# Atomic Oxygen and Hydroxyl Radical Generation in Round Helium-Based Atmospheric-Pressure Plasma Jets by Various Electrode Arrangements and Its Application in Sterilizing *Streptococcus mutans*

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***Abstract*— The reactive chemical species generated in an atmospheric-pressure plasma jet (APPJ), such as hydroxyl radical and atomic oxygen, are generally regarded as the major sources for several biomedical applications. In this paper, we propose an APPJ with a 25-kHz high-voltage powered stainless steel tube fixed inside a quartz tube and an aluminum grounded electrode that is wrapped around the quartz tube. The results show that the radical intensities of atomic oxygen and hydroxyl radical, gas temperature, and power absorption increase with increasing distance of discharge gap distance under the same applied voltage. Increasing the width of grounded electrode increases gas temperature and causes higher power absorption but only slight change of radical generation. We have applied the APPJ with the conditions of highest radical generation (grounded electrode width = discharge gap distance = 15 mm) for effectively sterilizing *Streptococcus mutans* within 15 s, which shows a highly potential application for root canal disinfection.**

***Index Terms*— Atmospheric-pressure plasma, dental sterilization, discharge gap, electrodes arrangement, plasma medicine.**

## I. Introduction

A SUCCESSFUL oral treatment relies on the eradication of bacteria from the root canal ($\sim$0.1 mm in diameter) and the complex 3-D networks of dentinal tubules ($\sim$1–3 $\mu$m). The tubular infection usually occurs to the depth of $\sim$1 mm [1]; however, conventional disinfection agent (sodium hypochlorite and chlorhexidine) penetrates merely $\sim$0.13 mm into the dentinal tubules due to the problem of surface tension [2], [3], even by enlarging the root canal up to $\sim$3–5 mm. Gas discharge jet plume can potentially penetrate the pores easily without the issue of surface tension. This feature gives plasma jet a great potential in root canal disinfection by enlarging the root canal to a minimal extent. In this specific application, operation at atmospheric pressure with low-gas temperature is necessary to ensure the convenience and safety that is strongly required.

Reactive oxygen species (ROS)/reactive nitrogen species have significant central roles in the process of sterilization, cancer therapy, and wound healing [4]. Most of the studies used ROS, such as hydroxyl radical and atomic oxygen, generated in atmospheric-pressure plasma jets (APPJs) to kill the bacteria [5]–[9]. Previous studies show that the ROS generation and sterilization rate could be enhanced by several means, including oxygen addition [7], [10]–[14], hydrogen peroxide addition [15], and using different power sources (frequency, voltage waveform, and power) [16]–[20]. Among these, there seem to very few studies focusing on systematic understanding of ROS generation and sterilization using different kinds of electrode arrangement, even though the rearrangement of the electrodes may be the easiest and cost effective way for achieving these goals.

Thus, in this paper, we would like to report the development and characterization of a helium-based round APPJ. We enhanced radical generations, especially atomic oxygen and hydroxyl radical, through various kinds of electrode arrangement under the same powered condition. We then applied the APPJ with the optimal operating condition that has the highest radical generation to sterilize *Streptococcus mutans*, which is the primary causative agent in the formation of dental cavities or secondary dental caries after dental filling surgery [5], [6].

## II. Experimental Methods

### A. Plasma JET

Fig. 1 shows the schematic diagram of the experimental setup of the dielectric barrier discharge type APPJ along with a typical photo of the downward plasma jet plume. The APPJ device consists of a quartz tube, a stainless steel capillary tube, an aluminum-made stepped nozzle, and an aluminum foil. The quartz tube with an inner and outer diameter of 4 and 6 mm, respectively, served as a dielectric layer. The powered electrode, including a stainless steel capillary tube (1.5 mm in diameter) and an aluminum-made stepped nozzle (0.5 mm in diameter), was inserted into the quartz tube tightly

