Original Paper

Parameter Dependence of C_{60} Production by Means of the $J \times B$ Arc Discharge Method

Md. Khairul HASSAN BHUIYAN and Tetsu MIENO†

Department of Physics, Faculty of Science, Shizuoka University
836 Ooya, Shizuoka-shi 422-8529
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Abstract: Parameter dependence of production rate of carbon soot including fullerenes, C_{60} content and production rate of C_{60} prepared by the $J \times B$ arc discharge method is investigated. The production rate of C_{60} is optimum at 40 kPa of He pressure, 120 A of discharge current and arc gap distance of about 5 mm, when an 8 mmϕ graphite rod is used as a source material.

Key words: fullerene, C_{60}, the $J \times B$ arc discharge method, optimum production rate, UV-absorbance, C_{60} content.

Introduction

The arc sublimation of carbon rods in helium gas is successful method for mass production of fullerenes [1-2]. But the content and the production rate of C_{60} along with those of higher fullerenes are insufficient by the conventional arc technique. In this connection, the $J \times B$ arc discharge fullerene reactor has been developed to produce fullerenes efficiently [3-4]. Though there are many studies about fullerenes production [5,6], clear reports about the parameter dependence are few. There are many production parameters which are important factors for the production of C_{60} and higher fullerenes, and they are not independent each other [7]. Therefore, exact control of the high temperature chemical reaction in gas phase [8,9] is difficult in the usual arc discharge method. However, efficient increase of the production rate is possible by setting discharge parameters, such as gas pressure, current density, gas temperature gradient and

†E-mail: sptmien@ipc.shizuoka.ac.jp

gap distance. Present study is aimed to find out the optimum method for the production of C_{60} by means of the $J \times B$ method where a small amount of sampled soot is used to determine the C_{60} content and C_{60} production rate by the UV/Vis absorption measurement. Production properties of higher fullerenes by this $J \times B$ arc method are published elsewhere [10].

Experimental Procedure

A schematic diagram of the experimental setup (RIT-FP machine) is shown in Fig. 1. This consists of three parts, (a) a cylinder type arc discharge chamber of 27 cm in diameter and 70 cm high made of stainless-steel which is cooled by a water-jacket type cooling system, (b) a revolver type carbon rod magazine of 35 cm in diameter and 45 cm long set in a vacuum chamber in which as much as 50 carbon rods with 300 mm long size can be previously installed and (c) a control panel where a mini-computer automatically feeds every stored carbon rods to the arc region [4]. Discharge power is maintained by a DC power supply with constant current control, which has maximum voltage, 65 V and

