

A plasma reactor based on the forced constricted type dc plasma jet and its application to thermal plasma processing in low pressure

Satoshi Sakiyama, Tetuya Hirabaru, and Osamu Fukumasa

Department of Electrical and Electronic Engineering, Faculty of Engineering, Yamaguchi University, 2557 Tokiwadai, Ube 755, Japan

(Presented on 30 September 1991)

A reactor consisting of a forced constricted type dc plasma jet generator and a feed ring has been developed. In order to demonstrate the application feasibility of this reactor to low pressure plasma processing, the characteristics of the forced constricted arc in low pressure, and production of ultrafine particles of refractory materials (Al_2O_3 , ZrC, TiC) are studied.

I. INTRODUCTION

There has been increasing interest in the application of plasma processing. Two types of plasmas, i.e., thermal plasma and cold plasma, are used in plasma processing. Thermal plasma can process materials at a high rate, but it is not easy to do processing uniformly in large areas. On the other hand, although large area deposition can be realized by using cold plasma, deposition rate is much lower. The purpose of this study is to realize plasma processing that can successfully combine the merits of those two types of plasmas, i.e., to realize the plasma processing with both high rate and large area deposition by using thermal plasma under low pressure.

The plasma jet is one of thermal plasmas with high speed and high gas temperature, so it has the ability to produce high quality sprayed coatings or synthesis of ultrafine particles of refractory materials. The keys to applying the plasma jet to low pressure plasma processing are to develop a plasma source that can generate a stable and large heat capacity plasma under wide operating conditions, and to clarify the process of particle heating and cooling in the plasma flow.

We have developed a processing reactor composed of the forced constricted type plasma jet generator and feed ring, and confirmed that this reactor generates a stable plasma jet with large heat capacity.¹ We have also made a performance test of the reactor in production of ultrafine particles and sprayed coatings of refractory materials^{2,3} and in synthesis of a diamond.⁴

In this paper, we will discuss the effectiveness of the arc in the forced constricted type plasma jet generator, i.e., stability of the forced elongated arc, thermal efficiency, etc. Further, in order to demonstrate the application feasibility of the forced constricted type plasma jet for low pressure plasma processing and then to study the interaction between powder materials and thermal plasma, we have produced ultrafine particles of refractory materials using three feed rings with different axial length. Preliminary results⁵ show that the feed ring works as a mixing plenum after injecting powders and gas mixture into plasma flow in the present reactor.

II. EXPERIMENTAL APPARATUS

A schematic drawing of the experimental setup is shown in Fig. 1. The reactor is divided into three parts: the

forced constricted (F.C.)-type plasma jet generator,⁶ the feed ring and the pressure vessel. The F.C.-type plasma jet generator is composed of the nozzle anode, the rod cathode, and the insulated constrictor nozzle. The nozzle anode (5 mm diam, 4.5 mm length) is made of copper and the rod cathode (5 mm diam) is 2%Th-W. The insulated constrictor nozzle (5 mm diam, 8 mm length) consists of three copper disks and four BN plates, which insulate each of three copper disks, as is shown in Fig. 1. The signs G1, G2, and G3 denote the three copper disks in order of the distance from the anode and the signs V1, V2, and V3 denote the voltage differences between each copper disk and anode, which is at the ground, respectively. By measuring the potential of the copper disk, we can estimate the electric field strength of the arc column. The electrode loss is also estimated from the difference between the temperature of the cooling water at the inlet of the electrode and that at the outlet.

In this experiment, we have used three different feed rings, which are denoted by the signs FR1, FR2, and FR3 in order of axial length. All feed rings have the same nozzle diameter (5 mm), and two powder feeding ports (1.4 mm diam) that are positioned at 3.5 mm below the feed ring entrance. But the distance (1 m) from the powder feeding port to the feed ring exit is different in each ring. These lengths of FR1, FR2, FR3 are 6, 12, and 18 mm, respectively.

