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Continuous Production of Nanotubes and Carbon Clusters by Use of the Revolver-Injection-Type Arc Jet Producer

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Abstract

Efficient and continuous production of fullerenes, single-walled carbon nanotubes (SWNTs), and carbon nano-particles has been developed by the JxB arc jet method, where the Lorentz force of magnetic field jets out gaseous carbon particles to the JxB direction. In order to realize continuous mass production of these carbon clusters, the revolver-injection-type arc jet producer (RIT-AJP) has been developed. Production of fullerenes, endohedral metallo-fullerenes, SWNTs, metal-encapsulated carbon nano particles has been examined.

Abstrak

Produksi Tabung Nano dan Gugus Karbon secara Terus-Menerus dengan Menggunakan Penghasil *Arc Jet* Tipe-Injeksi-Revolver. Produksi fulerena, tabung nano karbon berdinding tunggal (SWNT), dan partikel nano karbon secara efisien dan terus-menerus telah dikembangkan dengan metode *arc jet* JxB. Di dalam metode ini, daya Lorentz pada medan magnet menyemburkan partikel-partikel karbon gas ke arah JxB. Agar dapat memproduksi gugus-gugus karbon ini secara massal dan terus-menerus, penghasil *arc jet* tipe-injeksi-revolver (RIT-AJP) telah dikembangkan. Produksi fulerena, metalo-fulerena endohedral, SWNT, dan partikel-partikel nano karbon yang terenkapsulasi metal telah diamati.

Keywords: automatic and continuous production, carbon cluster, carbon nanotube, JxB arc jet discharge

1. Introduction

After the discovery of the mass production methods of fullerenes and single-walled carbon nanotubes (SWNTs) [1,2], applications of these carbon clusters such as transistor devices, bio-sensing devices, double-layer-type capacitors, transparent electrode films, EMS absorbers, material hardener etc. have been studied [3]. However, efficient production of high-quality and defect-free SWNTs, and metal/semiconductor selected or diameter-controlled production of SWNTs have not been achieved. Therefore, I think that basic study of various production methods of SWNTs and carbon clusters is still important, by which new excellent roots to produce desired carbon clusters will be found.

Here, we have studied about continuous production of fullerenes, SWNTs and carbon nano particles by the arc discharge method including magnetic field, which is called "the JxB arc-jet discharge method" [4]. Continuous mass production of carbon clusters by this method has been developed by our group. As a result, more efficient production compared with usual arc discharge methods has been obtained [5].

By applying a weak magnetic field (B0= 10 m ~ 50 mT) perpendicular to the discharge current in the arc discharge, the Lorentz force (JxB force) jets out the arc plasma and surrounding gas to the JxB direction as shown in Figure 1. By this effect, sublimated carbon

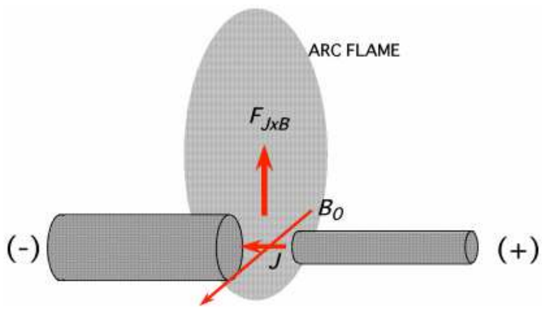

Figure 1. Model Figure of the JxB arc Jet

atoms are efficiently jetted out to the selected direction, and good efficiency of the production can be obtained reducing the loss of carbon material. This method also would reduce the effects of electrode direction and chamber configuration. Continuous production has been developed by utilizing revolver-type carbon stock and computer control.

2. Experiment

A schematic of the developed reactor (RIT-AJP) is shown in Figure 2. The left side of the machine is an arc discharge chamber, which consists of a vacuum vessel made of stainless steel 25 cm in diameter, 70 cm high and is uniformly cooled by so called "water jackets". About 5 L of water is stored in the jackets and total flow rate of water is about 2 L/min. At the central part of the chamber, a cathode electrode (30 mm in diameter), an anode electrode, an exhausting port, a viewing port and an electrode-cleaning hand are mounted. Top and bottom parts of the chamber are soot collectors, inner diameter of which is about 25 cm and 24 cm high, where discharged soot deposits and it can easily be collected after operation. Using these collectors, as much as 25 L of soot can be collected at one operation. The right side of the apparatus is a revolver type carbon rod magazine. In the cylindrical metal vacuum vessel, 34 cm in diameter and 49 cm long, there is a rotatable cylindrical magazine, in which 50 carbon rods 6-10 mm in diameter, 300 mm long can be loaded. A photo of this machine is shown in Figure 3. A schematic figure of the material feeding mechanism using the material magazine is shown in Figure 4, and a photo of the rotatable carbon rod magazine (for 50 carbon rods) is also shown in Figure 4. Under the vacuum chamber, there is a vacuum pump, an electric-controlling circuit and a micro-computer. Discharge power is supplied by an inverter type DC power supply (Daihen Co., ARGO-300P).

