# Preparation of Carbon Nanotubes by CVD using Metal-Phthalocyanines

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As one of the simplest and most effective method for producing multiwalled carbon nanotube (MWNT) on various substrates, we utilized chemical vapor deposition (CVD) in vacuum or $H_2$/Ar gas flow using (Ni, Fe, Co) -phthalocyanines (Pcs), as a source material.

In this study, we examine the influence of the surface of substrate materials such as a quartz plate, metal (Ni, Fe, SUS, Ta and W) foils, porous Si, and the oxidized surface of Al (OS-Al) plate on the growth of nanotubes. Growth of nanotubes on conductive substrate is required for the application of nanotubes as cathode of various devices.

The morphology of the deposites were observed by scanning electron microscope (SEM).

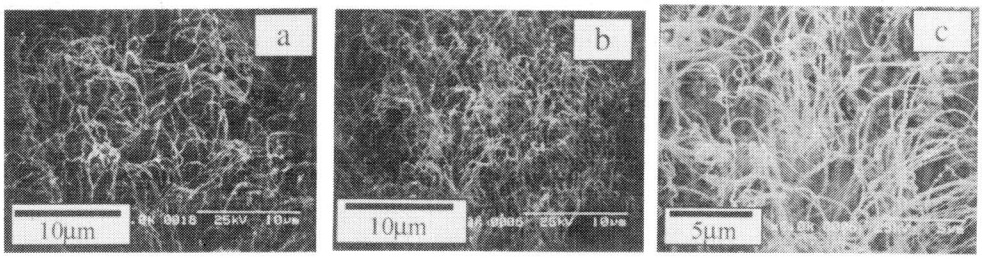


Fig. 1. SEM images of MWNT grown on a quartz plate or (OS-Al) plate using source materials (a) NiPc, (b) FePc and (c) NiPc.

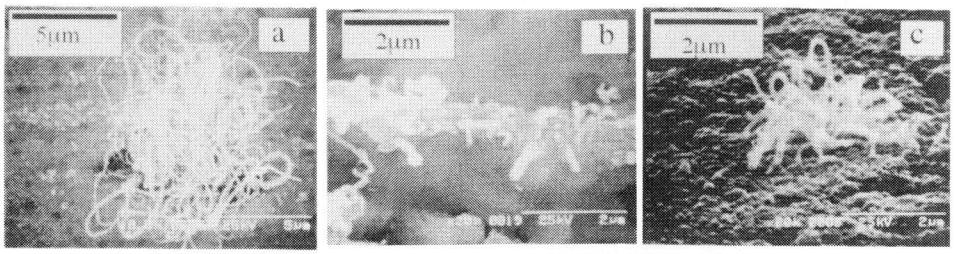


Fig. 2. SEM images of MWNT grown on uneven site of (a) a Ni foil from NiPc source, (b) a Fe foil from FePc source, and (c) a SUS foil from FePc source.

High density of MWNT was obtained on a smooth surface of a quartz plate on a (OS-Al) plate (Fig. 1), certain metal plates, on a uneven sites of metal foils such as Ni, Fe and SUS (Fig. 2), and at scratches on (Ta and W) foils or at their foil edges (Fig. 3).

On the other hand, low density of MWNT was obtained on a smooth surface of Ni, Fe and SUS foils and on a porous Si (Fig. 4).

MWNT was not formed at all on a smooth surface of foils of inactive metal such as Ta and W (Fig. 4).