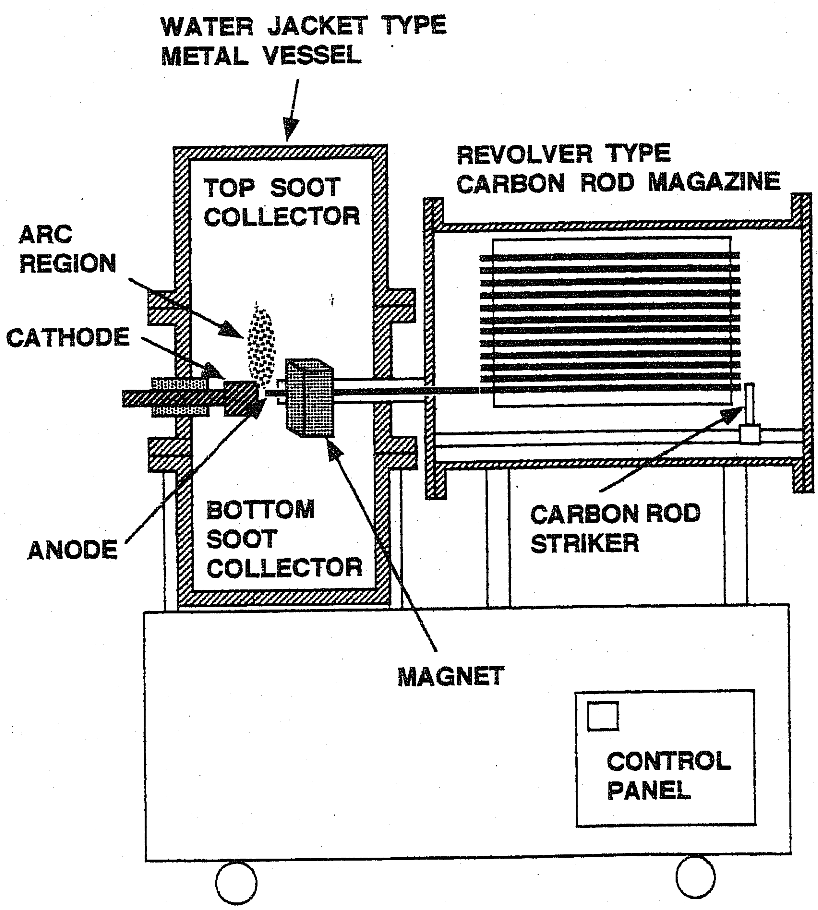

Fig. 1. Schematic of the experimental setup (RIT-FP Machine)

maximum current, 160 A. The whole chamber is evacuated well to less than 1 Pa to avoid contamination before starting the experiment, and a rectangular type ferrite-magnet (4 cm × 4 cm × 10 cm) is set at the outside of the middle chamber, which makes about 1.2 mT of magnetic field at the arc region. Applying the magnetic field, the arc plasma is accelerated to upper direction by the $J \times B$ force. The anode carbon rod (8.0 mmϕ × 300 mm long) is fed from the revolver chamber to the arc region and the carbon cathode is 30 mm ϕ × 30 mm long. The gap distance between two electrodes is maintained by computer control.

For the standard arc condition, He pressure $p = 40$ kPa, discharge current $I_d = 120$ A and gap distance $d_G \sim 5$ mm. For pressure dependent data, pressure variation from 6 kPa to 100 kPa at $I_d = 120$ A and $d_G \sim 5$ mm. In the case of current dependent data, discharge current range from 80 A to 160 A at $p = 40$ kPa and $d_G \sim 5$ mm. For the case of gap distance dependent examination, variation of gap distance, $d_G = 1$, 5 and 10 mm, $I_d = 120$ A and $p = 40$ kPa,.

A UV/Vis Spectrophotometer (UV-1200, Shimadzu Co.) is used to measure the C_{60} content for each parameter dependent condition. The absolute C_{60} content is scaled by using a scaling-factor equation obtained by a standard sample. C_{60} content in 1 mg soot is calculated from the absorption measurement as follows,

$$C(C_{60}) = A \frac{I_{ab}}{W_{soot}} \quad , \tag{1}$$

where A is constant obtained from the standard sample, I_{ab} is absorbance at $\lambda = 328.5$ nm and W_{soot} is dissolved soot weight.

After the dischage, all produced soot is collected and mixed well and a part of the spot is used to measure the C_{60} content. 2.0 mg of carbon soot is dissolved in 7.0 ml of hexane and its solution obtained by filtering is analyzed by the UV/Vis spectrometer, where absorbance at $\lambda = 328.5$ nm [11] is used to decide C_{60} content. A laser desorption time of flight mass spectrometer (LD-TOF-MS, Shimadzu-Cratos Maldi III) is also used to confirm the extracted fullerenes molecules.

Results and Discussion

A comparative experiment has been carried out between the cases with magnetic field ($J \times B$) and without magnetic field ($B = 0$) where He pressure $p = 40$ kPa, discharge current $I_d = 120$ A and arc gap distance $d_G \sim 5$ mm. Soot production rates for the two cases are shown in Fig. 2(a). C_{60} Content and production rate of C_{60} calculated from the UV absorbance are shown in Fig. 2(b) and (c) where open rectangles show experimental errors from 6 measurements. The typical UV/Vis spectrum is shown in Fig. 3, where $I_d =$ 120 A, $p = 40$ kPa and $d_G \sim 5$ mm. Clear 3 peaks of C_{60} are obtained in this graph. For the soot production rate in the $J \times B$ case, sublimated carbon soot is accelerated to