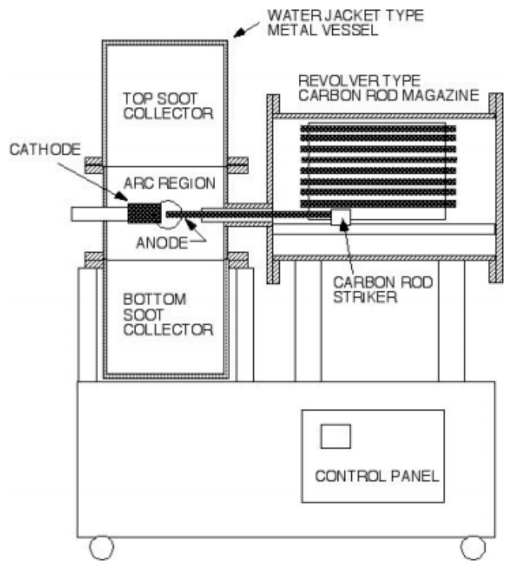

Figure 2. Schematic of the Revolver-injection-type JxB Arc Jet Producer (RIT-AJP)

A sequence of the production is as follows. At first, as much as 50 carbon rods are loaded in the magazine and evacuated by the vacuum pump. After sufficient evacuation less than 1 Pa, pure helium gas is introduced and the chamber is closed. By turning on the electrical

Figure 3. Photo of RIT-AIJ

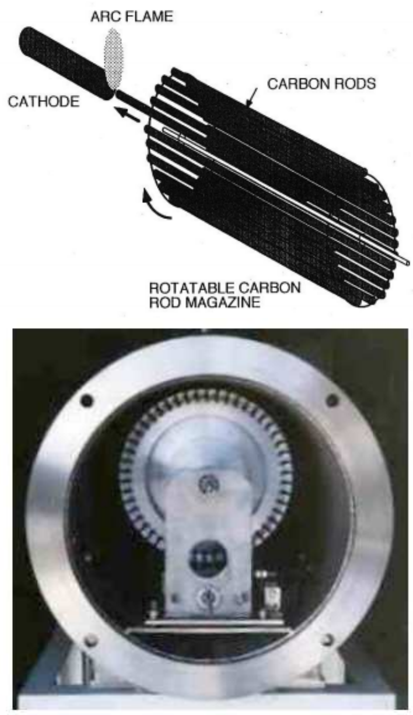

Figure 4. The Feeding Mechanism and a Photo of the Rotatable Carbon Rod Magazine

controller, a metal striker pushes one carbon rod towards the cathode, and by electrical contact the discharge starts. As the discharge condition is decided by discharge voltage and discharge current, a carbon rod is automatically moved until the both parameters match set values. When one carbon rod is consumed, the cylindrical carbon magazine rotates 1/50 rotation and the next carbon rod is inserted by the striker. A magnetic field can be applied by a block-type ferrite magnet located outside of the chamber, by which about 2.5 mT of magnetic field is applied horizontally in the discharge space. Deposition of carbon on the cathode, is removed by a cathode-cleaning hand. After the discharge, produced soot deposited is carefully collected.

Mass distribution of the produced carbon clusters is measured by a laser desorption time-of-flight mass spectrometer (LD-TOF-MS) (Shimadzu Co., Kratos Kompact MALDI III and Bruker Co., Autoflex). Production of SWNTs is analyzed by a TEM (Hitachi Hi-Tech. Co., H-7500 etc.) and a Raman spectrometer (Jasco Co., NR-1800). Content of fullerenes is measured by a UV/visible spectrometer (Jasco Co., V-630) and by a high pressure liquid chromatograph (HPLC) (Jasco Co., Gulliver Series, PU980).

3. Results and Discussion

The JxB arc discharge. The effect of a steady magnetic field in the arc discharge is already shown in Ref. 6, in which deposition rate of carbon vapor on a cathode decreases a half value and production rate of C60 increases by 1.5 times. Change of the arc discharge by the magnetic field can be observed by naked eyes. Photos of the arc flames (side view) are shown in Figure 5. By applying the magnetic field, the arc flame extends toward upper direction, which is a clear evidence of the plasma jet acceleration by the JxB force.

