Experimental study and sterilizing application of atmospheric pressure plasmas

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Abstract

The atmospheric pressure surface barrier discharge (APSBD) in air has been used in killing \textit{Escherichia coli}. We have developed two similar dielectric barrier discharge (DBD) structure types of nonthermal plasma jets (PJ) driven by 5–20 kHz audio-frequency power at atmospheric pressure. At a flow rate of 200 L/h (argon), a stable, arc-free discharge was produced. At 1.5 cm from the nozzle, the gas temperature was kept at 47 °C for PJ-1 source and 38 °C for PJ-2 source. Some research on sterilization has been carried out and results show that such a plasma jet source as PJ-2 is very effective in the disruption of \textit{E. coli}.

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1. Introduction

Atmospheric pressure discharges have been widely investigated in recent years because of a variety of advantages they offer, such as low-cost equipment, no vacuum operation and the discharge reactivity. Atmospheric pressure plasma, such as the arc, torch produced by arc discharge have also been investigated and applied to a number of industrial fields. However, its application is limited yet, because these plasma sources are thermal plasma sources having high gas temperature that leads to indiscriminate “burning” rather than selective chemical reactions. A discharge generated at or upon a dielectric surface can generate highly active non-equilibrium plasma at atmospheric pressure and even at room temperature. Roth et al. [1], Koinuma et al. [2–6] and Park et al. [7–9] have developed kinds of non-equilibrium atmospheric pressure plasma sources, such as symmetric pectinate electrodes panel driven by audio-frequency power and plasma jet (or cold torch) source driven by 13.56 MHz RF power, respectively. Those atmospheric pressure plasma sources can be used for environmental protections, deposition of thin films, etching materials and surface treatment of materials.

We have already studied atmospheric pressure surface barrier discharge (APSBD) in air using kinds of pectinate electrodes panels driven by audio-frequency power and experimentally proved that such APSBD plasma was a very simple, effective and innocuous tool for sterilization [10]. We found that if we offered a gas flow to blow out the APSBD plasma and concentrated the highly active non-equilibrium plasma into one line, we could obtain atmospheric pressure plasma jet driven by audio-frequency power. In the present study, we have developed two structure types of atmospheric pressure plasma jet sources named PJ-1 and PJ-2, which are driven by easily generated audio-frequency power variable range from 5 kHz to 20 kHz. Using PJ-1 or PJ-2, we can obtain a stable, near-homogeneous, arc-free dielectric barrier discharge (DBD) with argon at atmospheric pressure, without water-cooling device.

2. Experimental setup

A schematic drawing of the experimental setup is shown in Fig. 1. The common ground of the main structure of PJ-1
and PJ-2 is that they both consist of two concentric electrodes. The outer electrode is coupled to an audio-frequency power supply at variable frequency range from 5 kHz to 20 kHz, and the inner one is grounded with the discharge gas such as argon or helium passing it through. The key differences between these two structure types of plasma jet source are the surfaces of inner electrode and the shapes of outer electrode. For PJ-1, the grounded inner electrode is a stainless steel pipe closing on one end and only opening the other end as gas inlet. There are several holes used as spray nozzle of gas along a circular helix on the wall of the inner electrode having 8 mm inner diameter. The outer electrode of PJ-1 is a metal pipe (inner diameter 15 mm) covered with both sides insulating coat. For PJ-2, there are no holes on the wall of the inner electrode, except one nozzle at each side of the grounded inner stainless steel pipe. The outer electrode of PJ-2 is just a coil around an insulating pipe.

The gas flow rate was measured by a mass flow meter. Fig. 2 shows the electrical circuit of audio-frequency power-driven atmospheric pressure plasma jet source. A sinusoidal voltage up to several tens of kilovolts peak to peak was applied to the outer electrodes, and the source frequency was varied in the range from 5 kHz to 20 kHz. The current and voltage waveforms below were recorded by using a digital oscilloscope (Tektronics TDS220).

3. Results and discussion

First, we present the results of PJ-1 source. When argon gas flow rate was kept at 200 L/h and the applied voltage increases up 13.1 kV_{pp} at frequency 15 kHz, a soft blue glow was produced between the concentric outer and inner electrode, which then exited the narrow space through the nozzle of outer pipe electrode. At 1.5 cm from that nozzle, the gas temperature was kept at 47 °C. A photograph of atmospheric pressure discharge of PJ-1 with argon is shown in Fig. 3(a). Visual inspection of the discharge at the nozzle seems like a uniform glow with no apparent arcing. In Fig. 3(b), we show the typical waveforms of DBD current and voltage in PJ-1 source, where the discharge gas stream comes out from several holes along the circular helix of the grounded inner electrode. Although it does not appear to revolve flow of discharge gas like some condition under the arc discharge jet, the PJ-1 structure results in the inconsistent direction of discharge gas slowly spraying at the nozzle of the outer electrode.

