Investigation of Basic Properties of a Fullerene Ion Source Using Sublimation and Electron Attachment

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Abstract

This article reports basic properties of a prototype fullerene ion source that is being under development at Tokyo Tech. We adopt a sublimation electron irradiation method to ionize C_{60} and successfully obtain a 1.6-nA negative fullerene beam. The results suggest that irradiated electron energy has a key to improve the ionization efficiency of fullerene.

1 Introduction

The cluster, an aggregate of several to several million atoms or molecules, has been attracting much attention during the last two decades. There are various types of clusters depending on constituent atoms and bonding structures. In particular, C_{60} fullerene, which has a soccer-ball-like structure composed of sixty carbon atoms, is the most famous one. A lot of studies on the fullerene have been conducted since its discovery by Kroto in 1985 [1] and the establishment of the industrial mass production method in 1990.

One of the most successful applications of the cluster is time-of-flight secondary-ion mass spectrometry (TOF-SIMS) with a C_{60} primary ion beam [2]. The use of C_{60} drastically enhanced secondary-ion emission efficiency compared with the case using monoatomic ions. The kinetic energy of C_{60} ions in this application is typically 10 keV, so they interact only with the sample surface. On the other hand, the induction microtron was recently proposed to accelerate large cluster ions having low specific charges to much higher energies than ever achieved [3]. This new type accelerator will enable us to investigate the interaction of energetic clusters with bulk solid materials.

To supply a large amount of fullerene ions to the accelerator, the ion source need to adopt an efficient ionization scheme. The ionization of fullerene has been investigated by electron impact ionization for generation of relatively low charge-state fullerene ions [4, 5] and multi-photon ionization for higher charge-state ions [6]. However, the ionization of fullerene has not been investigated in terms of efficient ion supply to the accelerators.

The purpose of this study is to develop a fullerene ion source that can supply a high-current fullerene ion beam and examine its basic properties. The ion source uses a sublimation and electron irradiation method for ionization of fullerene. This method allows us to produce both positive and negative ions by controlling the kinetic energy of irradiated electrons. When the electron energy is lower than approximately 10 eV, negative fullerene ions are formed by electron attachment [4]. With electron energy more than 10 eV, positive fullerene ions are formed by electron impact [5]. This article reports experimental results on basic properties of the fullerene ion source that we originally developed at Tokyo Tech.

2 Experimental Setup

Figure 1 shows a schematic of the test bench of the fullerene ion source. The ion source components are installed in a vacuum chamber evacuated down to 10^{-5} Pa. Commercially available fullerene powder is contained in an oven and heated up to 500 °C to obtain fullerene gas through a nozzle. Elec-

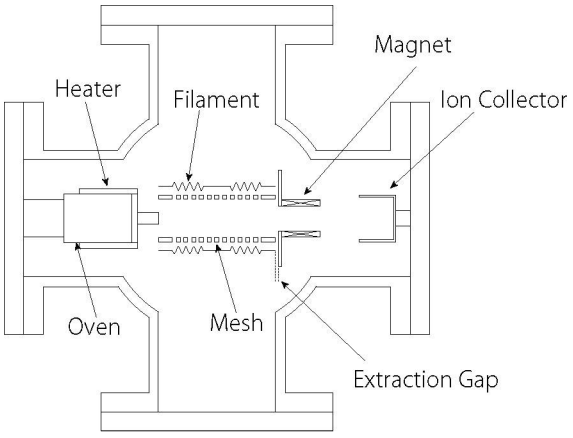

Figure 1: A schematic of the test bench of the fullerene ion source.

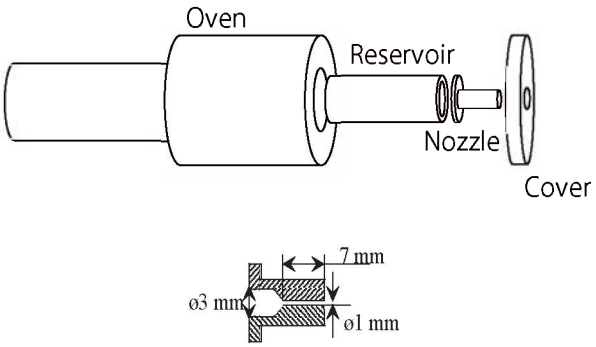

Figure 2: Inner structure of the sublimation component (upper) and cross-sectional view of the nozzle (lower).

trons emitted from tungsten filaments are accelerated by biased mesh electrodes and introduced to the space where the evaporated fullerene gas flows downstream. Fullerene molecules are irradiated by the incoming electrons and negatively ionized by electron capture reactions. The fullerene ions are extracted by an extraction voltage of typically 1 kV and detected by a Faraday cup. Electrons are also extracted at the same time, but they are suppressed by a transverse magnetic field applied just after the gap.

