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Development of a new atmospheric pressure cold plasma jet generator and application in sterilization

Cheng Cheng(程 诚), Liu Peng(刘 鹏), Xu Lei(徐 蕾),
Zhang Li-Ye(张力叶), and Zhan Ru-Juan(詹如娟)

Department of Modern Physics, University of Science and Technology of China, CAS Key Laboratory of Basic Plasma Physics, School of Science, Hefei 230026, *China*

Zhang Wen-Rui(张文锐)

School of Life Science, University of Science and Technology of China, Hefei 230026, *China*

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This paper reports that a new plasma generator at atmospheric pressure, which is composed of two homocentric cylindrical all-metal tubes, successfully generates a cold plasma jet. The inside tube electrode is connected to ground, the outside tube electrode is connected to a high-voltage power supply, and a dielectric layer is covered on the outside tube electrode. When the reactor is operated by low-frequency (6 kHz–20 kHz) AC supply in atmospheric pressure and argon is steadily fed as a discharge gas through inside tube electrode, a cold plasma jet is blown out into air and the plasma gas temperature is only 25–30°C. The electric character of the discharge is studied by using digital real-time oscilloscope (TDS 200-Series), and the discharge is capacitive. Preliminary results are presented on the decontamination of E.colis bacteria and Bacillus subtilis bacteria by this plasma jet, and an optical emission analysis of the plasma jet is presented in this paper. The ozone concentration generated by the plasma jet is $1.0\times10^{16}\mathrm{cm}^{-3}$ which is acquired by using the ultraviolet absorption spectroscopy.

Keywords: atmospheric pressure cold plasma jet, sterilization, spectral measurement
PACC: 5280, 5275R

1. Introduction

Although the use of an electrical discharge for disinfection was suggested and applied more than a hundred years ago, basic and applied research on sterilization by using plasma at atmospheric pressure has been carried out only relatively recently.[1] Atmospheric pressure plasma devices can provide an advantage over low pressure plasmas system because they can operate without vacuum equipment. Recently, a few novel atmospheric pressure plasma sources have been developed, such as the one-atmosphere uniform glow discharge plasma (OAUGDP),[2] the atmospheric pressure surface discharge,[2] the cold plasma torch[3] and the atmospheric pressure plasma jet (APPJ).[4] For the discharge regions of the atmospheric pressure cold plasma sources such as OAUGDP, surface discharge are so limited that they are not suitable to treat bulky or complex objects. The cold plasma jet operating in open air can resolve this problem perfectly. Koinuma *et al* report that microbeam plasma has been successfully generated by a cold plasma torch in the air powered by RF generator.[3] They use this torch for applications such as Si etching,[3] surface treatment of rubber,[5] SiO_2 and TiO_2 film deposition using alkoxysilanes[6] and tetraethoxytitanate,[7] respectively. Recently, atmospheric pressure plasma jet (APPJ) has been developed by Park *et al*, and they use this plasma jet for deposit[8] etching[9] and decontamination.[10] The plasma gas temperature of the microbeam plasma and APPJ is about 50–300 °C . [5−10] In this paper, we will show a new atmospheric pressure plasma generator supplied by the low-frequency (6 kHz–20 kHz) AC electric source and obtain a stable plasma source. The gas temperature of the cold plasma jet which we have develop is only 25–30 °C, and the cold plasma jet is more suitable for treating those vulnerable objects. Preliminary results are presented on the decontamination of E.colis bacteria and Bacillus subtilis bacteria by this cold plasma jet.

2. Experimental

Figure 1 schematically shows the experimental set-up for the atmospheric pressure cold plasma jet (APPCJ). The inside tube electrode is an all-metal tube connected to the gas supply and the ground. The outside tube is an all-metal tube connected to a low-frequency (6–20 kHz) AC supply and covered by an insulation layer. The thickness of the dielectric layer is about 1 mm. Feed gas (Ar) flow through inside tube at the gas flow rate of 50 l/h–2500 l/h, and the plasma jet forms at the outlet of the inside tube. The gap width between the inside tube and the outside tube is an important factor for generating a stable plasma, and we adjust the gap width according to the diameter of the inside tube and the outside tube.