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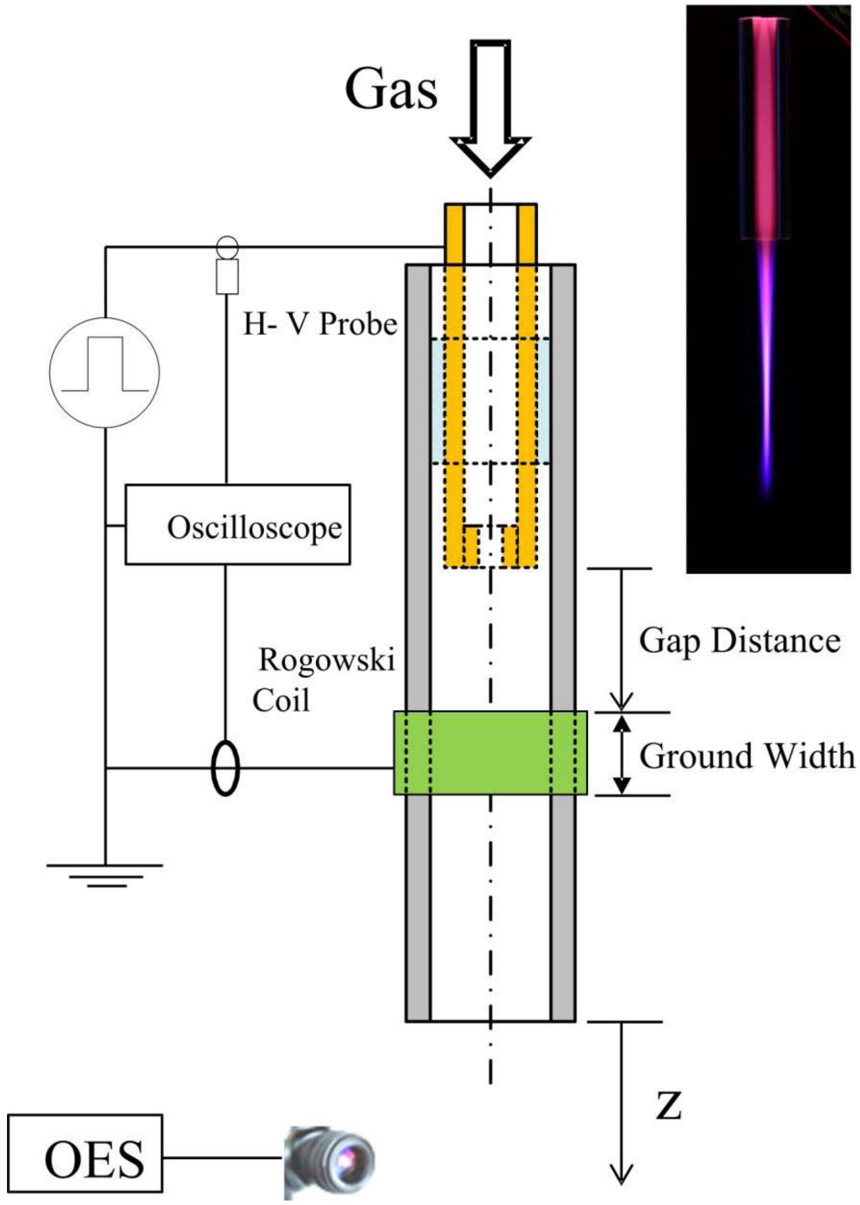



Fig. 1. Schematic diagram of the experimental setup of helium round APPJ.

with polytetrafluoroethylene (PTFE) sealing tape. The stepped nozzle was used to accelerate the gas speed for elongating the jet plume length and reducing the gas temperature. An aluminum foil with a thickness of 0.3 mm was wrapped outside the quartz tube as the grounded electrode. In this paper, the width and location (or gap distance with powered electrode) of grounded electrode were varied from 5 to 20 mm and $-5$ to 15 mm, respectively. Note the gap distance is defined as the position of the leading edge of the grounded electrode from the end of the stepped nozzle in the gas flow direction. When it becomes negative, it means that the grounded electrode overlaps with powered stepped nozzle with a quartz tube in between.