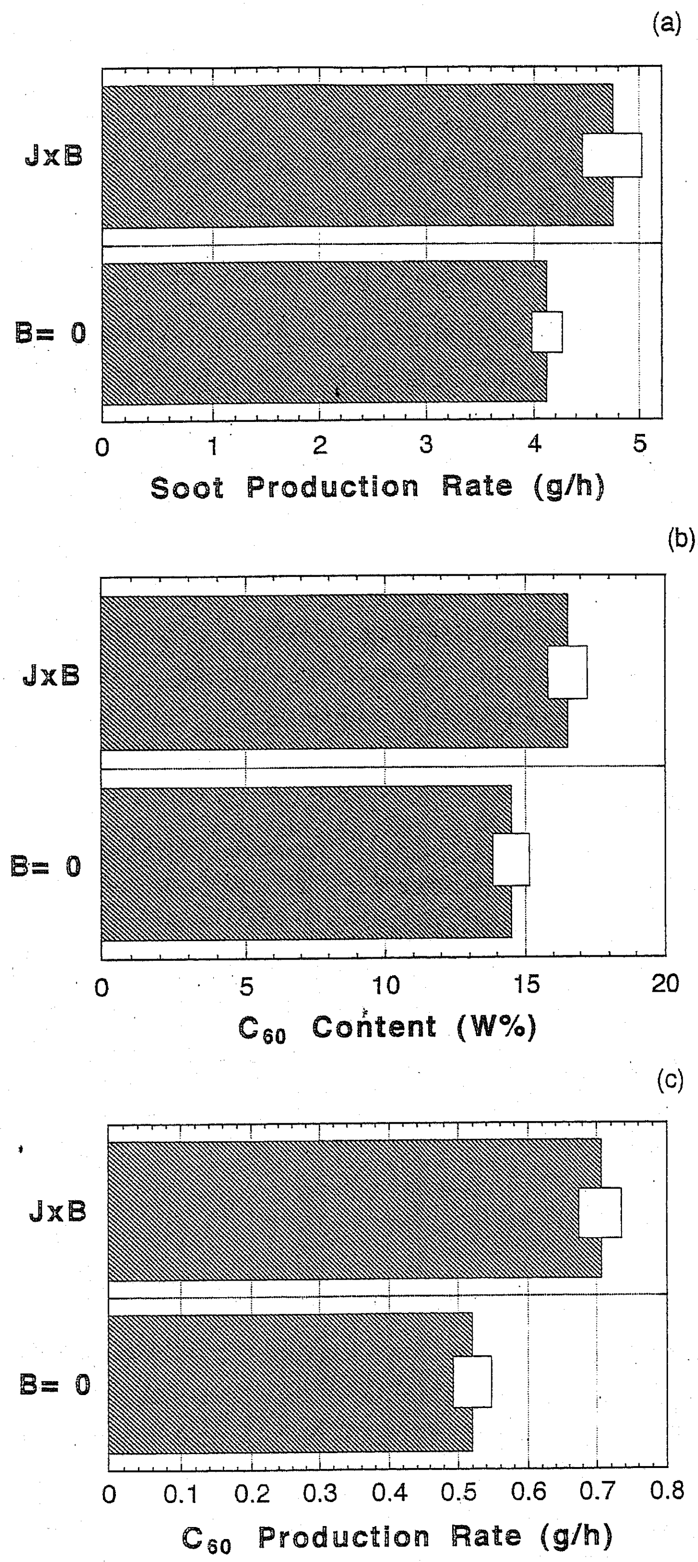

Fig. 2. (a) Production rate of carbon soot, (b) C_{60} content and (c) production rate of C_{60} for the $J \times B$ and the $B = 0$ cases, where $I_d = 120$ A, $p = 40$ kPa and $d_G \sim 5$ mm.

upper direction and carbon deposit at the top part is very large which includes much amount of fullerenes, while, for the $B = 0$ case, deposition at the bottom part is considerably large, which includes less amount of fullerenes. Considerable increase of the production rate of C_{60} is obtained for the $J \times B$ condition. The magnetic field can modify the equilibrium arc condition, and it effectively flows out sublimated carbon atoms to the low temperature gas region. The collision time and cooling time of carbon clusters are conjectured to be modified by the magnetic field.

Soot production rate, C_{60} content and production rate of C_{60} as a function of discharge current are measured and shown in Figure 4(a)-(c) where $p = 40$ kPa, $d_G \sim 5$ mm with the $J \times B$ arc. The error bars are calculated from 3 measurements. At $I_d = 120$ A, optimum C_{60} content is obtained, whereas, production rate of C_{60} is the highest around discharge current of 120-160 A. At $I_d = 140$-160 A, the large production rate of carbon soot also increases the production rate of C_{60}. Lower current density seriously decreases the sublimation rate of carbon vapor and excess current would increase the sublimation carbon clusters, which do not take role of the C_{60} production. As a result, optimum condition of production rate of C_{60} is found at the discharge current of 120-160 A. Because discharge voltage is almost constant with an increase of the discharge current, discharge power is almost proportional to the discharge current, C_{60} production rate increases almost proportional to the discharge current, when a proper rod diameter is selected.