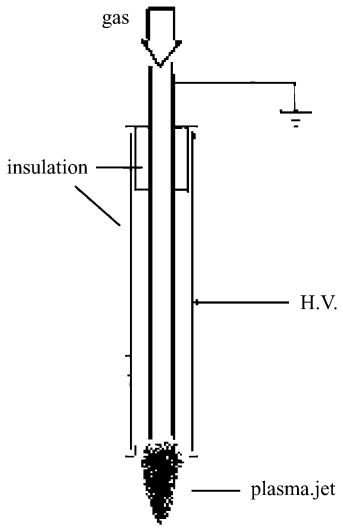


Fig.1. Schematic illustration of the cold plasma jet apparatus.

Under these conditions, we can obtain the steady and homogeneous plasma jet, when the peak-to-peak voltage is 30–80 kV and the frequency is 6–20 kHz. Two APPJ generators with different nozzle sizes are developed, whose diameters were 4mm and 20mm respectively. The pictures of the cold plasma jet are shown in Fig.2. When the discharge is stable, the plasma gas temperature is measured by an infrared radiation detector and a thermometer, and it is only 25–30 °C.

The circuit for measuring the voltage waveform and the current waveform of the discharge is shown in Fig.3. Using the digital oscilloscope we monitor the discharge electrical circuit for the applied potential through the probe of CH1 and the electrical circuit current of discharge through an electric potential drop in a 25 Ω resistor recorded by the CH2 probe.

Ultraviolet absorption spectroscopy is used to acquire the ozone concentration in the plasma and the downstream afterglow in APPJ work,[11,12] and we use

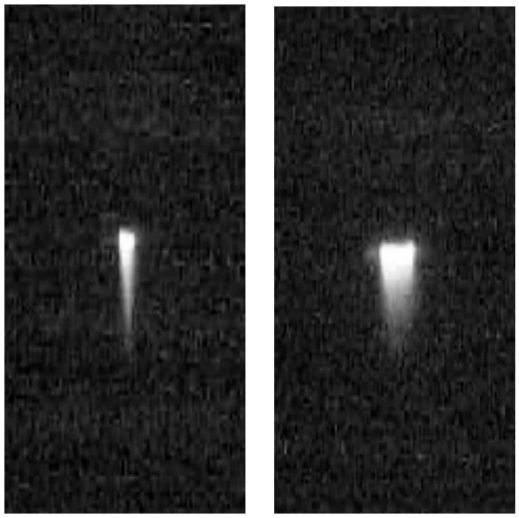


Fig.2. The plasma jet generated by the different diameters nozzle.

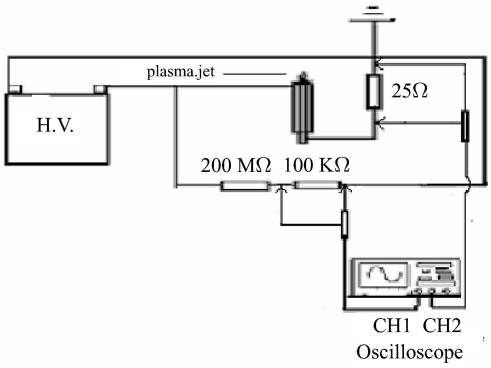


Fig.3. The circuit of the plasma jet generator.

the same method to measure the ozone concentration produced by cold plasma jet. A schematic diagram of the optical setup is shown in Fig.4. A mercury lamp is mounted on the side of the plasma jet, while the lenses for gathering light are placed on the other side. Pinholes, approximately 2 mm in diameter, are placed in front of the lamp and the lenses to eliminate any stray light. Light pass through the lenses into a photomultiplier tube (PMT). The average absolute concentration of the ozone generated by plasma is calculated from the reduction in the Hg I emission intensity at 253.6 nm.