The APPJ device was driven by a unipolar pulsed power source with typical voltage and current waveforms shown in Fig. 2 (grounded electrode width = gap distance = 15 mm). Throughout the study, the applied voltage was fixed at 6 kV in amplitude and 25 kHz in frequency unless otherwise specified. Voltage and current signals across the electrodes were measured by a high-voltage probe (Tektronix P6015A) and a Rogowski coil (IPC CM-100-MG, Ion Physics Corporation Inc.) with a digital oscilloscope (Tektronix TDS1012B). The gas temperature and optical emission spectroscopy (OES) in the postdischarge region were measured by an alcohol thermometer and a monochromator (PI Acton SP 2500) with a photomultiplier tube (Hamamatsu R928), respectively. Plasma plume visualization was performed by a digital SLR camera (NIKON D7000).

We used helium (99.99%) as the discharge gas at a flow rate of 5 slm throughout the study. The corresponding Reynolds number based on the inside diameter of the quartz tube (4 mm) and the average gas speed at the exit ($\sim$6.6 m/s) is 217.5, which is a typical laminar flow in the quartz tube. Fig. 1 also shows a typical photographic visualization having a plume

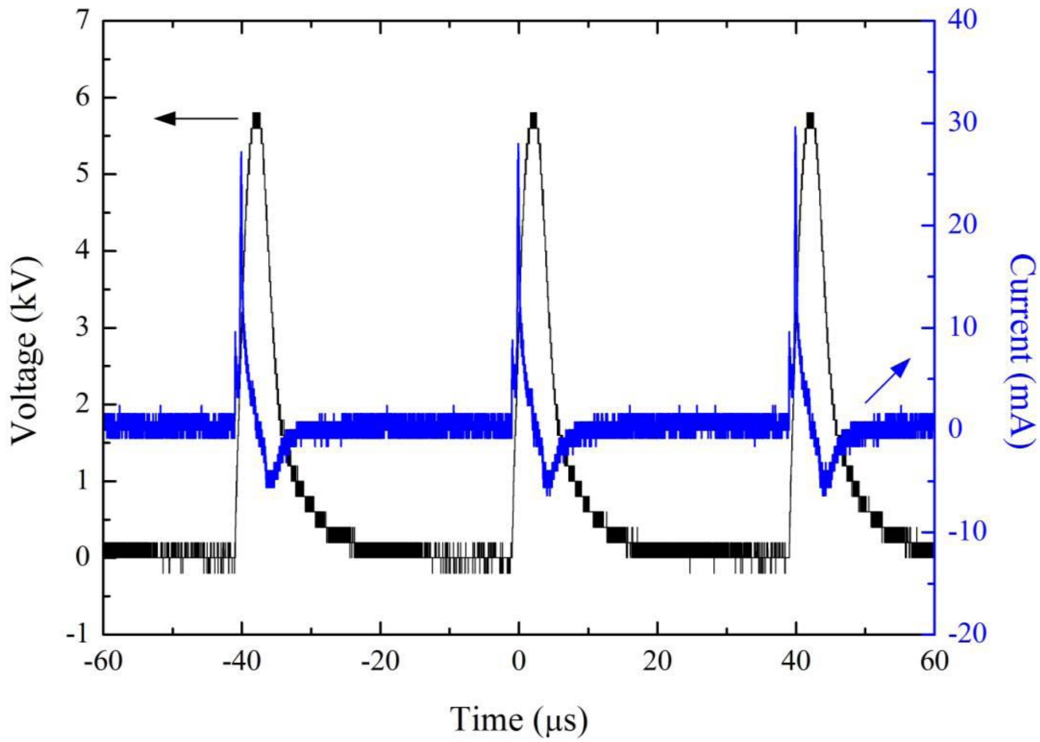
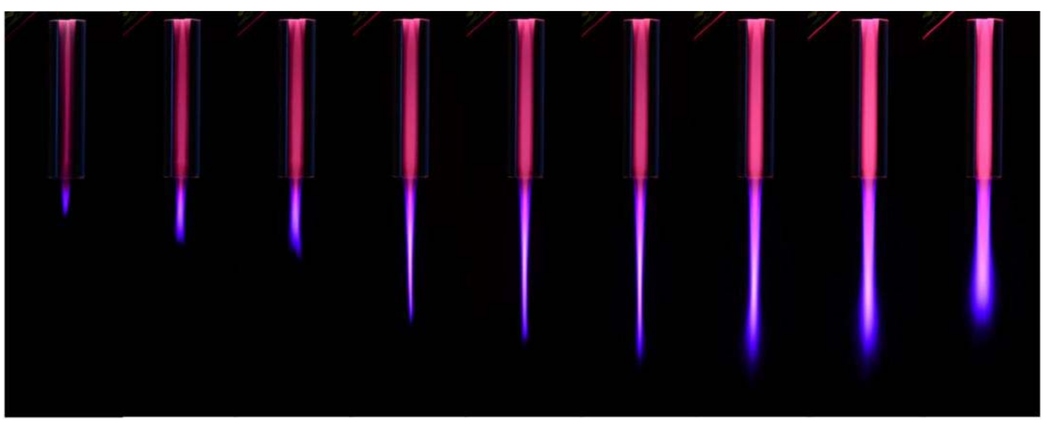



