Carbon Nanomaterial Synthesis from Sucrose Solution without Using Graphite Electrodes

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Carbon nanomaterials such as carbon nanotubes (CNTs) and carbon nanospheres (CNS) were synthesized from sucrose solution using an arc discharge method without graphite electrodes. In this process, the nanocarbon products were formed from carbon atoms supplied by the sucrose in solution rather than from the graphite of the electrodes. One of the advantages of this method is that the nanomaterials are produced in agglomerated forms, and there are no generated toxic gases during arcing process.

Recent research on carbon nanotube (CNT) production using an arc discharge method in water,[1–8] has used graphite rods as both anode and cathode. The carbon atoms for the carbon nanomaterials such as CNTs, carbon nano-onions, and carbon nanospheres (CNS) so produced were supplied from the graphite of which the rods were composed. Sano et al. were successful in producing related carbon nano-onions using this method.[9] Hsin et al. first presented a novel low-temperature, condensed-phase production of CNTs using carbon plasma produced by an electrical discharge in water.[1] Zhu et al. reported that when using a solution containing several metal and sulphate ions instead of deionized water as the medium, CNTs with a higher purity and yield were obtained.[3]

Carbon nanomaterials are one of the most promising materials currently being investigated owing to their unusual mechanical and electrical properties. Various methods for production of nanocarbon products involving arc discharge, laser ablation and chemical vapor deposition (CVD) have been employed. In almost all of the research employing an arc discharge method, the nanomaterials produced are composed of carbon atoms supplied from the graphite electrodes. According to Shibata et al.'s report,[10] nanosized carbon particles could be prepared in ultrasonically irradiated liquid benzene, using an arc method without graphite electrodes. The products formed in the volatile solvent were, however, amorphous carbon (a-C). This method has the added disadvantage of employing the dangerously inflammable benzene as the production medium.

This paper introduces a novel method for the synthesis of carbon nanomaterials including CNTs and CNS where the carbon is supplied from sucrose in solution rather than from the graphite of the electrodes. One of the advantages of this method is that the nanomaterials are produced in agglomerated forms generally free from other contaminants. On the other hand, arc discharge methods using graphite electrodes generate a-C around the nanocarbon products. Although a-C was also generated using the sucrose method and distributed thoughout the solution, most of the agglomerated nanocarbon products were a-C free. Since the molecular composition of readily soluble sucrose is $C_{12}H_{22}O_{11}$, it is able to provide a vast number of carbon atoms without any contaminants other than water molecules. Furthermore, even highly refined sucrose is generally cheaply available, and there are no toxic gases generated during the arcing process.

Figure 1 shows a schematic diagram of the experimental apparatus. Sucrose (Wako Pure Chemical Industries, Ltd.) was dissolved in boiled distilled water (Otsuka Pharmaceutical Co., Ltd.) in a glass container, and 160 mL of 1.64 M (mol/L) sucrose solution was prepared. Copper rods (5-mm diameter, 30-mm long) were submerged in the container and then connected to a DC power supply able to provide a potential difference of 30 V and a continuous current discharge of 20 A. The gap between the electrodes was set to less than 0.5 mm, and the apparatus was allowed to operate for an hour. While the equipment was operating, carbon soot production was visible between the rods. The anode was vaporized at the rate of about 3 g/h. It is supposed that this generation was a result of carbon atoms condensed from the hot plasma produced from the sucrose between the electrodes. During the arcing, some of the electrode copper was vaporized, and the material was subsequently precipitated in the solution mixed with the carbon soot. These copper particles were not removed prior to microscopic investigation. The products in the container were filtered using a filter paper, and the filter paper was then air dried at room temperatures for 2 days. The resulting residue was then observed using a Hitachi H-9500 transmission electron microscope (TEM) with an operating voltage of 200 kV.

Figure 2a shows the low-magnification TEM image of the agglomerated products. All products in Figure 2a are graphitized and have hollow inside the products. When carbon nanomaterials were synthesized by arc discharge using graphite rods as both electrodes, almost all of the nanomaterials were surrounded with a-C, as well as with CNS. However, when nanocarbon

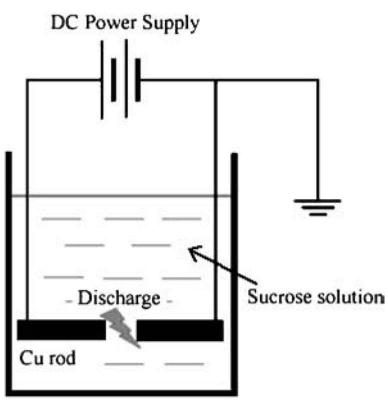

Figure 1. Schematic diagram of the apparatus for arc discharge in sucrose solution.

