the effect of pulse frequency on crystalline carbon nitride synthesized using pulsed microwave plasma CVD.

2. Experimental method

Fig. 1 presents a schematic diagram of microwave plasma CVD apparatus. Carbon nitride was synthesized using an improved microwave plasma CVD apparatus equipped with a

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