Advanced Materials '93, I / B: Magnetic, Fullerene, Dielectric, Ferroelectric,
Diamond and Related Materials, edited by M. Homma et al.
Trans. Mat. Res. Soc. Jpn., Volume 14B

Continuous production of fullerenes by evaporation of carbon powder and carbon chips in an arc discharge

T. Mieno, T. Asano and A. Sakurai

Department of Physics, Shizuoka University, Shizuoka 422 Japan

In order to produce fullerenes efficiently, continuous supply of carbon raw material is necessary. Here, a new apparatus is developed, which utilizes carbon chips, carbon powder and discharged soot as raw materials, and produces fullerenes continuously in an arc discharge by supplying the raw material without breaking the discharge atmosphere.

1. INTRODUCTION

The arc discharge method is convenient to produce many kinds of fullerenes. [1, 2] Length of the carbon rod, however, is shortened by the discharge and we must break the discharge atmosphere to change the carbon rod, which makes continuous production of fullerenes very difficult. This led to our study which uses carbon chips, carbon powder and discharged soot as raw material to produce fullerenes. As a positive electrode, a metal oven filled with carbon material is used and the arc discharge between the oven and a carbon rod (negative electrode) is carried out, where the discharge condition is automatically kept almost constant by using a controlling system. [3] The carbon raw material can be supplied quickly by dropping it into the oven from a material stock without breaking the discharge atmosphere. For all of the materials; carbon chips, carbon powder and the discharged soot, the production of the soot with high C_{60} content is obtained, and continuous and mass production of many kinds of fullerenes is expected by this method.

2. EXPERIMENTAL SETUP AND METHOD

Schematic of the experimental setup is shown in Fig. 1. A stainless steel vessel 184 mm in diameter and 400 mm high (made of two 200 mm high vessels) is at first evacuated by a diffusion pump to less than 10^{-3} Torr and filled with 300 - 500 Torr of helium gas. Then, the chamber is closed from the pump and a gas feed. A positive electrode is a metal oven 72 mm in diameter and 60 mm high, which is filled with a carbon raw material, and is insulated from the vessel. Here, the raw materials are cylindrical carbon chips, 6.5 mm in diameter and length l= 2, 5 and 10 mm, 3 mm x 3 mm x 3 mm triangle-prism style chip, carbon powder with size of about 0.1 mm, and discharged soot, from which C_{60} is already extracted. A negative electrode is a carbon rod 6.5 cm in diameter and about 20 cm long and is connected to a metal pipe, which is pushed in or pulled out by a motor drive system. Applying DC voltage between the two electrodes, an arc discharge is carried out (discharge voltage V_d= 15 - 50 V, discharge current I_d= 50 - 100 A). By measuring the

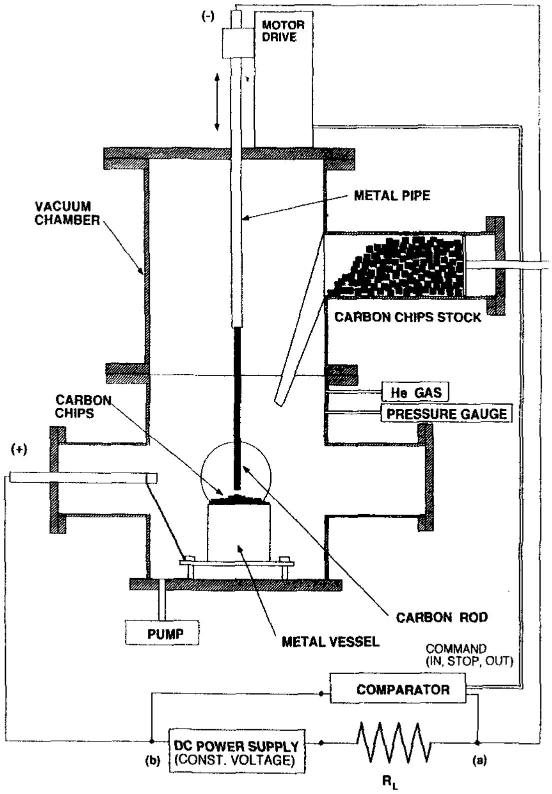

Figure 1. Schematic of experimental setup.

voltage between the two electrodes, a comparator automatically pushes in or pulls out the negative electrode to keep the discharge condition constant. [3] The carbon raw material is dropped into the oven from a carbon material stock by pushing it using a piston rod, when the carbon material in the oven is consumed enough. Produced soot is deposited on inner-wall of the chamber and is collected after the discharge. Its weight W_{soot} is measured (the soot is separately collected at the upper flange, the upper side vessel, the lower side vessel and the bottom). The soot is sufficiently mixed and 1 mg of it is dissolved in 7 ml hexane. After filtration, the UV/visible spectrum of the absorbance I_{ab} of the solvent is measured to obtain C_{60} content in the soot. [4, 5] In this measurement, the absorbance peak of C_{60} at λ= 329 nm [1] is adopted to decide the C_{60} content. The production rate of C_{60} is obtained from the product of W_{soot} x I_{ab}. [5] Total sum of W_{soot} x I_{ab} for 4 positions in the vessel wall is adopted to show the total production rate.

