6th World Conference on Experimental Heat
Transfer, Fluid Mechanics, and Thermodynamics
April 17-21, 2005, Matsushima, Miyagi, Japan

Synthesis of Single-Walled Carbon Nanotube Film on Quartz Substrate from Carbon Monoxide

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Keywords *: Single-Walled Carbon Nanotubes, CCVD, Quartz Substrate, Carbon Monoxide*

1. INTRODUCTION

The discovery of single-walled carbon nanotubes (SWNTs) in 1993 by Iijima and Ichihashi [1] has stimulated many researchers to utilize for the innovative technology, especially applications to electronic or optic devices in which SWNTs are used as grown on substrates. However, the technical development for decreasing impurities and defects, controlling physical properties, shapes and sizes, and economical mass-synthesis is necessary for utilizing the unique physical properties of SWNTs [2].

Bulk production techniques of SWNTs were developed as HiPco [3] (decomposing high-pressure CO with floated Fe particles), ACCVD [4] (decomposing alcohol with Co/Fe supported on zeolite), and CoMoCAT [5] (decomposing CO with Mo/Co supported on silica particles). On the other hand, direct synthesis of high-purity SWNTs on silicon and quartz substrates using catalytic chemical vapor deposition (CCVD) technique from alcohol was demonstrated [6]. Later, by the efficient CVD reaction, vertically aligned SWNTs film was also achieved [7]. Recently, Hata et al. [8] demonstrated much more efficient and longer growth of SWNTs on a Si substrate from ethylene. However, physical properties are not fully controlled by these methods.

Electric Power Development Co.,Ltd. (J-Power) is studying on utilizing carbon monoxide gas to the carbon source of carbon nanomaterials such as SWNTs, graphite nanofibers (GNFs), etc. Carbon monoxide gas is produced in numerous industrial plants. Furthermore, carbon dioxide gas is exhausted from most industrial plants as one of Greenhouse gases, and it can be converted to carbon monoxide gas using Bosh reaction [9].

In the present report, SWNTs were synthesized directly on a substrate using a liquid-based approach for the catalytic mount, in combination with the carbon monoxide CCVD method. We here report characteristics of synthesized SWNTs using above method.

2. EXPERIMENT

A detailed description of the catalyst loading process is presented in previous report [6]. Bimetallic Co-Mo catalyst was supported on a quartz substrate by dip-coating the substrate in an acetate solution (i.e. ethanol solution of Co and Mo acetates, each with a metal content of 0.01 wt%).

The schematic of our CCVD apparatus is shown in Fig, 1. It is composed of a gas supplying system, a quartz tube (19 mm i.d., 1 m long), a gas pre-heater, an electric furnace and a gas cooler / drain separator. This apparatus is a one-through open-system, therefore the reaction field is close to atmospheric pressure.

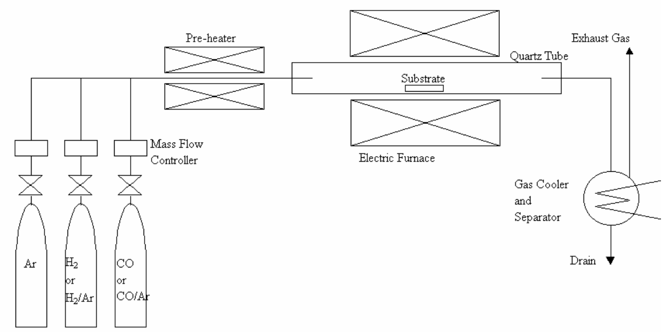

Fig. 1 Schematic of our CCVD apparatus for synthesis of SWNTs

Our experiments consisted of the warming and reducing process by hydrogen gas (1 NLM), the CCVD process by carbon monoxide gas (0.5 NLM) and hydrogen gas (0.5 NLM), and the cooling process by pure argon gas. The purity of hydrogen was 99.99999 % (or 39.9 % balanced argon), one of carbon monoxide was 99.95 % (or 40.1 % balanced argon). Each process patterns are described in Fig. 2.