The sublimation component consists of an oven, a reservoir, and a nozzle. Figure 2 shows the inner structure of the sublimation component and the cross section of the nozzle. To heat the oven, we used a commercially available band-heater. The maximum operation temperature of the heater is 760 °C. All parts except for the heater are made of brass. Fullerene powder in the reservoir is heated up to a temperature of $\sim 450\,^{\circ}\mathrm{C}$ typically. Fullerene molecules evaporates and flows into the ionization section through the nozzle. The flow rate of the fullerene vapor is regulated by the oven temperature and the nozzle conductance. The flow rate Q of fullerene vapor is given by

$$Q = C \times P, \tag{1}$$

where C is the nozzle conductance and P the vapor pressure of fullerene. Assuming the molecular flow, the conductance C_m is given by

$$C_m = \frac{2\pi a^3 v}{3L}, \tag{2}$$

where a is the nozzle diameter, L the nozzle length, and v the averaged thermal velocity of fullerene molecules. On the other hand, the conductance of viscous flow C_v is given by

$$C_v = \frac{\pi a^4 P}{16\eta L}, \tag{3}$$

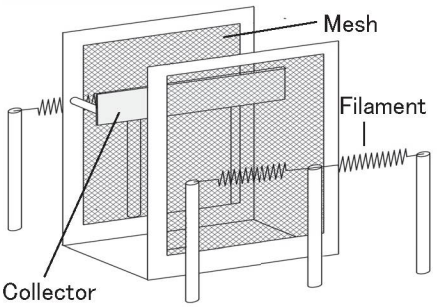

Figure 3: An illustration of the ionization component.

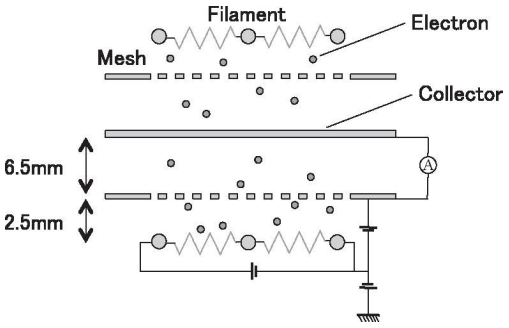

Figure 4: Circuit diagram of the ionization component.

where η is the coefficient of viscosity. We estimate the amount of consumption of fullerene from the difference of the reservoir weight before and after the operation.

Figures 3 illustrates the ionization component. The ionization component consists of four tungsten filaments and two counter-facing mesh electrodes made of stainless steel. The electron current density emitted from the filament is described by the Richardson-Dashman's equation,

$$J_{\mathrm{RD}} = \frac{4\pi m e k^2 T^2}{h^3} \exp\left(-\frac{\phi}{kT}\right), \tag{4}$$

where m is the mass of electron, e the elementary charge, k the Boltzmann constant, h the Planck constant, T the temperature of filament, and ϕ the working function. As shown in Eq. (4), the electron current is proportional to the square of the filament temperature. Thermal electrons from the filaments are accelerated up to 18 eV by the biased mesh electrodes and introduced to the space between the electrodes, where the fullerene vapor flows. In evaluating the electron flux usable for the ionization of fullerene, a charge collector (10 mm×40 mm) was mounted on the fullerene beam axis.

The circuit diagram of the ionization component

is shown in Fig. 4. The distance between the filament and the mesh electrode is 2.5 mm, and the distance between the mesh electrodes is 13 mm. Because both the filaments and the mesh electrodes are biased with respect to the ground potential, fullerene ions generated between the electrodes are accelerated downstream.

3 Results and Discussion

Figure 5 plots the flow rate of fullerene vapor as function of the oven temperature. The flow rate increases with increasing oven temperature. In particular, it increases remarkably when the oven temperature exceeds approximately 400 °C. Solid lines in the figure show the calculated flow rates assuming molecular flow conductance C_m and viscous flow conductance C_v. The measured flow rates above 400 °C almost coincide with the calculated ones, indicating that the fullerene vapor behaves as viscous flow in the nozzle in this temperature regime.

Figure 6 shows electron current densities measured as a function of mesh electrode voltage for filament voltages of 25, 30, and 35 V. The electron current density increases both with increasing mesh electrode voltage and with increasing filament input power. Since no saturation is observed in the electron current density even with the highest filament voltage (35 V), there is a room for increasing electron current density by increasing filament input power.