Production of C60. The production rate of C60 is examined in this reactor, by consuming 134 carbon rods in continuous JxB arc discharge, where p(He)= 40 kPa, discharge current Id= 120 A and voltage between electrodes, Vrod~ 33 V, the gap distance dG~ 5 mm. Insertion speed of the carbon rods is about 30 cm/h. After the discharge, carbon soot from three parts (top collector, center chamber and bottom collector) is collected separately and their weights are measured. Amount of soot deposited on the top wall considerably increases by applying the magnetic field, because the carbon molecules are blown up to the top wall. After sufficient mixing, C60 content in the soot is measured by the UV/visible spectrometer. At the top collector, C60 content is the highest and about 14 w% of C60 presents, whereas, 4.2 w% at the center wall and 2.9 w% at the bottom wall. In total, about 105 g of soot is produced in 12 h operation and it includes about 7 g of C60 in the soot.

Contents of higher fullerenes in the soot are measured by using the HPLC [5]. Contents of C76, C78, C82 and C84 relative to the C60 content are compared as shown in Figure 6. By applying the magnetic field, yields of the higher fullerenes considerably increase.

Production of metallo-fullerenes. The production of endohedral metallo-fullerene [7] is examined using the JxB arc discharge method. Carbon rods including La2O3 (9 w% of La atom included) are used as a carbon material, and fullerenes are produced in the same way, where Id= 50 A. After Soxhlet extraction of the soot by xylene for 3 h, fullerene content is measured by the HPLC, as shown in Figure 7, where eluent is xylene, 1 mL/min. There is a strong peak corresponding to La@C82, mass of which is confirmed by the LD mass spectrometer. However, La@C82 content is estimated to be around 0.1 w% in the soot. The optimum production condition of the endohedral metallo-fullerenes (La@C82 and Gd@C82) by this reactor is under investigation.

Production of SWNTs. This reactor can produce a large amount of carbon nano-tubes (SWNTs). When carbon material rods including 4.2 w% of Ni and 0.9 w% of Y (6 x 6 mm rectangular type) are used, and p (He) = 60 kPa, Id= 40-50 A, SWNTs are effectively produced by the JxB arc discharge method [8]. The

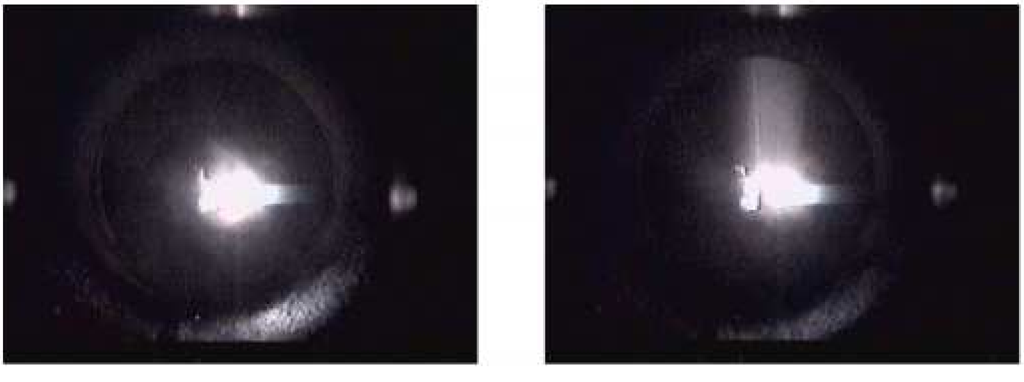

Figure 5. Arc Flames for B0= 0 (a) and B0= 2.0 mT (b) (Side View), where p(He)= 40 kPa and Discharge Current Id= 80 A

production rate of the soot is 1.0 g/h for B0= 0 mT, while it is 2.5 g/h for B0= 3.0 mT, which also shows the effect of the magnetic field. Typical TEM image of the produced SWNTs by this method is shown in Figure 8, in which bundles of long SWNTs, catalyst particles and carbon particles are observed. Though the SWNTs are under cotton-like condition, SWNTs content is high. Typical Raman spectra for the sample of B0= 2.1 mT are shown in Figure 9. The G band/D band ratio is very high, which means good quality of SWNTs. Serious reduction of this ratio by applying the magnetic field is not observed. The diameters of SWNTs are 1.40, and 1.26 nm from the radial breathing mode in Figure 9. The diameters of SWNTs are consistent with those observed in TEM images.

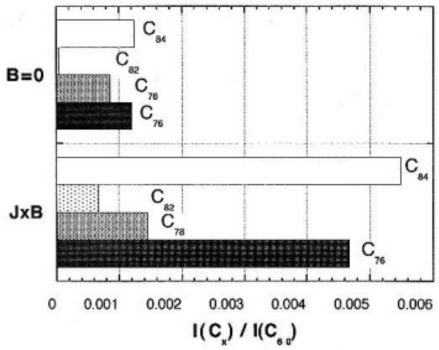

Figure 6. Relative Content of Higher Fullerenes Relative to C60 Content. p= 40 kPa, Id= 120 A