3. RESULT AND DISCUSSIONS

At first, the discharge characteristic of this new apparatus is measured. Figure 2 shows discharge voltage V_d as a function of gap distance d_G, where I_d= 80 A, p= 400 Torr and carbon material of l= 10 mm-chip. V_d increases with an increase in d_G. This d_G dependence is almost same as that of the ordinary arc discharge configuration. [4]

The UV/visible spectrum of absorbance I_{ab} for the solvent is measured to obtain C_{60} content. Figure 3 shows spectrum for the soot deposited on the upper flange, where $V_d \approx$ 25 V, I_d= 80 A, p= 400 Torr, and carbon material of l= 10 mm-chip. We can confirm

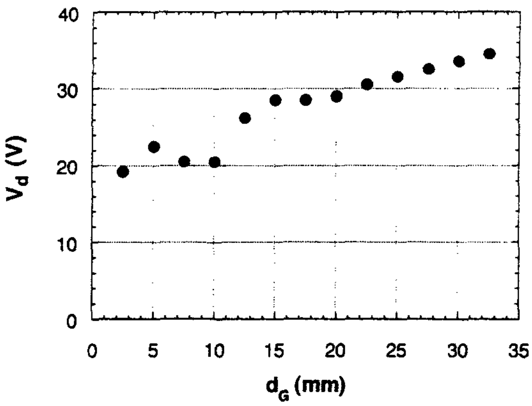

Figure 2. Discharge voltage V_d versus gap distance d_G. Discharge current I_d= 80 A, helium pressure p= 400 Torr and carbon material, l= 10 mm-chip.

the three peaks for C_{60} and small C_{70} peak. Therefore, C_{60} and C_{70} are successfully produced by this apparatus and the quality of the spectrum is as good as that of the ordinary arc discharge. [4]

As mentioned in the section 2, 6 kinds of carbon materials are used for the C_{60} production. Figure 4 shows the total soot weight W_{soot} obtained by the discharge for each material, where V_d=20 - 30 V, $I_d \approx$ 80 A p= 400 Torr, d_G= 5-10 mm and the discharge time T_d= 4 hr. It is interesting that W_{soot} increases with the size of the chips. For the carbon powder, W_{soot} is twice as large as that for l= 10 mm-chip. It is conjectured that part of the carbon is not evaporated as a carbon atom but only blown up as a carbon powder by strong convection of the helium gas.

C_{60} content at the upper flange for each source materials is measured by the UV/visible spectrometer and the absorbance at λ= 329 nm I_{ab} is shown in Fig. 5. Except for the powder, the C_{60} contents are almost the same each other and are also almost same as that for the ordinary arc discharge. [5] From the measurement for pure C_{60} sample, 0.1 scale of the abscissa corresponds to the C_{60} content of 2.2 %. Thus, for the discharged soot, the data show that the soot contains 12 % of C_{60}. For the powder, it is conjectured that the blown up carbon powder also deposits on the wall, which reduces the C_{60} content in the soot. It is also necessary to consider about impurity gas evaporated from the raw material. Especially, the powders and the discharged soot have large surface area, which can absorb the air, water and the solvent, and the material should be warmed up in a vacuum before the discharge to reduce this effect.

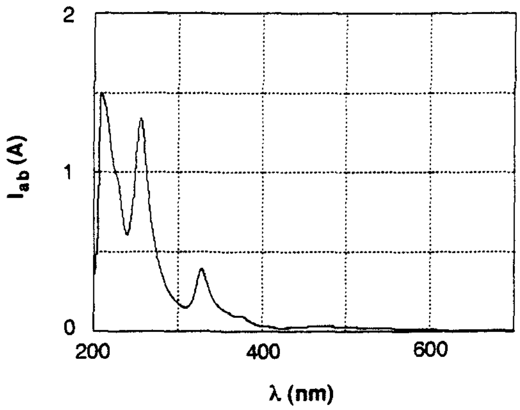

Figure 3. UV/visible spectrum of the absorbance of the soot. Production conditions; $V_d \approx$ 25 V, I_d= 80 A, p= 400 Torr, discharge time T_d= 4 hr and carbon material, l= 10 mm-chip.