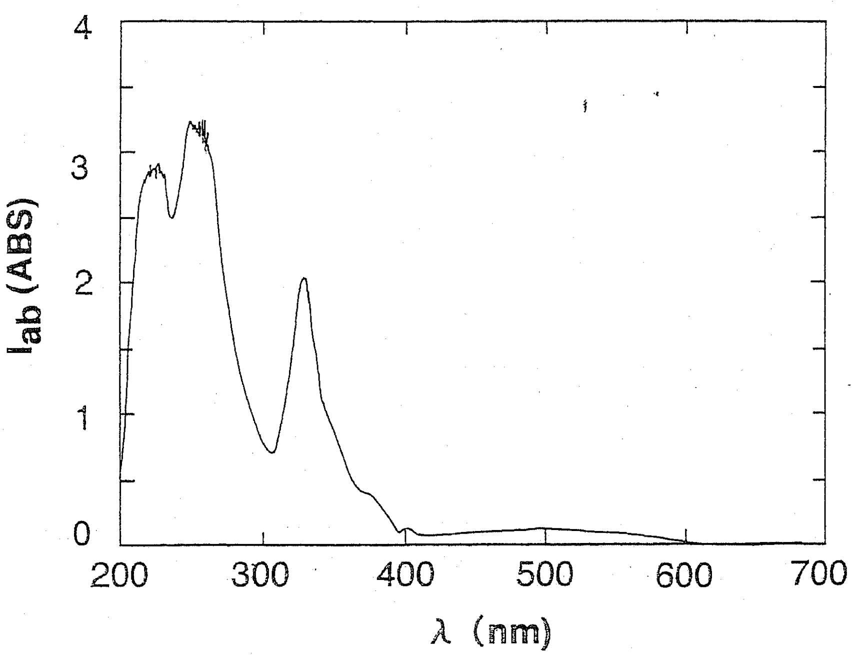

Fig. 3. Typical UV/Vis spectrum of the hexane extract from the produced carbon soot.