Fig. 2. Typical electrical properties of the helium APPJ.



| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| *F | 2.5 | 3 | 3.5 | 4 | 4.5 | 5 | 5.5 | 6 | 6.5 |
| *L | 8 | 14 | 17 | 32 | 35 | 40 | 42 | 40 | 33 |

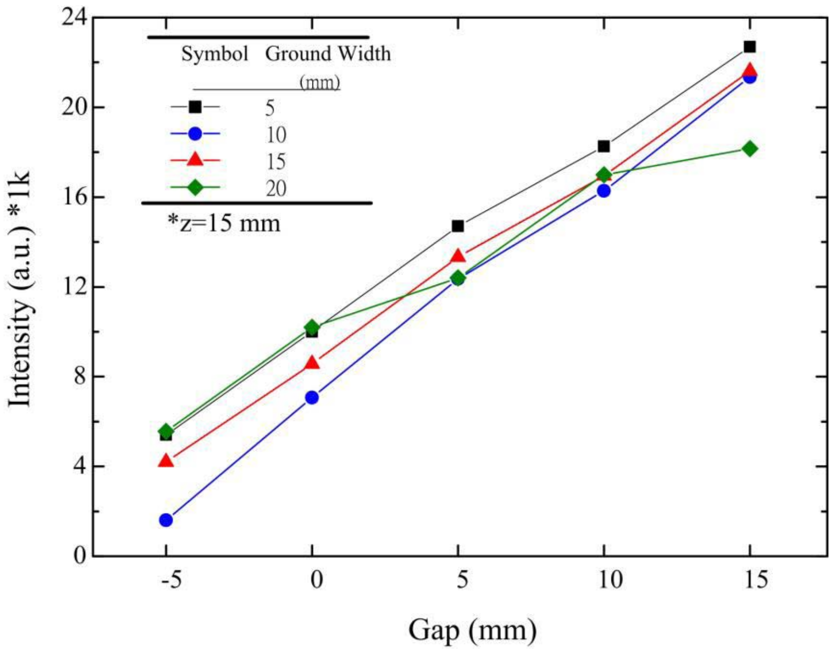
*F: Flow Rate (slm)
*L: Plasma Length (mm)

Fig. 3. Photographic visualizations of the APPJ plume with ground width = gap distance = 15 mm.

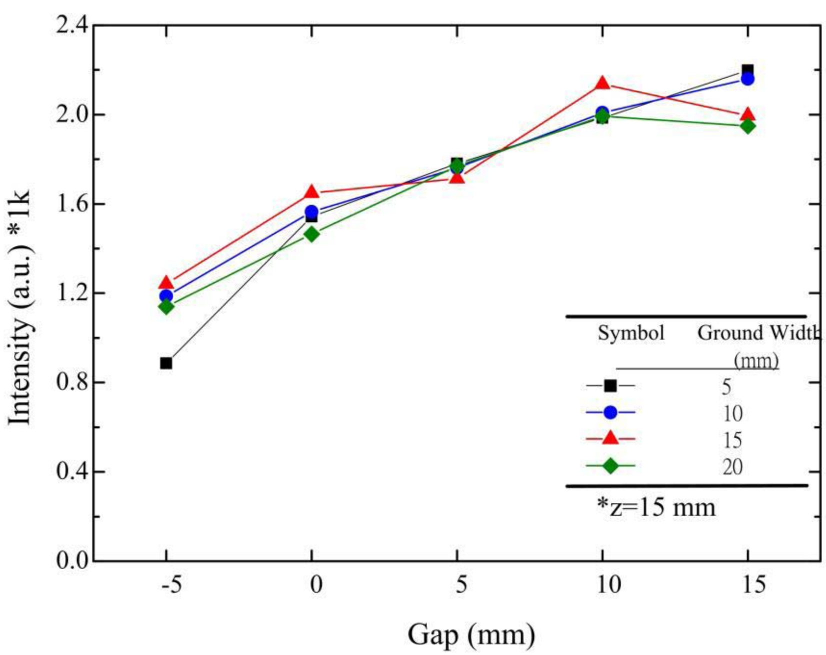
length of ∼40 mm with a grounded width of 15 mm and a gap distance of 15 mm. The jet appearances under other electrode kinds of arrangement remain similar, which will be shown in detail later.