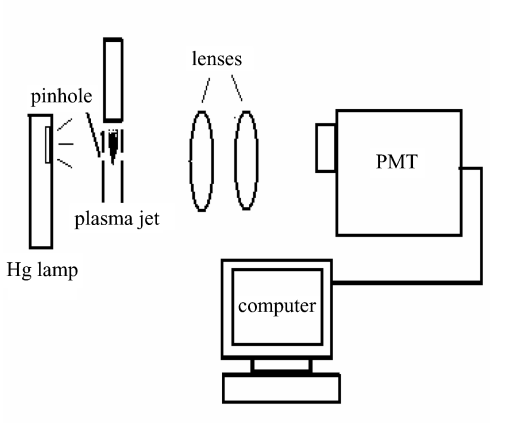


Fig.4. Schematic diagram of the apparatus used for measuring the ozone.

3. Results

3.1. The electricity character of the discharge

We use the digital real-time oscilloscope (TDS 200-Series) to record the waveform of the discharge peak-to-peak voltage (dashed) and peak-to-peak current (solid), the typical voltage and current waveforms of the cold plasma jet are shown in Fig.5. The discharge peak-to-peak voltage is 33.6 kV and the discharge peak-to-peak current is 41.6 mA. The nearly

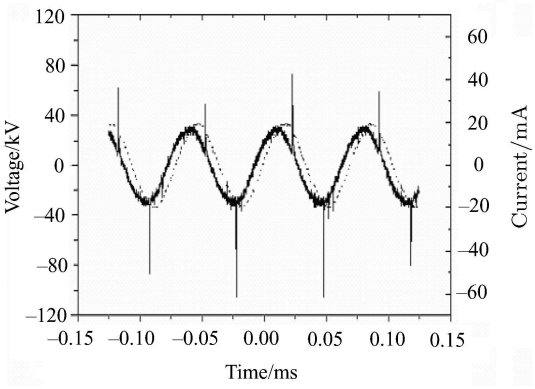


Fig.5. Waveforms of the discharge voltage (dashed) and the current (solid).

sinusoidal waveforms indicate a mostly linear response of the discharge, while the capacitive nature of the cold plasma jet discharge is clearly demonstrated by the current waveform which leads the voltage waveform. The current waveform is composed of the sinusoidal current waveform and the spike-like current pulses. The sinusoidal current waveform is the current which flows through the electrodes and the dielectric, the spike-like current pulses represent the effective discharge current. If we presume that the sinusoidal waveforms are the plasma current, according to horse-power formula

$$P = \frac{\int_0^{\frac{T}{2}} U(t)I(t)\mathrm{d}t}{\int_0^{\frac{T}{2}} \mathrm{d}t} \tag{1}$$

we can calculate that the power supplied to the plasma is about 150–200W, but it is too high for the low temperature (30°C) of the plasma gas. On the contrary, if we consider the spike-like current pulses to be the discharge plasma current, the power is about 14–20 W, and it is more reasonable.

3.2. The active species generated by the cold plasma jet

The ozone plays an important part in disinfection,[1,13] so we do quantitative measurement for it. Using the ultraviolet absorption of Hg 253.6 nm line emission from a mercury lamp, we obtain the absolute concentration of ozone of the cold plasma jet, and this absorption spectrum is shown in Fig.6.

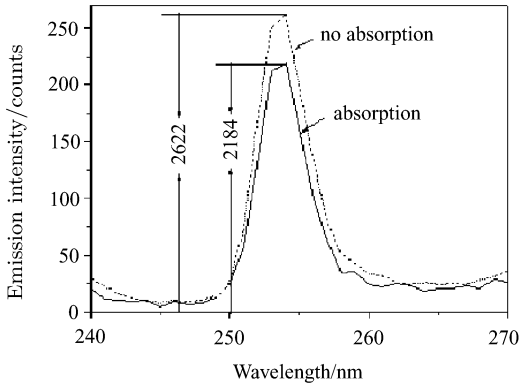


Fig.6. No absorption (dashed) and absorption (solid) of the Hg 253.6 nm line emission.