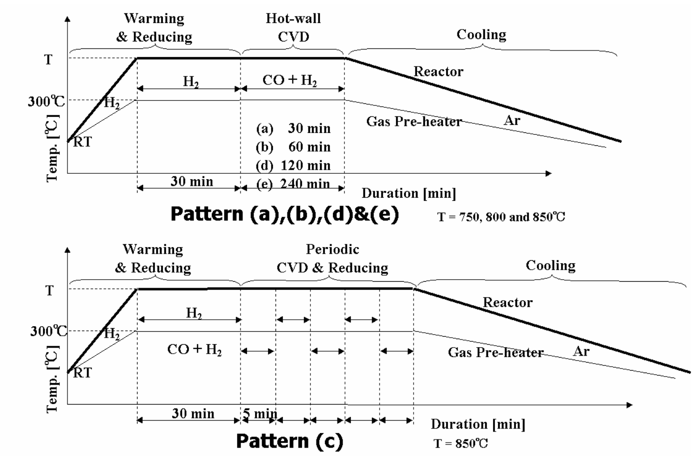

Fig. 2 Process patterns for synthesis of SWNTs

3. RESULTS AND DISCUSSION

3.1 Microscopic analyses

Fig. 3 shows SEM images. The picture (a) shows that SWNTs crept on the substrate, and the (b) shows that SWNTs grew up to more than 1 mm long in 30 min CCVD times.

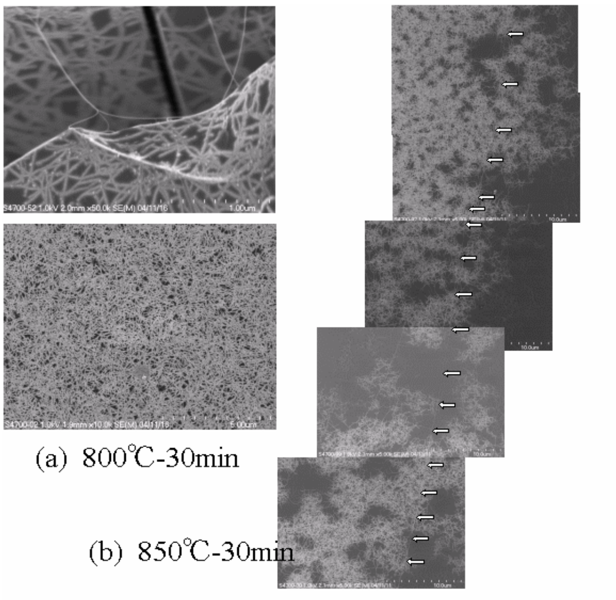

Fig. 3 SEM images of SWNTs

3.2 Raman scattering analyses

The Raman spectra of SWNTs synthesized on quartz substrates are presented in Fig. 4.

Pattern (a), (b), (d) and (e) show that SWNTs grew as CCVD times, but dissociation happened at 240 min. Furthermore, the RBM peak of SWNTs growing stage is limited to about 200 cm^{-1}. This means that diameter of SWNTs is uniform, therefore their physical properties based on the chirality are uniform, too.

On the other hand, there are no peaks in the RBM region of the pattern (c). This means that diameter of SWNTs dispersed by the periodic processing of reducing and CCVD.

4. CONCLUSIONS

SWNTs were synthesized directly on a substrate using a liquid-based approach for the catalytic mount, in combination with the carbon monoxide CCVD method.

SWNTs crept on a substrate, the diameter was uniform and the growth speed was relatively high (1mm over / 30min).

These results suggest that the CCVD method from carbon monoxide have a potential of mass-synthesizing high-quality SWNTs at relative low cost.

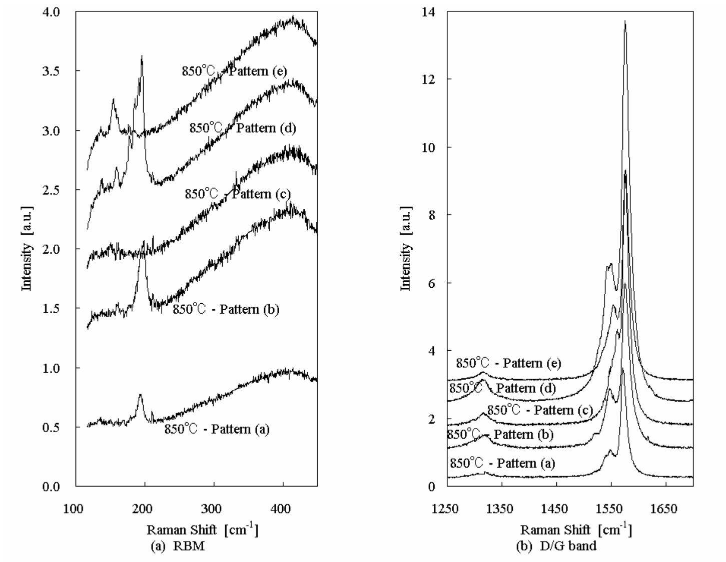

Fig. 4 Raman spectra (taken by 488 nm) of SWNTs

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