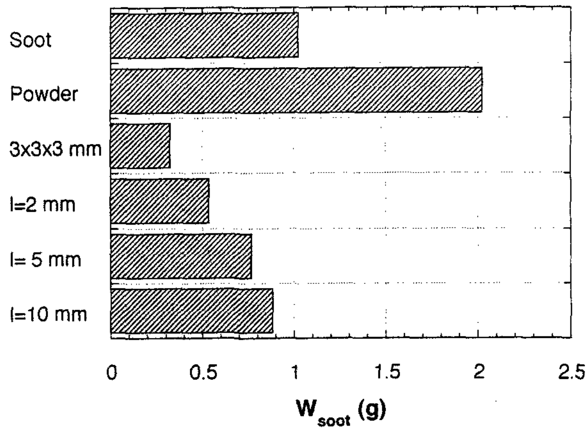

Figure 4. Total soot weight W_{soot} for 6 kinds of carbon materials. Discharge conditions; $V_d \approx$ 20 - 30 V, $I_d \approx$ 80 A, p= 400 Torr and T_d= 4 hr.

From the results in Fig. 4 and Fig. 5, the total production rate of C_{60} (total sum of I_{ab} x W_{soot} measured for 4 places in the vessel) is obtained for each material and shown in Fig. 6. The total production rate for the discharged soot, l= 5 mm-chip and l= 10 mm-chip are almost same and high. For

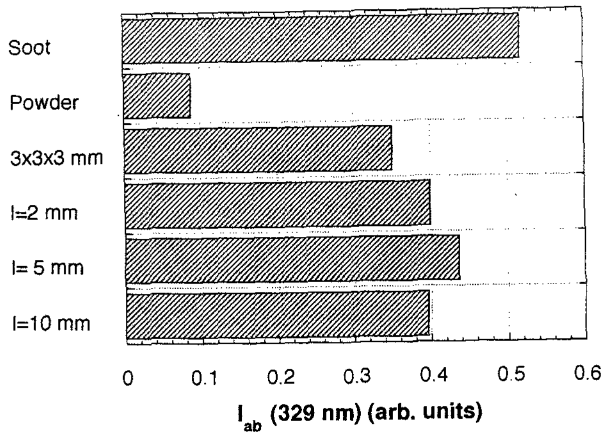

Figure 5. C_{60} content (absorbance I_{ab}) in the soot obtained at the upper flange for 6 kinds of carbon materials. V_d= 20 - 30 V, $I_d\approx$ 80 A, p= 400 Torr and T_d= 4 hr.

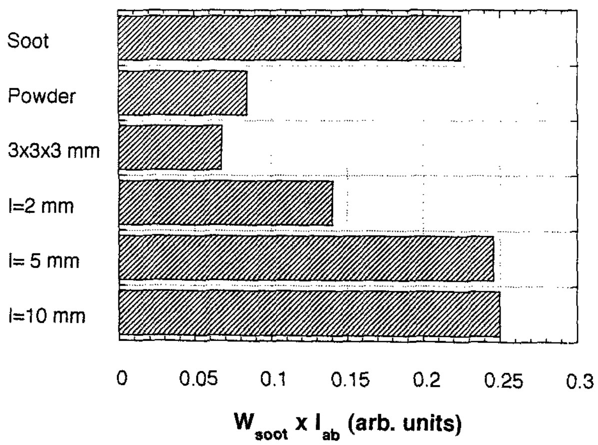

Figure 6. Total production rate (W_{soot} x I_{ab}) for 6 kinds of carbon materials. V_d= 20 - 30 V, $I_d\approx$ 80 A, p= 400 Torr and T_d= 4 hr.

the evaporation rate is larger for the larger chips. For the discharged soot, it is expected that, as the material has cage-like structure in part, it is easy to evaporate and construct fullerenes, which may has new production process of fullerenes. From the measurement of pure C_{60} sample, 0.1 of horizontal scale corresponds to 48 mg of C_{60}. As the discharge time is 4 hr, the production rate for l= 10 mm-chip is about 30 mg/hr in this experiment and is one order smaller than ordinary arc discharge. [5] As the reason is attributable to the low production rate of the soot, further investigation is necessary to increase production rate of the soot.

When the carbon raw material in the oven is consumed, the material is dropped successfully into the oven and the discharge can be continued.

4. CONCLUSION

C_{60} is produced by using the carbon chips, the discharged soot and the carbon powder as raw material. During these materials, l= 10 mm and l= 5 mm cylindrical carbon chips and the discharged soot have high production rates of C_{60}. The carbon material can be supplied by dropping it from the material stock without stopping the discharge, which enables us the continuous C_{60} production.

REFERENCES

1. J. P. Hare, H. W. Kroto and R. Taylor Chem. Phys. Lett. 177 (1991) 394.
2. R. F. Curl and R. E. Smalley, Sci. Am., Oct. (1991) 32.
3. T. Mieno, J. Phys. Soc. Jpn., (1993) (submitted).
4. T. Mieno et al., Rep. Fac. Sci., Shizuoka Univ. 26 (1992) 17: J. Plasma and Fusion Res. 69 (1993) (to be published in Japanese).
5. Y. Miyauchi and T. Mieno, Proc. 6th Sympo. Plasma Sci. Material (SPSM-6) Tokyo (1993) (to be published).