### *B. Microbe Inhibition Experiment*

In this paper, we applied the developed helium APPJ to sterilize bacteria, named *S. mutans*, which is commonly found in root canal and dentinal tubules of dental treatment. *S. mutans* was first cultivated in a 3-ml brain heart infusion (BHI) medium for 12 h. A hundred microliters of the microbe suspension was cultured on 90-mm BHI agar plates with the density of approximately 6log10 c.f.u./cm$^2$ and dried for 15 min at 37°. The agar plates are divided into four equal partitions for comparison based on the treating time: control (without any treatment), 15 s, 30 s, 60 s, and the treating distance was kept as 15 mm away from the exit of APPJ throughout the study. After treatment, all of the plates were incubated at 37° for 12 h.




(a)




(b)

Fig. 4. Measured OES of (a) hydroxyl radical and (b) atomic oxygen.

## III. Results and Discussion

### *A. Plasma JET Characterization*

Fig. 3 shows a series of photographic visualizations of the plasma jet plume with various gas flow rates under the same arrangement of electrodes (grounded electrode width = gap distance = 15 mm). The results show that, with increasing helium gas flow rate, the plasma jet length increases first, then reaches a maximum (∼42 mm) at 5–5.5 slm and decreases afterward. In addition, the jet plume diameter first shrinks,

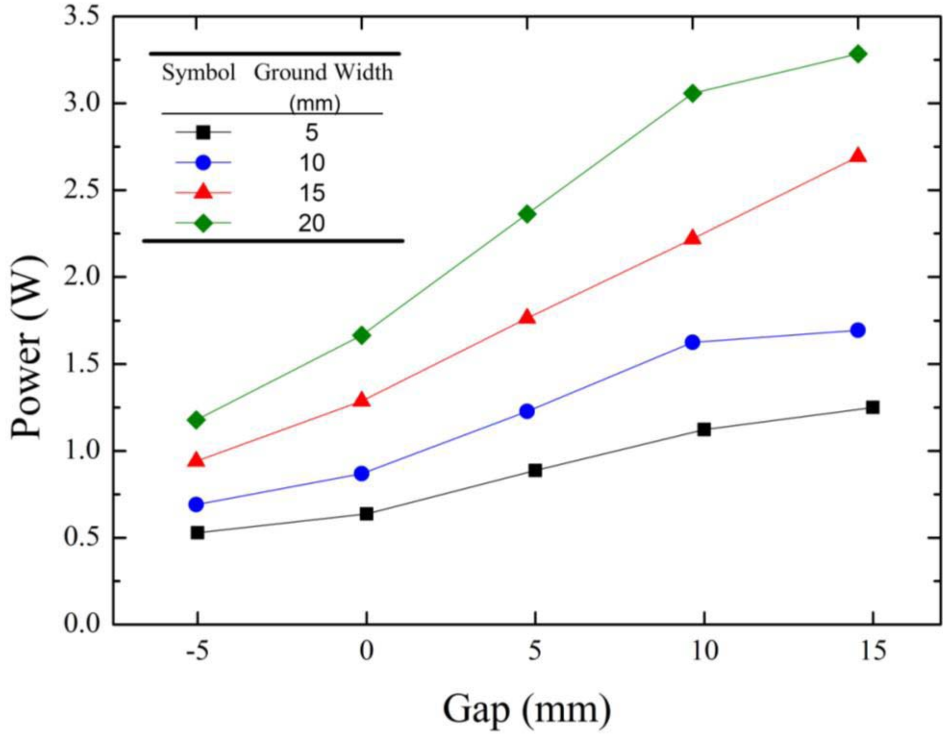



Fig. 5. Power consumptions of the APPJs.