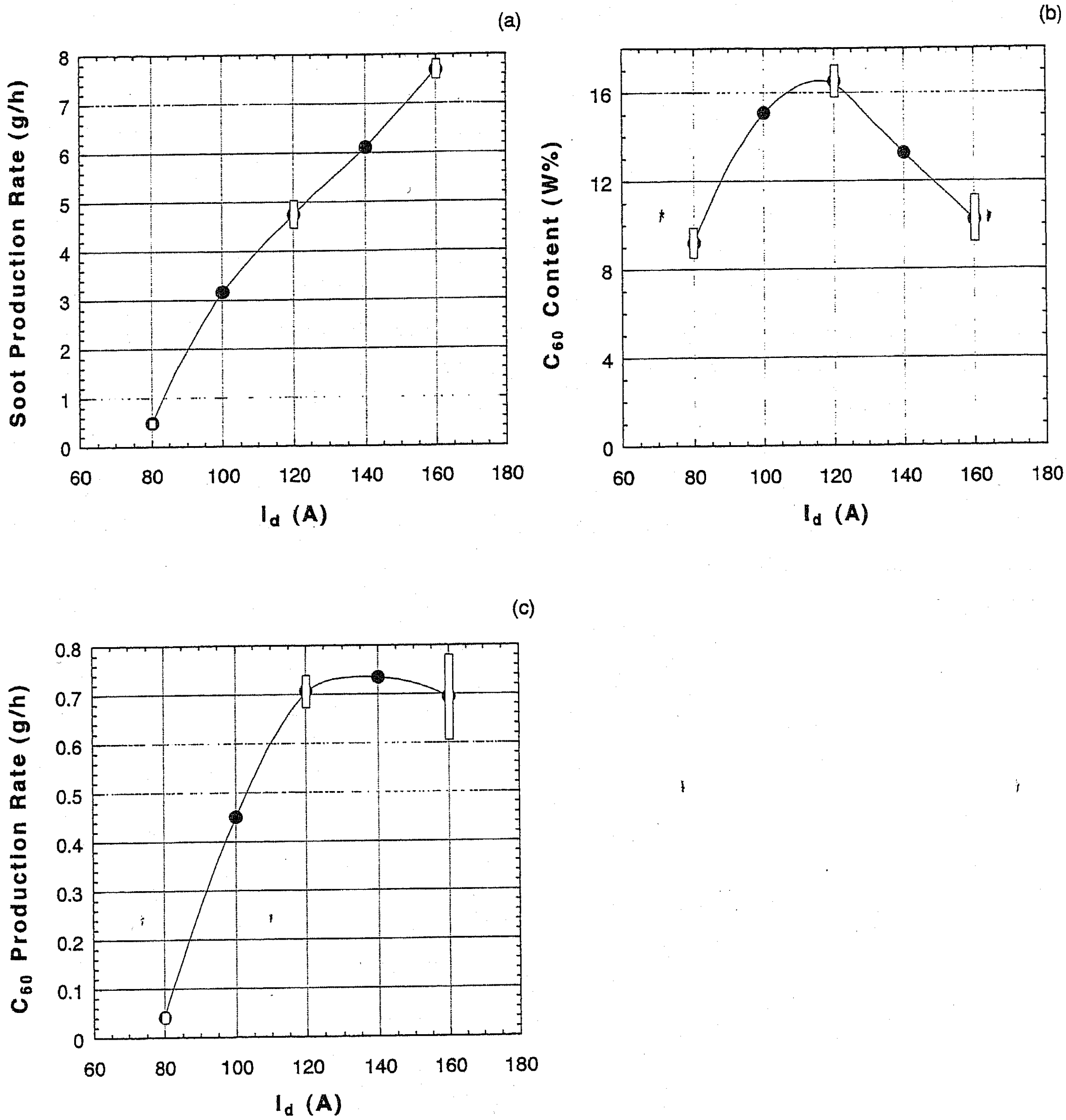

Fig. 4. (a) Production rate of carbon soot, (b) C_{60} content and (c) production rate of C_{60} versus current in the $J \times B$ method, where $p = 40$ kPa and $d_G \sim 5$ mm.