Powder was injected into the plasma with carrier gas through the feeding ports and melted or vaporized. Prepared fine particles are quenched by the cold finger and

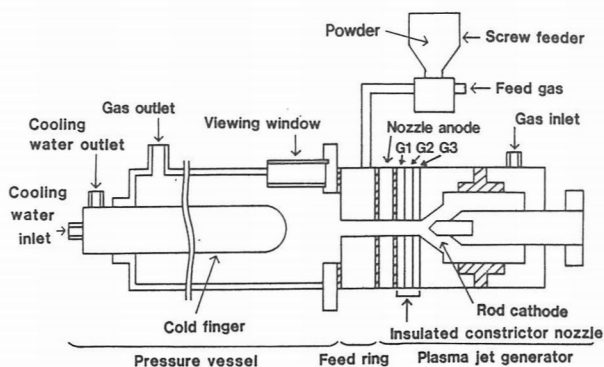


FIG. 1. Schematic diagram of the plasma jet reactor system.

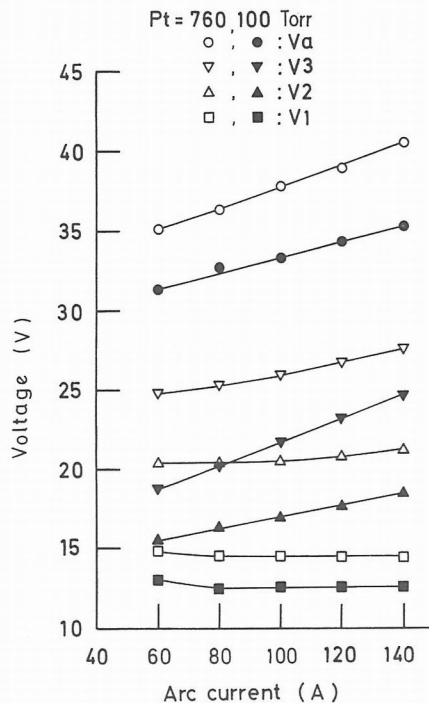


FIG. 2. Voltage versus arc current for two different pressures. V_a denotes arc voltage. V_1 , V_2 , and V_3 denote the voltage differences between the copper disk and anode that is earthed.

adhered to the plane glass set on the cold finger.

Experiments were made under the following conditions: Working gas (Ar) flow rate is 20 l/min. Pressure (P_t) in the reaction chamber is kept at 760 and 100 Torr, respectively. Net arc input power is 3 kW. Carrier gas (Ar) flow rate is 6 l/min. Powder flow rate is 0.5 g/min. Powder materials are Al_2O_3 (mean diameter 20 μm , diameter distributions 10–44 μm , melting point 2040 $^\circ\text{C}$), TiC (21 μm , 10–44 μm , 3140 $^\circ\text{C}$), and ZrC (22 μm , 10–53 μm , 3570 $^\circ\text{C}$).

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Electrical and thermal characteristics of the plasma jet generator

As is shown in Fig. 2, arc voltage (V_a) versus arc current (I_a), the characteristic of the forced constricted arc is a rising characteristic both under atmospheric and low pressure and V_a decreases with decreasing P_t . We have already confirmed that the decrease in the electric field strength of the arc column causes this decrease in V_a by measuring the potential of V_1 , V_2 , and V_3 .⁷ The reason why the arc in this type of generator has a rising characteristic, though the arc in the conventional type one has a drooping characteristic in the range of arc current 60–140 A, is as follows: With the use of the insulated constrictor nozzle, the arc column is elongated and fixed at constant length. The increase of arc diameter with increasing arc current is also restricted. As a result, the thermal pinch effect is so strong that the electrical conductivity is almost saturated, even in the range of low current. So the arc voltage increases with arc current. Because of this V_a vs I_a characteristic of the arc, the F.C.-type plasma jet generator