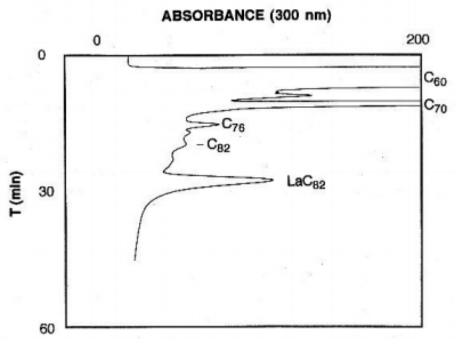

Figure 7. The HPLC Chart of Fullerenes Extracted from the Soot by Xylene. Wave Length of 300 nm

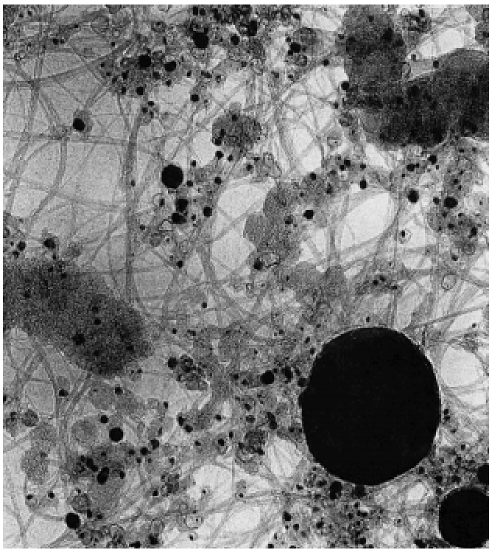

Figure 8. TEM Image of Produced SWNTs

Production of metal-encapsulated carbon particles. When metal powder is included in the carbon material rods, many kinds of metal-encapsulated carbon nano particles can be produced by this arc discharge method.

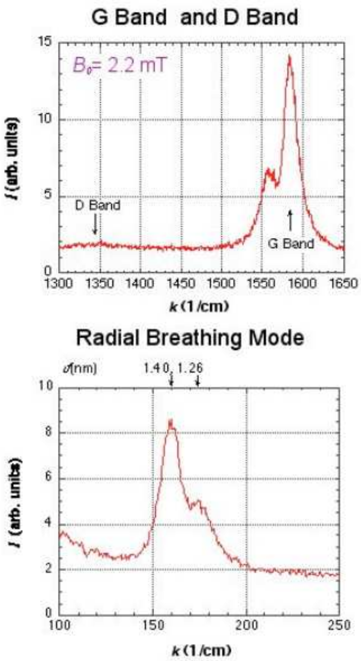

Figure 9. Raman Spectra of the Soot Including SWNTs for B0= 2.2 mT, p(He)= 60 kPa, Id= 50 A. The Diameters of SWNTs are Written in the Figure of Radial Breathing Mode

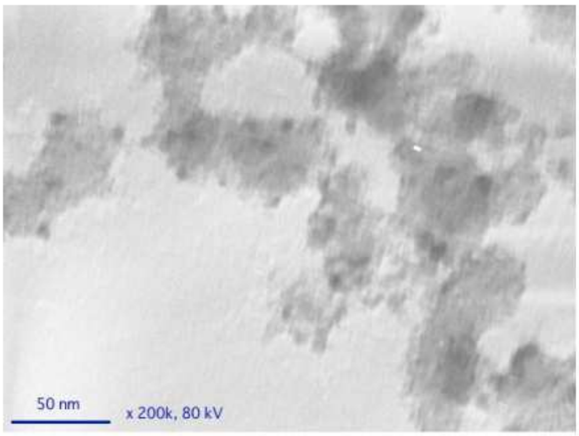

Figure 10. TEM Image of Gd Included Carbon Particles

For example, Gd-included carbon nano particles are produced, where 6x6 mm rectangular rods are used p(He)= 30 kPa, Id= 58 A. Production rate of the soot is about 2.4 g/h. Typical TEM image of the soot is shown in Figure 10. In the case of gadolinium (Gd), size of Gd particles is small (5-10 nm), and the amorphous carbons (20-50 nm of size) include Gd particles. Magnetic nano particles like Co particles are also effectively produced by this method. In this case, the size of metal is much larger and spherical. These nano magnets are very stable and have potential applications.

4. Conclusions

The JxB arc jet producer with a revolver-injection type automatic material injector has been successfully developed. By this reactor, as much as 50 carbon rods are arc-sublimated into carbon soot by the DC discharge automatically and continuously. As well as production rate of C60, the content of higher fullerenes such as C76, C78, C82 and C84 in the soot is considerably increased. By this reactor, metallo-fullerenes like La@C82, which have endohedral structure, are also produced efficiently. SWNTs and metal-encapsulated carbon nano particles are also effectively produced. It is expected that this reactor would be useful for production of many kinds of carbon clusters to develop new engineering materials.

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