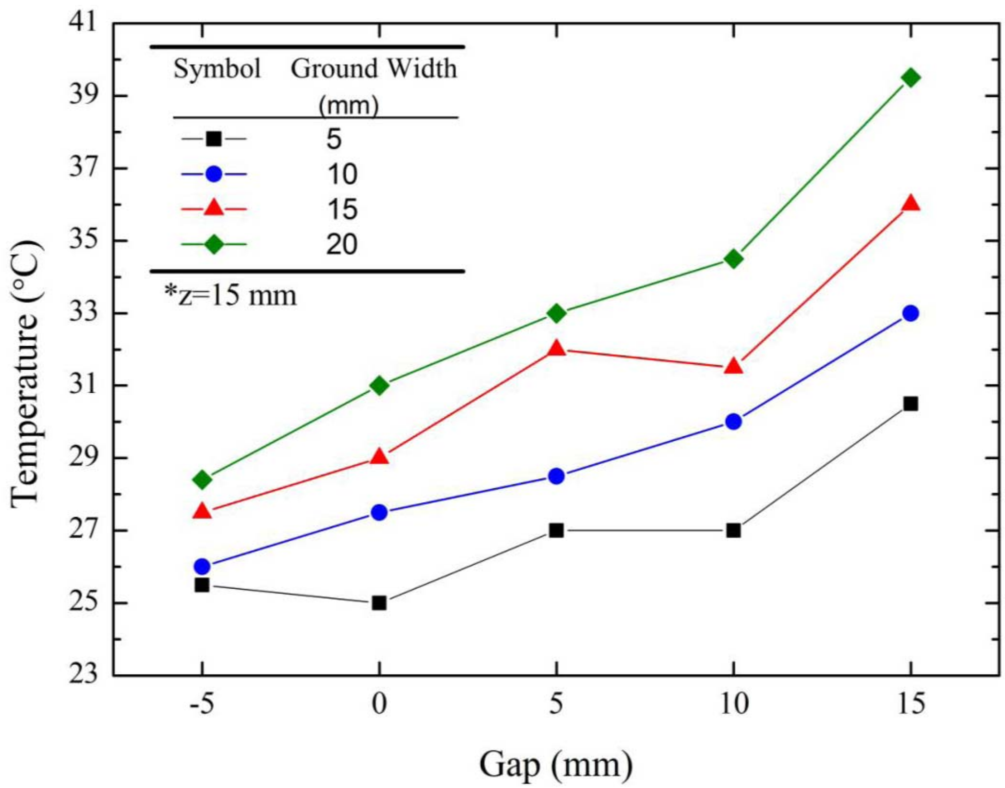
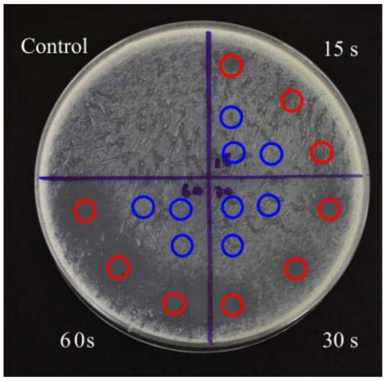


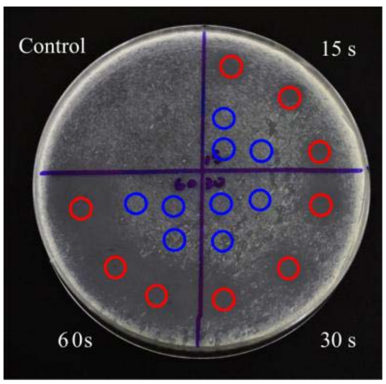

Fig. 6. Temperature measurements at postdischarge region ($z = 15$).

and then expands rapidly and becomes unstable for increasing larger gas flow rates (>5.5 slm) mainly because of efficient mixing with ambient air when the Reynolds number becomes larger. Thus, for all the results presented next we have fixed the gas flow rate as 5 slm.