Fig. 1. Schematic drawing of the atmospheric pressure plasma jet sources (a) PJ-1 and (b) PJ-2.

Fig. 2. Audio-frequency power-driven atmospheric pressure plasma jet source.
In order to clearly know the discharge status between the inner electrode and the outer electrode, we modified the outer electrode of PJ-1 and used a glass tube as an insulating medium. The image of plasma discharge is shown in Fig. 4(a), where visual inspection of the discharges between the electrodes at each hole along the inner electrode is revealed obviously. Fig. 4(b) shows the waveforms of current and voltage waveforms in the modified PJ-1 source. Even though the structure is slightly different from PJ-1 source, we can see from Fig. 4(a) that the discharge plasma appears around the inner electrode nearly at the same time when gas streams sprayed out from several holes along the circular helix of the grounded inner electrode. As shown in Fig. 4(a), the plasma

![Fig. 3. (a) Image of discharge of PJ-1 and (b) time-depending current and voltage waveforms.](image)

![Fig. 4. (a) Image of discharge, and (b) time-depending current and voltage waveforms.](image)

![Fig. 5. (a) Image of discharge of PJ-2 and (b) time-depending current and voltage waveforms.](image)

![Fig. 6. Schematic setup of sterilization.](image)
spray at the nozzle of PJ-1 looks like a flame and not straight. For applications such as etching, deposition, surface modification and sterilization, however, the present plasma shape is not satisfactory to achieve a rapid material processing. Hence, we designed PJ-2 to improve the characteristics of plasma spray at the nozzle.

In the case of PJ-2 source, argon gas flow rate was kept at 200 L/h and the applied voltage was increased up to 49.3 kVpp at frequency 15.9 kHz. As shown in Fig. 5(a), a bright white glow was produced at the nozzle of the grounded inner electrode and spraying outside the nozzle of outer electrode for almost 2 cm long. At 1.5 cm from the nozzle, the gas temperature was kept at 47 °C. Fig. 5(b) shows typical current and voltage waveforms of the PJ-2 source.

Although the atmospheric pressure plasma jet has wide applications, in this paper, we studied the sterilizing characteristics as one of its applications. Since the highest temperature for living Escherichia coli is 47.5 °C, we tested the sterilization effect to E. coli using plasma jet of PJ-2 at 1.5 cm away from the nozzle, so as to keep gas temperature at 38 °C.

Fig. 6 shows the experimental setup of the sterilizing application. We placed the culture media into which E. coli was implanted beforehand, at 1.5 cm from the nozzle of PJ-2. After the plasma processing, the culture mediums were then kept at 37 °C for 14 h for the incubation. By contrast with untreated culture medium growing E. coli, we plotted the relative area ratio S of colonies versus plasma treatment times in Fig. 7. It is obviously found from Fig. 7 that such a plasma jet source as PJ-2 is very effective in the disruption of E. coli.

In the case of PJ-2, we could produce higher density plasma jet compared with that in the case of PJ-1, because we could apply higher voltage and current in the case of PJ-2 under lower gas temperature, such as 38 °C. Such lower temperature in the PJ-2 was realized due to strong gas flow from one exit of the inner pipe differently from the case of PJ-1, where gas flow through each hole in the inner pipe was weak because of several gas through-holes.

In the present sterilization experiment, we consider that one of the possible sterilization mechanisms is ozone generated by the Ar plasma jet under the air circumstance at atmospheric pressure. The production yield of ozone in the case of PJ-2 may be stronger than that in the case of PJ-1. Therefore, PJ-2 is more effective in the sterilizing E. coli compared with PJ-1.

Although we have also carried out the mass spectroscopic measurement to study the by-products by the Ar plasma jet, we could not detect the ozone directly because of their unstable characteristics. However, it was no doubt that the ozone was produced under the present atmospheric plasma jet, since we could recognize their existence from the strong smell of ozone.

4. Conclusion

In the present paper, we have developed two similar DBD structure types of nonthermal plasma jets driven by 5–20 kHz audio-frequency power at atmospheric pressure named PJ-1 and PJ-2. Experimental results showed that at a flow rate of 200 L/h (argon), a stable, arc-free discharge was produced. At 1.5 cm from the nozzle, the gas temperature was kept at 47 °C for the PJ-1 source and 38 °C for the PJ-2 source. The sterilizing application experiments showed that such a plasma jet as PJ-2 was very effective in the disruption of E. coli.

References