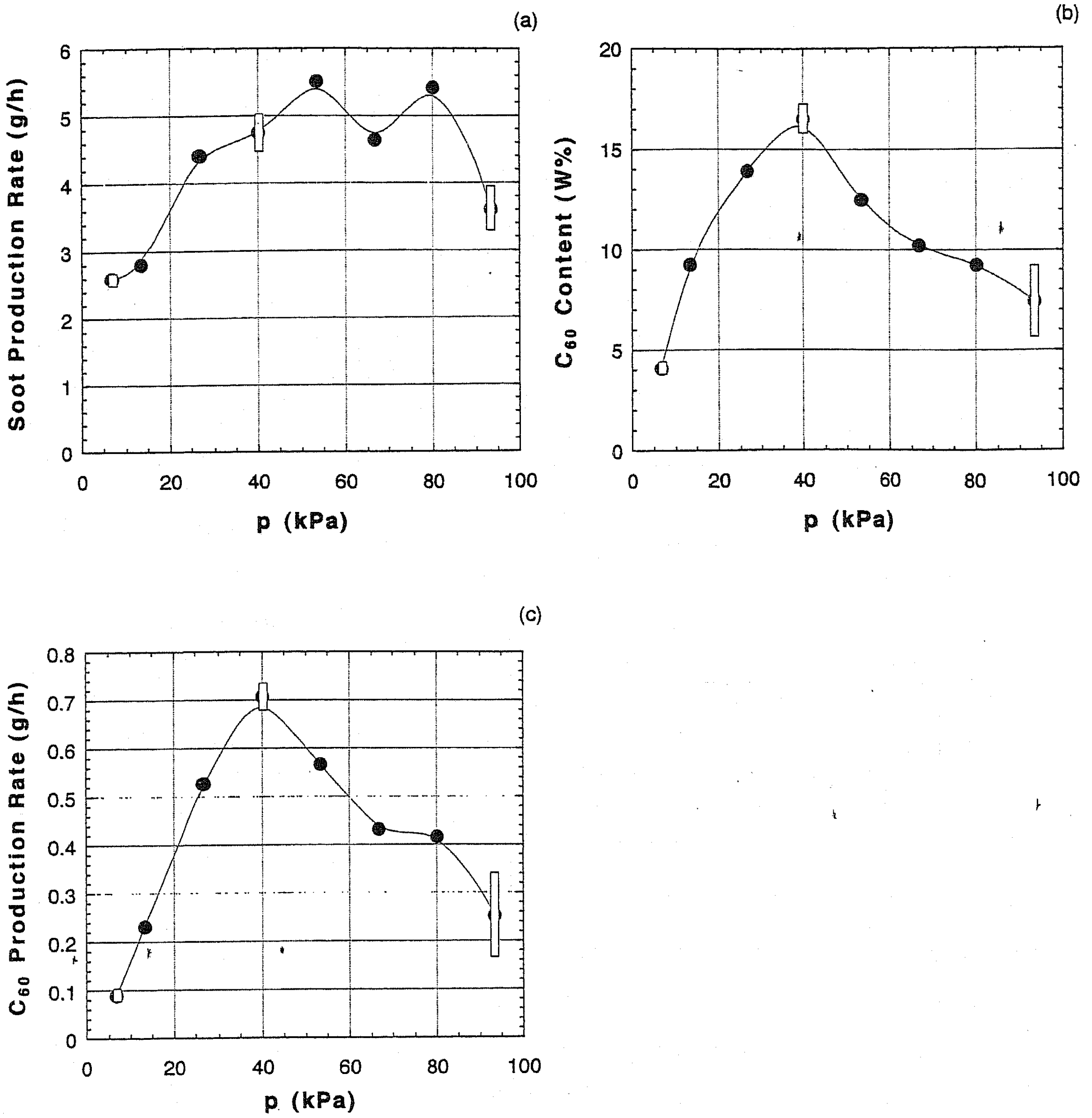

Fig. 5. (a) Production rate of carbon soot, (b) C_{60} content and (c) production rate of C_{60} versus He pressure in the $J\times B$ method, where I_d= 120 A and d_G ~ 5 mm.