In the present study, we performed an experiment on the production of fullerene negative ions. Time evolutions of the oven temperature and the negative ion current measured by the Faraday cup are shown in Fig. 7. An ion current drastically increases when the oven temperature exceeds approximately 300 °C, which indicates that the sublimation of fullerene begins around this temperature. Thus, the observed negative current is obviously attributed to fullerene vapor. However, there still remains the possibility that we observed the negative ions originating from fragments of fullerene molecules.

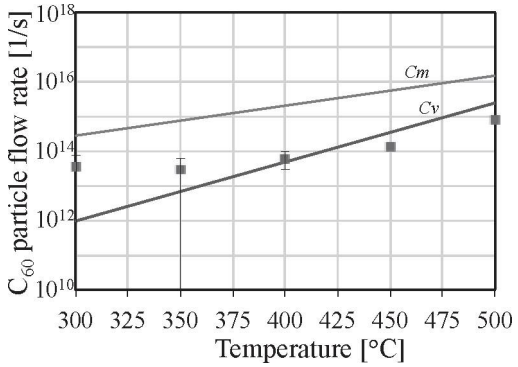

Figure 5: Fullerene vapor flow rate as a function of oven temperature.

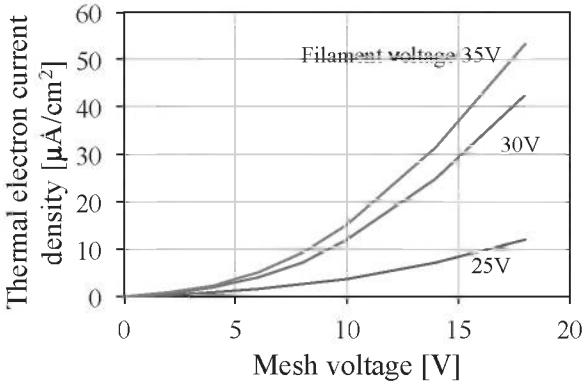

Figure 6: Dependencies of electron current density on mesh electrode voltage and filament voltage.

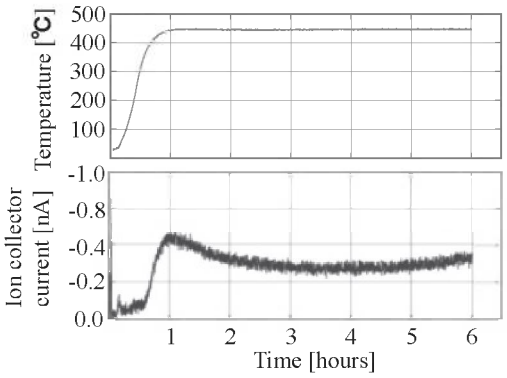

Figure 7: Time evolutions of oven temperature and negative ion current in a typical operation.

The negative ion current flowing into the ion collector was measured as a function of the mesh voltage for different filament voltages from 25 to 35 V as shown in Fig. 8.

We obtained a maximum negative ion current of 1.6 nA at a filament voltage of 35 V and a mesh voltage of 18 V. The irradiated electron energy is determined by the mesh voltage. It is quite curious that negative ion current continues to increase when the electron energy increases beyond 10 eV. Since the cross section of electron attachment to a fullerene drops sharply for electron energies above 10 eV, there is almost no chance for primary electrons to attach fullerene molecules. To explain this result, we need to additional processes such as secondary electron attachment.

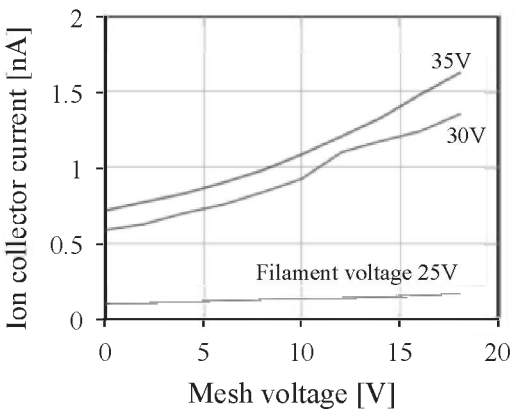

Figure 8: Dependencies of negative ion current on mesh electrode voltage and filament voltage.

4 Concluding remarks

This article reported basic properties of a prototype fullerene ion source using sublimation and electron irradiation. We successfully obtained 1.6-nA negative ion beam current from the fullerene ion source. The ion beam current was found to depend not only on the fullerene vapor flow rate but on the energy of irradiated electrons. In particular, electrons having an energy higher than 10 eV obviously contribute to the enhancement of the negative ion current, which implies that the secondary electrons generated by electron impact ionization of fullerene causes negative ion production.

References

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