Effects of Radio-Frequency Glow-Discharge Reactive Plasma on Carbon Nanotube Growth Process

T. Hirata , N. Satake, G. -H. Jeong,
R. Hatakeyama, and K. Tohji *

Department of Electronic Engineering, Tohoku University, Sendai 980-8579, Japan
*Department of Geoscience and Technology, Tohoku University, Sendai 980-8579, Japan

Since their discovery, carbon nanotubes (CNTs) are hoped to provide new possibilities for application in various fields because of their novel properties. Although plasma enhanced chemical vapor deposition (PECVD) method has become a common way for synthesizing CNTs, there are still many unclear points in the relationship between the CNTs production and plasma condition. Here our aim is to clarify essential experimental parameters in the CNTs production using relatively low-energy controlled reactive plasmas generated by a magnetron-type radio-frequency (RF) glow discharge [1, 2].

In our experiment various parameters can be controlled externally, which are CNTs growth time, RF electrode DC voltage (V_{RFE}), DC current density toward the RF electrode (J_{RFE}), strength (B_z) of magnetic-field lines parallel to a surface of the RF electrode, etc. For the CNTs production, a mixture of CH_4 and H_2 is introduced into the chamber in the total-pressure of 0.5 Torr.

A time evolution of the CNTs growth is shown in SEM images of Fig.1. A Ni substrate on the RF electrode is firstly exposed to an Ar plasma for 15minutes prior to deposition. This pre-treatment has two purposes. One is to heat the substrate up to around 700 ℃ before the CNTs growth and the other is to form small projecting islands in the nano scale on the RF electrode surface, as shown in Fig1(a). It is assumed that the Ar plasma sputtering of the RF electrode is accompanied with the development of these islands. We can find a tendency that CNTs start to grow from the islands on the RF electrode surface as shown in Fig1(b), (c). Thus these islands are considered to play an important role in the CNTs formation, especially at the primitive stage of growth. After 15 minutes, well-aligned CNTs are individually formed on the substrate and their length becomes around 10 μm as shown in Fig.1(d).

It is also found that V_{RFE} and J_{RFE} have crucial effects on the CNTs structure formed by our method. Figure 2 shows dependences of CNTs growth on different J_{RFE}, V_{RFE}, and B_z. When J_{RFE} is 0 mA/cm^2, CNTs grow only scatteredly on the RF electrode as indicated in Fig.2 (a). On the other hand, CNTs uniformly grow in the distinct form in the case of J_{RFE} = 1.5 mA/cm^2, as shown in Fig.2 (b). In any case the CNTs are observed to grow along local electric-field lines due to the strong potential drop in the plasma sheath formed by V_{RFE}. However, when V_{RFE} becomes extremely low (≤-500 V) and an increase in J_{RFE} is accompanied, the CNTs formed once are damaged probably due to the sputtering by the surplus of ion flux from the plasma, as seen in Fig.2 (c) These results indicate that the diffusion of hydrocarbon precursors on the RF electrode surface and the electric field around the electrode play an important role in synthesizing CNTs.

In conclusion, the initial condition of the RF electrode, amount of ion flux toward the RF electrode, and potential drop in the plasma sheath are highly effective for the CNTs growth.

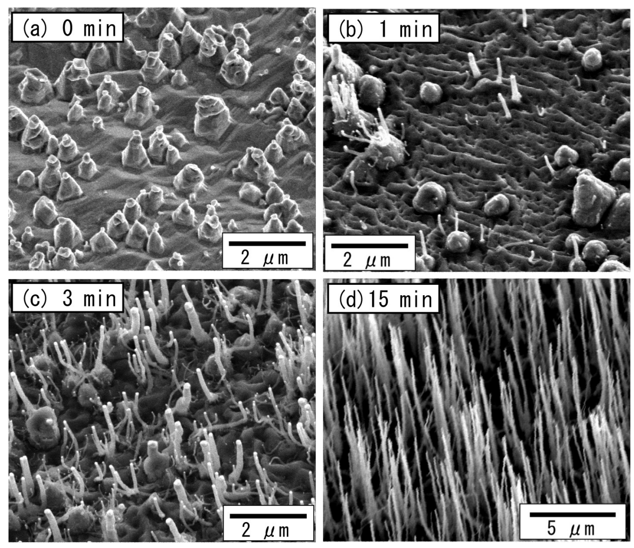

Figure 1. SEM images showing a time evolution of the CNTs growth.

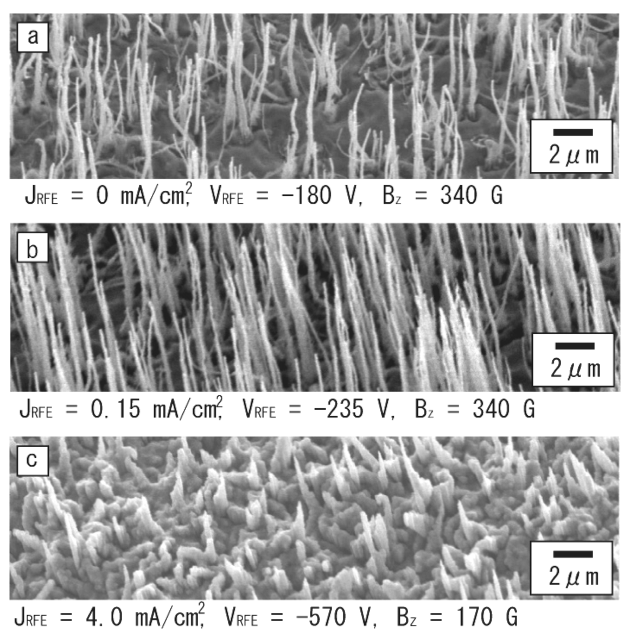

Figure 2. CNTs growing under various conditions.

References
1. H. Ishida et al., Thin Solid Films **407,** 26 (2002).
2. N. Satake et al., Physica B **323,** 290 (2002).