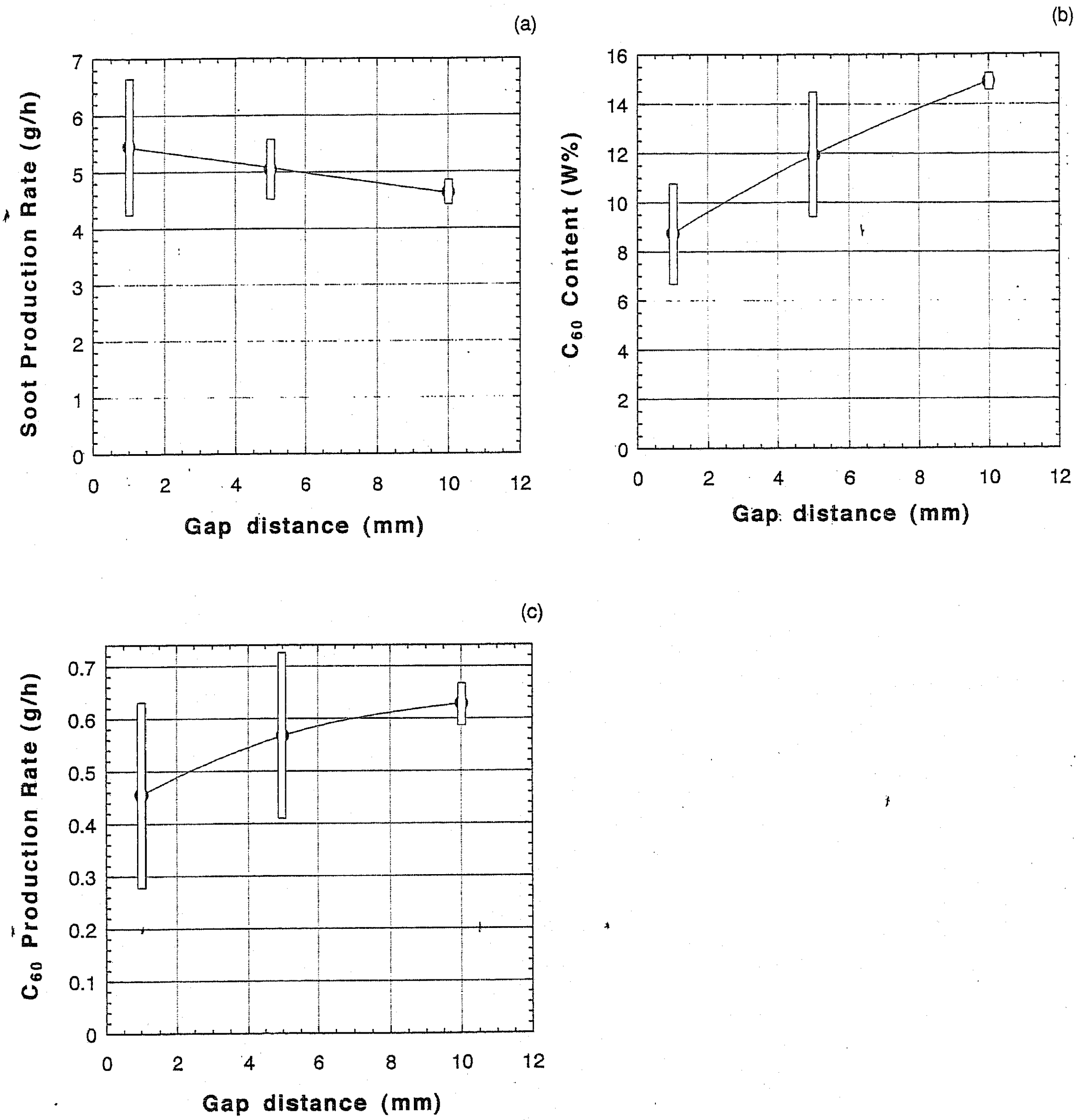

Fig. 6. (a) Production rate of carbon soot, (b) C_{60} content and (c) production rate of C_{60} versus gap distance in the $\boldsymbol{J} \times \boldsymbol{B}$ method, where $I_d = 120$ A and $p = 40$ kPa.

Soot production rate, C_{60} content and production rate of C_{60} as a function of He pressure is measured by the UV spectrometer and the results are shown in Figure 5(a)-(c), where $I_d = 120$ A, $d_G \sim 5$ mm with the $J \times B$ arc. The measurement errors are calculated from 6 measurements. At $p = 40$ kPa, optimum C_{60} content and C_{60} production rate are obtained. At lower pressure, insufficient collision decreases sufficient reaction time for fullerene synthesis. At the excess pressure, high plasma density would reduce the proper atomic sublimation rate of carbon.

Finally, effect of gap distance is examined. For gap distances, $d_G = 1$, 5 and 10 mm, fullerene is produced, where $I_d = 120$ A, $p = 40$ kPa and the $J \times B$ arc is used. Soot production rate, C_{60} content and production rate of C_{60} as a function of gap distance are measured and shown in Fig. 6(a)-(c). For $d_G = 5$-10 mm, preferable production rate of C_{60} is obtained. Narrower arc space limits free-space reaction, obstracting effect of the gas phase reaction and much amount of sublimated carbon deposit on the cathode. For wider gap, large plasma volume absorbs heating energy and the heating efficiency is insufficient for efficient fullerenes production. These effects influence on C_{60} production.

Conclusion

Production rate of C_{60} considerably increases by applying the magnetic field (the $J \times B$ arc discharge). In this $J \times B$ method, the production rate of C_{60} is maximum at the discharge current of 120 A, the He pressure of 40 kPa and the arc gap distance of 5-10 mm, using 8 mmϕ size carbon rods. It is conjectured that the $J \times B$ force modifies heat flow and high temperature reaction in the arc gas space and influences on the formation of fullerene families, while the complete control of the arc synthesis cannot be done by applying the magnetic field. By increasing both the carbon rod diameter and the discharge current, the production rate of C_{60} can be linearly increased with the discharge power.

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$J \times B$アーク法を用いたC_{60}合成におけるパラメーター依存性

モハメド・カイラル・ハッサン・ブイヤン，三重野　哲

静岡大学理学部物理学科
〒422-8529 静岡市大谷836

$J \times B$ アーク法を用いた C_{60}合成において，フラーレンを含むすす合成率，C_{60}含有率および C_{60}合成率のパラメーター依存性を実験的に調べた．直径 8 mmϕ の原料炭素棒を用いた場合，C_{60}合成率は，ヘリウム圧力 40 kPa，放電電流 120 A，ギャップ長約 5 mm の時に最大となった．