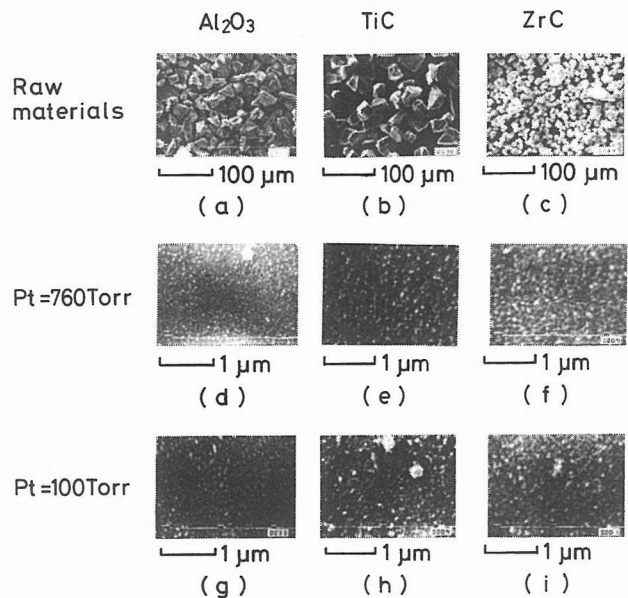


FIG. 3. SEM photographs of collected ultrafine particles and raw materials.

can increase the input power effectively compared with the conventional type by changing arc current under not only atmospheric but also low pressure.

Net arc input is obtained after subtracting the total heat loss of electrodes from total input power ($I_a \times V_a$). Electrode loss and net arc input decrease with decreasing P_t . But even at 100 Torr thermal efficiency of the generator [(net arc input/total input power) \times 100] is kept at 57%–64%, which is nearly equal to or larger than the value of the efficiency at 760 Torr (54%–62%). Further, these values are larger than those of a conventional-type plasma jet generator by 10%–20%. We have also obtained preliminary results that the insulated constrictor nozzle that is used to keep the arc stable and to increase the input power in this generator does not decrease the thermal efficiency of the generator.

It is found that the F.C.-type plasma jet generator can increase the net arc input without reducing the thermal efficiency of the reactor by elongating the axial length of the insulated constrictor nozzle, even in low pressure.

B. Application of the reactor for production of ultrafine particles

At first, the influence of the feed ring length on the performance characteristics of the forced constricted-type plasma jet generator was studied. We found that the reactor produced a stable plasma jet under the various operating conditions, even if any feed ring within three different ones was installed.⁵

SEM photographs of fine particles prepared under the condition of $P_t = 100$ and 760 Torr and raw powder materials are shown in Fig. 3. For both pressures, many ultrafine particles (diameter $D < 0.1 \mu\text{m}$) are prepared. These samples are analyzed by x-ray diffraction. It is confirmed that impurities, for example, electrode materials, are not detected.

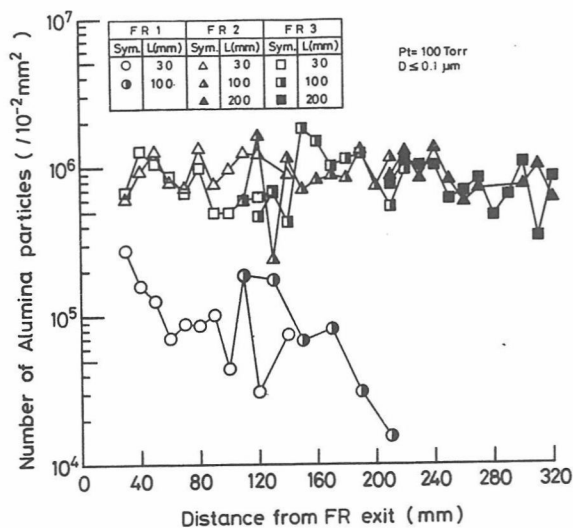


FIG. 4. Density of collected Al_2O_3 fine particles ($D < 0.1 \mu\text{m}$) versus distance z from the feed ring exit at 100 Torr. L represents the distance from feed ring exit to the tip of the cold finger.

In order to study the effect of the feed ring length on the production of fine particles, we obtain the relationship between the feed ring length and the number of collected fine particles. We collect the fine particles setting the cold finger at some different positions L , where L is the distance from the feed ring exit to the tip of the cold finger. According to SEM photographs, the number density of fine particles per unit area, i.e., $0.1 \times 0.1 \text{ mm}^2$ is estimated.