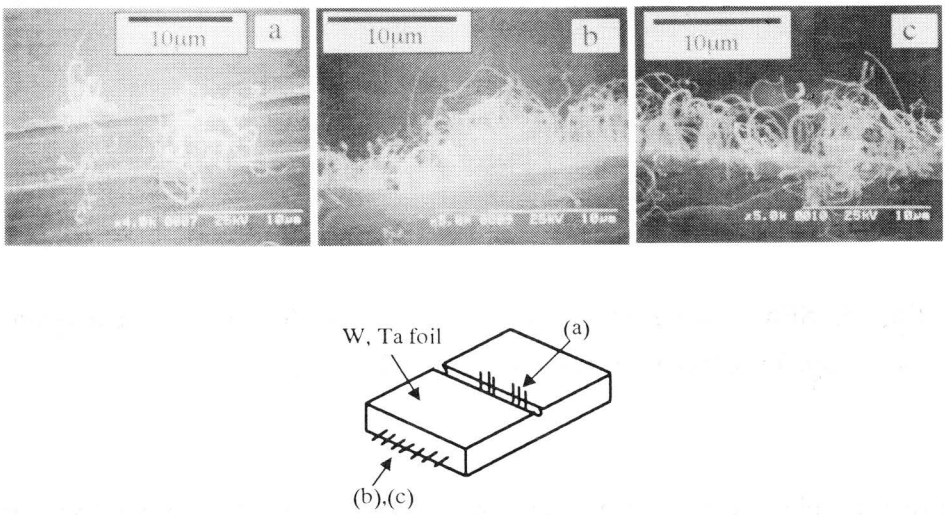


Fig. 3. SEM images of MWNT grown at (a) a scratch on a Ta foil, (b) a Ta foil edge, and (c) a W foil edge.

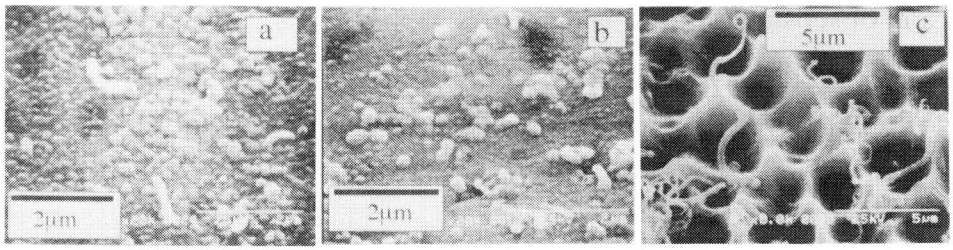


Fig. 4. SEM images of MWNT grown on a smooth area of (a) a Ni foil, (b) a SUS foil NiPc source or (c) on a porous Si from NiPc source.

Furthermore, very attractive phenomena were found that figh density of uniformly alliged MWNT grew at edges of a quartz plate (Fig. 5), when FePc source was used. Uniformly alligned MWNT was also obtained from inner wall of a quartz tube. The inner tube of these MWNT was closely packed with Fe metals at the base.

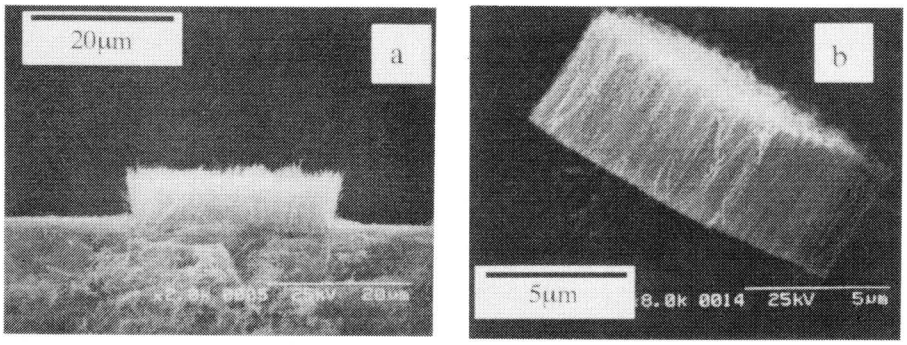


Fig. 5. SEM images of uniformly aligned MWNT grown at a quartz plate edge by CVD in (a) vacuum and (b) $H_2$/Ar gas flow.

In above results, high density of NT was obtained when material with a low surface tension or surface energy was used as substrate. As is well known, Ni, Fe and Co metals act as catalysis. Therefore, above facts suggest that when vapors of these metals impinge onto the substrate from vapor phase, they may aggregate to a nanometer-sized particle with a high contact angle against the substrate because of their hard wetting, and nucleation of MWNT may occur on the particle.

Amount of work done per unit surface area when the substrate is wetted with liquid, W, is expressed by a relation $W = \sigma_S + \sigma_L - \sigma_{SL} = \sigma_L(1 + \cos\theta)$, where $\sigma_S$, $\sigma_L$ and $\sigma_{SL}$ denote the surface tension of substrate, liquid, and interface between substrate and liquid, respectively, and $\theta$ is the contact angle at that interface. Hard wetting is due to a small W, that is, a small $\sigma_S$ and $\sigma_L$ in metal, and a large $\sigma_{SL}$ in the smooth interface. Therefore, the deposition of small metallic particle with high $\theta$ can be expected on the quartz plate, the (OS-Al) plate, and certain metal plate with a small $\sigma_S$.

Even if a metal substrate such as Ta or W foils has a very high $\sigma_S$, the growth of MWNTs can be realized when the surface is structurally modified so that micro-defects of uneven sites are formed.

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