We record the emission intensity of the Hg 253.6 nm by a PMT when the Hg lamplight pass through the plasma jet, and compare this emission intensity with another one not passing through the plasma jet. Assuming absorption cross-section of O_3 at this wavelength, 1.16×10^{-17} cm^2,[12] we can calculate the concentration of ozone by Beer's law:

$$\ln(I_\mathrm{o}/I) = ecL. \tag{2}$$

where I_o is original intensity, I is the intensity after absorption, e is the absorption cross-section of ozone at this wavelength, c is ozone concentration, and L is the path length. The emission intensity of the Hg 253.6 nm is not reduced when the Hg lamplight pass through the discharge region, but decrease downstream 1–2 cm of the discharge region. The feed

gas of the plasma jet is pure argon, so the discharge can not generate any ozone, and for this reason the emission intensity of the Hg 253.6 nm does not reduce in the discharge region. However, on the edge of the discharge region the oxygen molecules in air become oxygen atoms by electron impact dissociation, and these oxygen atoms (we can find the emission line of oxygen atom in Fig.7) recombine with other oxygen molecules of air outside discharge region and generate ozone. Consequently, the ozone concentration rapidly increases downstream 1–2 cm of the discharge region and it is about $1.0 \times 10^{16}\mathrm{cm}^{-3}$.

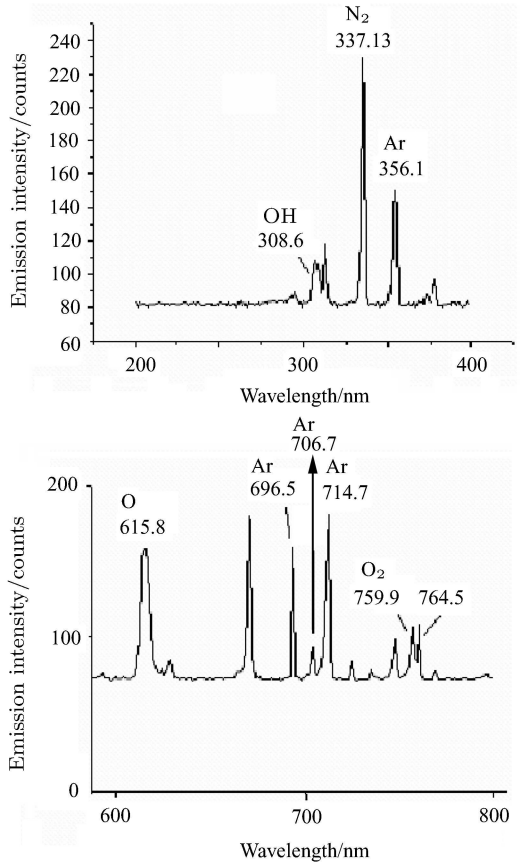


Fig.7. The optical emission spectrum of the plasma jet.

The optical emission from an argon cold plasma jet is analysed and the optical emission spectrum is shown in Fig.7. We can see the emission lines of OH (308.6 nm), O (615.8 nm) and O_2 ($^1\Sigma_g^+$) (Singlet-Sigma Metastable Oxygen) (759.9 nm, 764.5 nm) clearly, and these active species had effect on sterilization.[1,10]

3.3. Application of the cold plasma jet

Using a cold plasma jet generated from pure argon, we examine decontamination of E.coli bacteria placed 1.5 cm away from the plasma jet. The treated and untreated E.coli bacteria are cultured in incubator about 18–24 hours. After 18–20 hours' culture, we account the number of the treated and untreated E.coli bacteria to get the results of the decontamination. Comparing with cold plasma torch and APPJ, we develop this cold plasma jet possessing lower temperature (25–30 °C), and we can exclude the possibility of high temperature sterilization. Because we detect the ozone downstream 1–2 cm of the cold plasma jet, we estimate that the primary effect for sterilization is ozone. The surviving fraction versus the sterilization time is shown in Fig.8, and $S(\%)$ is the surviving fraction. In addition, the area of the cold plasma jet to the area of the E.coli is about 1/22, and it illustrates that using the cold plasma jet to sterilization can obtain fair result.