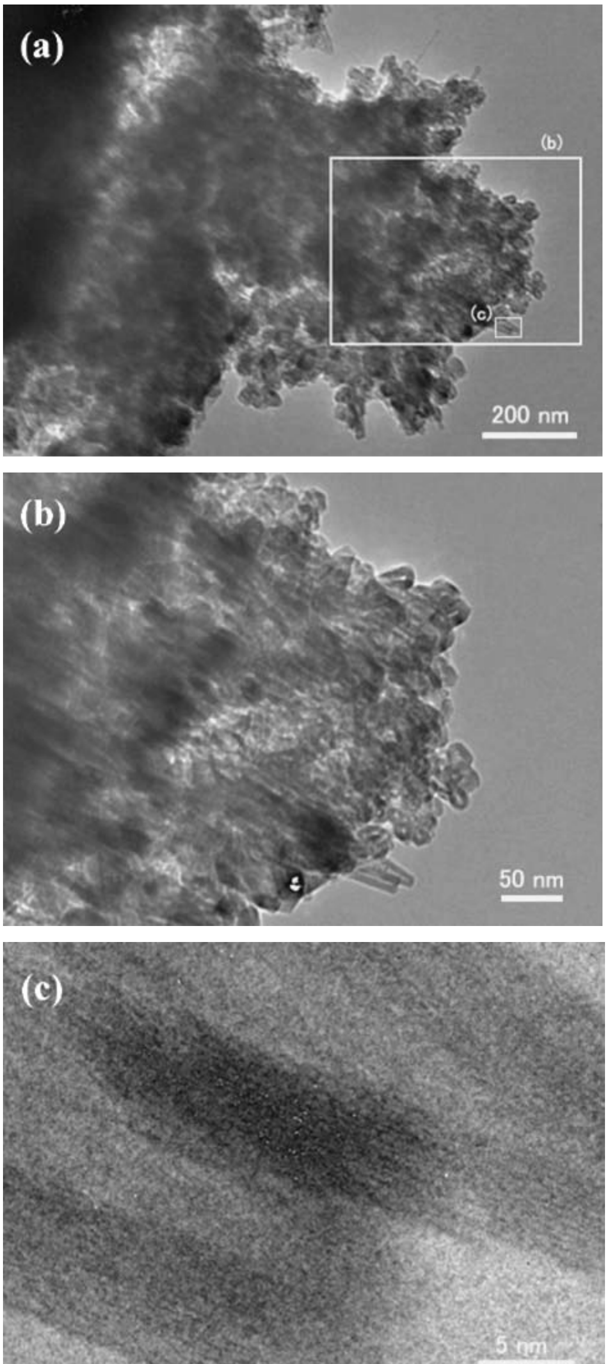

Figure 2. TEM images of nanocarbon products produced by arc discharge in sucrose solution. (a) Low-magnification image of the uncontaminated agglomerated products. (b) and (c) High-magnification image of the rectangle areas in Figure 2a. Figure 2b shows the agglomerated nanocarbon products, and Figure 2c is multilayered forms of the CNTs.

products were prepared using the method described in this paper, no such contamination occurred around the nanocarbon products. The lengths of the CNTs which accompanied the nanocarbon products are somewhat less than those produced using graphite rods. A report by Sano et al.[2] described the ionization of carbon atoms generated in hot plasma condition and the subsequent sublimation to form nanotubes. The plasma generated using carbon electrodes reaches a temperature of over 3000 K. A lower temperature of about 2000-K plasma was created using Cu electrodes, resulting in shorter nanotubes.

Figures 2b and 2c are high-magnification TEM images of the rectangle areas in Figure 2a. Figure 2b shows the agglomerated nanocarbon products. In this area, it seems the majorities are CNS, while Figure 2c is multilayered forms of the shorter CNTs.

In summary, it has been found that carbon nanomaterials such as CNTs and CNS can be synthesized from sucrose solution without using graphite electrodes. This letter describes production using only Cu rods as electrodes; however, there is nothing to suggest that other metals may be also employed in nanocarbon production. It is also assumed that aqueous solutions of other substances containing carbon could be used as a source for carbon atoms in carbon nanomaterials in place of sucrose.

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References

1 Y. L. Hsin, K. C. Hwang, F.-R. Chen, J.-J. Kai, *Adv. Mater.* **2001**, *13*, 830.
2 N. Sano, H. Wang, I. Alexandrou, M. Chhowalla, K. B. K. Teo, G. A. J. Amaratunga, *J. Appl. Phys.* **2002**, *92*, 2783.
3 H. W. Zhu, X. S. Li, B. Jiang, C. L. Xu, Y. F. Zhu, D. H. Wu, X. H. Chen, *Chem. Phys. Lett.* **2002**, *366*, 664.
4 H. Lange, M. Sioda, A. Huczko, Y. Q. Zhu, H. W. Kroto, D. R. M. Walton, *Carbon* **2003**, *41*, 1617.
5 X. Li, H. Zhu, B. Jiang, J. Ding, C. Xu, D. Wu, *Carbon* **2003**, *41*, 1664.
6 K. Imasaka, Y. Kanatake, Y. Ohshiro, J. Suehiro, M. Hara, *Thin Solid Films* **2006**, *506–507*, 250.
7 Y. Yao, R. Wang, D. Wei, D. Du, J. Liang, *Nanotechnology* **2004**, *15*, 555.
8 L. P. Biró, Z. E. Horváth, L. Szalmás, K. Kertész, F. Wéber, G. Juhász, G. Radnóczi, J. Gyulai, *Chem. Phys. Lett.* **2003**, *372*, 399.
9 N. Sano, H. Wang, M. Chhowalla, I. Alexandrou, G. A. J. Amaratunga, *Nature* **2001**, *414*, 506.
10 E. Shibata, R. Sergiienko, H. Suwa, T. Nakamura, *Carbon* **2004**, *42*, 885.

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