Figure 4 shows the relationship between the number density of ultrafine particles (Al_2O_3) and z (distance from the feed ring exit) at Pt 100 Torr. Roughly speaking, the density of collected fine particles decreases with increasing z when the reactor is operated with FR1 ($lm = 6 \text{ mm}$), where lm is the distance from the feeding ports to the feed ring exit. On the other hand, the density keeps a constant value when the reactor is operated with FR2 ($lm = 12 \text{ mm}$) or FR3 ($lm = 18 \text{ mm}$). Besides, the density for the latter case is higher, especially in the downstream region, than that for the case of FR1 ($lm = 6 \text{ mm}$). The increment of the density by increasing the feed ring length is so remarkable in low pressure that the density is equal to or greater than that in atmospheric pressure.

To clarify the effect of the feed ring length on processing further, the temperature of the plasma jet was measured. It was estimated from the ratio of the intensity of two spectral lines (4159 and 6965 Å). Figure 5 shows the radial distributions of the spectroscopic temperature of the plasma jet for two different feed rings at $z = 4 \text{ mm}$. The temperature for the case of the FR3 ($lm = 18 \text{ mm}$) is nearly equal to that for the case of FR1 ($lm = 6 \text{ mm}$), though the feed ring lengths are different from each other. That is to say, the increase of the feed ring length extends the high temperature region of the plasma flow.

Experimental results shown in Figs. 4 and 5 indicate that increasing the feed ring length extends the region in which powder materials are heated effectively by the

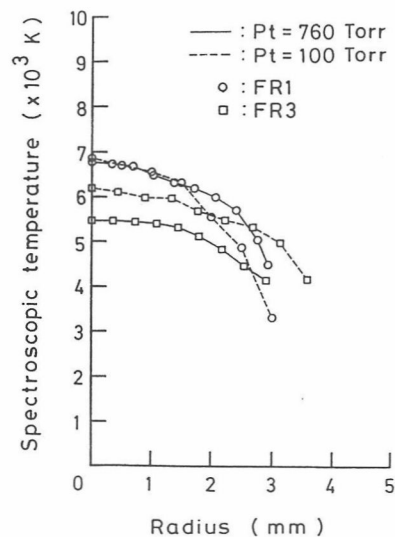


FIG. 5. Radial distributions of the spectroscopic temperature for two different feed rings measuring position $z = 4 \text{ mm}$.

plasma flow with high temperature. This might be one of the reasons why the number of fine particles increases with the increase of the feed ring length.

In summary, it is found that by using the insulated constrictor nozzle, this reactor can increase the net arc input without decreasing the thermal efficiency of the reactor, even in low pressure. Besides, with increasing feed ring length, the high temperature regions are extended and the number of ultrafine particles increases. So even in low pressure processing, this reactor can produce ultrafine particles, the density of which is nearly equal to or larger than that in atmospheric pressure processing by increasing the feed ring length. We have concluded that this type of reactor has high potential for low pressure plasma processing.

¹S. Saeki, O. Fukumasa, K. Matsubara, K. Osaki, and I. Yamada, Proceedings of the 9th Symposium on Ion Source and Ion-Assisted Technology, Tokyo, 1985, p. 31.

²S. Saeki, O. Fukumasa, and K. Osaki, Proceedings of the 8th International Symposium on Plasma Chemistry, Tokyo, 1987, p. 1989.

³O. Fukumasa, T. Onzuka, S. Sakiyama, K. Osaki, and S. Saeki, Proceedings of the 6th Symposium on Plasma Processing, Kyoto, 1989, p. 431.

⁴S. Sakiyama, T. Hirabaru, O. Fukumasa, H. Naitou, and K. Osaki, Proceedings of the 8th Symposium on Plasma Processing, Nagoya, 1991, p. 453.

⁵S. Sakiyama, M. Watanabe, O. Fukumasa, H. Naitou, and K. Osaki, Proceedings of the 7th Symposium on Plasma Processing, Tokyo, 1990, p. 49.

⁶S. Saeki and K. Uchiyama, Mem. Fac. Eng. Yamaguchi Univ. 27, 113 (1976).

⁷S. Sakiyama, T. Hirabaru, and O. Fukumasa, Proceedings of the 9th Symposium on Ion Source and Ion-Assisted Technology, Tokyo, 1991, p. 63.