When we use the same method (placing the Bacillus subtilis bacteria 1.5 cm away from the plasma jet) to decontaminate the Bacillus subtilis bacteria, the experimental results show that the treated Bacillus subtilis bacteria have almost no difference with the untreated, and we can deduced that the ozone generated by the cold plasma jet cannot decontaminate

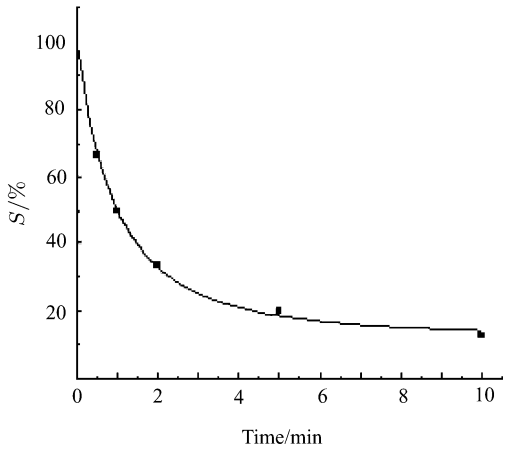


Fig.8. The result of E.coli treated by the ozone generated by plasma jet.

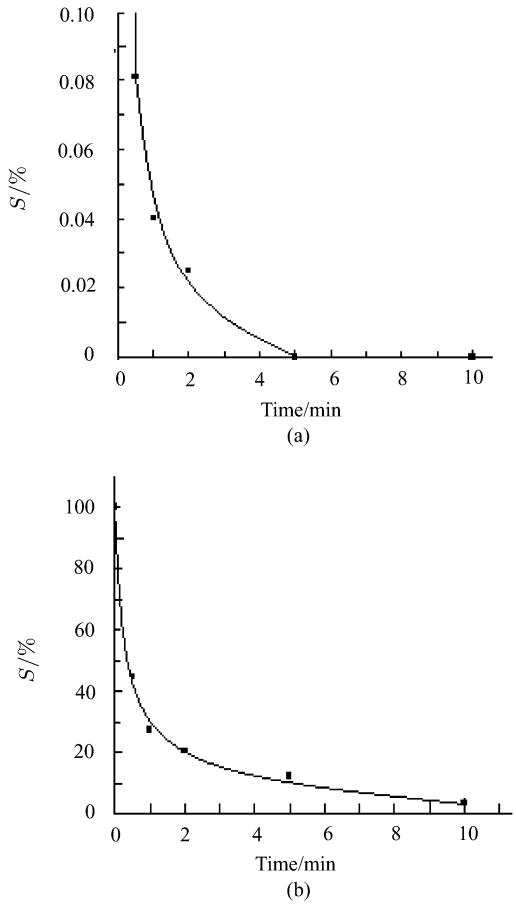


Fig.9. The result of E.coli (a) and Bacillus subtilis (b) treated by the charged particles and the active species (not including the ozone) generated by plasma jet.

Bacillus subtilis. The charged particles (produced by atmospheric pressure plasma) and the active species (such as OH, O and metastable oxygen) may play a role in very significant sterilization,[1] and we make the cold plasma jet (because the gas temperature of plasma jet is so low) contact with the cultures (E.coli and Bacillus subtilis). We get much great better results than before, as shown in Fig.9. These results elucidate that the charged particles and active species (not including ozone) are more effective than the ozone for sterilization.

4. Conclusions

A new plasma generator has been developed, which can produce a stable cold plasma jet at atmospheric pressure by using the pure argon, and the size of the cold plasma jet is varied with the diameter of the nozzle and the gas flow rate. The concentration of ozone generated by cold plasma jet is acquired by using the ultraviolet absorption spectroscopy, and the preliminary optical emission analysis is presented in this paper. We use the cold plasma jet for sterilization and get the good results. The cold plasma jet is expected to be used not only for sterilization reported here, but also for many other purposes such as surface modification, thin film deposition and etching.

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