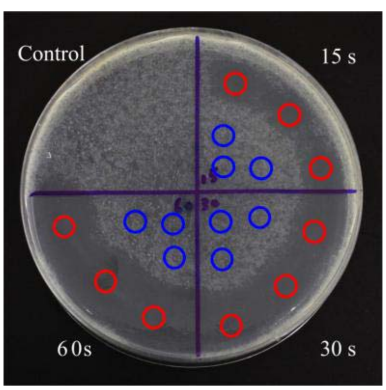
Fig. 4 shows the OES intensities (a.u.) of hydroxyl radical (309 nm) and atomic oxygen (777 nm) in the postdischarge region ($z = 15$ mm) with various kinds of electrode arrangement. The results show that both intensities increase with increasing discharge gap distance almost linearly and they do




(a)




(b)




(c)

Fig. 7. Sterilization results treated three plasma conditions. (a) Ground width/gap = 5/−5 mm. (b) Ground width/gap = 15/−5 mm. (c) Ground width/gap = 15/15 mm.

not change much when the grounded electrode width varies, except when the gap distance is very small or negative. This trend is especially true in the generation of atomic oxygen. OH radical and O atom intensities becomes six and two times larger, respectively, when the discharge gap distance increases from −5 to 15 mm. Surprisingly, the trend of radical generation related to the discharge gap distance contradicts general physical intuition that a larger gap distance induces a smaller electric field which should produce a weaker gas discharge. The reason may lie in the fact that a longer discharge gap distance provides a longer region for charge accumulation at the dielectric surface, which probably leads to a stronger charge overflow in the postdischarge region [21], [22]. This may in turn dissociate more the water vapor and oxygen in the postdischarge region, which results in higher concentrations of hydroxyl radical and atomic oxygen.

The power absorption of the gas discharge was calculated by the temporal cycle average of integrating voltage and current over time. Fig. 5 shows that the power absorption (0.5–3.2 W) increases with increasing grounded electrode width and gap distance. This observation supports our previous argument that a larger discharge gap distance provides a longer charge accumulation and thus higher current. Similar trends are also found for gas temperature in the postdischarge region ($z = 15$ mm), which is shown in Fig. 6. It is seen that appreciable amount of power absorption goes to gas heating through the ionic joule heating because of the low driving frequency (25 kHz) and collisions between ions and helium gas, even though a higher amount of radicals are generated with larger discharge gap distance.

### B. *Sterilization of Streptococcus mutans*

Fig. 7 shows the incubated agar plates containing *S. mutans* under different kinds of electrode arrangement with the same helium gas flow rate (5 slm) and treating distance (15 mm). Note the control partition stands for the case without any treatment, while the other partitions with time (seconds) represent the treatment time. For the same treatment time, the blue circles near the central region of the plate represent those cases treated using pure helium gas flow, while the red circles near the outer edge of the plate represent those cases treated with plasma plume. The results show that it took at most 15 s to totally remove the bacteria, considering the size of the jet plume impinging at the agar plate, when the grounded electrode width and gap distance are both kept as 15 mm [Fig. 7(c)]. For other arrangements of electrodes with lower levels of radical generation, it took longer time to remove the bacteria [Fig. 7(a), 30 s and Fig. 7(b), 15–30 s]. These observations show a strong correlation to the relative intensities of OH and O measured by OES (Fig. 4). This clearly demonstrates that the higher OH/O generation, the faster inactivation rate.

## IV. Conclusion

A helium-based APPJ is presented in this paper. OES analysis shows that the amounts of both hydroxyl radical and atomic oxygen, which are considered to be important agents in many biomedical applications, increase with increasing both grounded electrode width and discharge gap distance. Relative OH radical and O atom intensities present six and two times larger from gap of −5 mm to 15 mm. Measurements of power absorption and gas temperature exhibit a strong correlation to the grounded electrode width and discharge gap distance. *S. mutans* inactivation test indicates that sterilization rate increases with enhanced OH/O radical intensities. In this paper, when the grounded electrode width and discharge gap distance are both 15 mm, the low-temperature helium APPJ (∼35°) can effectively remove the *S. mutans* within 15 s, which may be highly potential applications in root canal sterilization.

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