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Length control of He atmospheric plasma jet plumes: Effects of discharge parameters and ambient air

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The effects of various discharge parameters and ambient gas on the length of He atmospheric plasma jet plumes expanding into the open air are studied. It is found that the voltage and width of the discharge-sustaining pulses exert significantly stronger effects on the plume length than the pulse frequency, gas flow rate, and nozzle diameter. This result is explained through detailed analysis of the *I-V* characteristics of the primary and secondary discharges which reveals the major role of the integrated total charges of the primary discharge in the plasma dynamics. The length of the jet plume can be significantly increased by guiding the propagating plume into a glass tube attached to the nozzle. This increase is attributed to elimination of the diffusion of surrounding air into the plasma plume, an absence which facilitates the propagation of the ionization front. These results are important for establishing a good level of understanding of the expansion dynamics and for enabling a high degree of control of atmospheric pressure plasmas in biomedical, materials synthesis and processing, environmental and other existing and emerging industrial applications. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3119212]

I. INTRODUCTION

Atmospheric-pressure thermally nonequilibrium plasmas (APTNPs) have recently been of significant interest for a large number of emerging applications such as surface modification and materials processing,[1–3] biomedical and chemical decontamination and sterilization,[4–16] thin film deposition,[17] synthesis of nanomaterials,[18–22] and water purification.[23–25] Among APTNPs, plasma jet devices have indisputable advantages owing to the absence of confining electrodes or chamber walls which are unavoidable in many traditional discharge configurations.[26–32] These advantages make the atmospheric pressure plasma jets (APPJs) very effective in many applications such as plasma medicine and bacterial inactivation.[33–37]

Some of these plasma jet devices generate plasma plumes that expand into the surrounding air, typically for a few centimeters and even longer. Even though electrons have energies in a few eV range, the gas stream remains at almost room temperature and the jet does not cause any thermal or electric shock upon contact with human skin. High-speed optical imaging reveals that some of the plasma plumes that resemble continuous plasma streams are actually bulletlike plasma clumps with typical propagation speeds of 10^4-10^5 m/s.[38–43] However, although observations and applications of APPJs are quite common, the physical mechanisms that lead to the generation of these relatively long nonthermal plasma plumes are not fully understood. In particular, the issue of determining the most effective control of the plume length and other characteristics still remains essentially open. It also remains unclear how the interaction of the surrounding air with the plasma stream through the large lateral plume surfaces that are normal to the ionization front affects the APPJ length.

This paper reveals the most effective discharge control parameters that affect the length of the plasma stream. The effects of variation of the applied voltage V_a, the pulse width t_{pw}, the pulse frequency f, the flow rate q of the working gas, and the diameter of the jet nozzle D on the APPJ length L_{pla} are systematically quantified and interpreted in terms of basic discharge maintenance phenomena. In a dedicated experiment, it is demonstrated that by enclosing the plasma plume by a thin glass tube, one can significantly reduce the effect of the surrounding air which results in remarkably longer APPJs.

The rest of this paper is organized as follows. The experimental details are described in Sec. II. Section III reports on the effects of the various discharge parameters on the length and current-voltage characteristics of the plasma plumes sustained in the open air and within the glass tube attached to the exit nozzle of the jet device. These results are interpreted in Sec. IV. This paper concludes with Sec. V where the main findings are summarized and the outlook for the future research is given.

II. EXPERIMENTAL DETAILS

Figure 1(a) is a schematic of the experimental setup. A high-voltage (HV) electrode is inserted into a 10 cm long quartz tube. The inner and outer diameters of the quartz tube are approximately 2 and 4 mm, respectively. The quartz tube together with the HV electrode is inserted into the hollow

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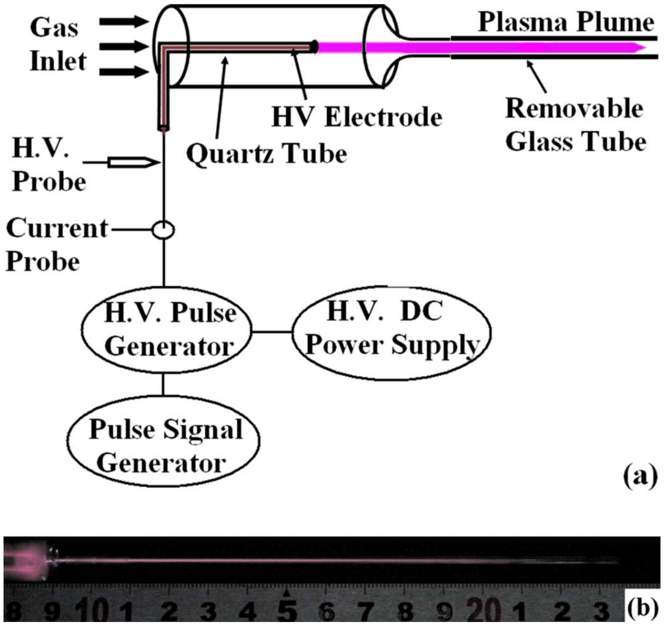

FIG. 1. (Color online) (a) Schematic of the experimental setup. (b) Photograph of the plasma. Glass tube is attached to the nozzle. Applied voltage V_a=9 kV, pulse width t_{pw}=4 μs, pulse frequency f=4 kHz, and He flow rate q=0.5 l/min.

FIG. 2. Photographs of the plasma for different applied voltage V_a. Pulse width t_{pw}=800 ns, pulse frequency f=4 kHz, and He flow rate q=0.5 l/min. Glass tube is attached to the nozzle.

barrel of a syringe. The inner and outer diameters of the hollow barrel are 13.4 and 15 mm, respectively. The inner diameter of the syringe nozzle is about 0.8 mm. The distance between the tip of the HV electrode and the gas entrance end of the nozzle is fixed at 5 mm. The HV electrode is connected to a pulsed dc power supply (amplitudes up to 10 kV, repetition rate up to 10 kHz, and pulse width variable from 200 ns to dc). The currents and the voltages are measured with current (Tektronix TCP202) and voltage (Tektronix P6015) probes, respectively. The signals collected by the probes are recorded by a digital oscilloscope (Tektronix DPO7104).

When He gas with a flow rate of a few l/min is fed through the hollow barrel and pulsed dc voltages are applied to the HV electrode, a homogeneous plasma plume is generated inside the syringe and in the surrounding air. To investigate the effect of the diffusion of the surrounding air into the He gas stream, experiments with a 14 cm long glass tube attached to the nozzle have also been conducted. The inner diameter of the glass tube is the same as the outer diameter of the nozzle near its opening. Figure 1(b) shows a photograph of the longest plasma achieved under conditions when the glass tube is attached. In this example, the plasma plume reaches a length of almost 13 cm. The APPJ length has been evaluated by high-resolution optical imaging of the discharge using a Canon digital camera with 50 ms exposure time.

III. EXPERIMENTAL RESULTS

In this section we study the effects of the most important process/system parameters such as the applied (discharge-sustaining) voltage V_a, pulse width t_{pw}, pulse frequency (repetition rate) f, He gas flow rate q, and the nozzle diameter D, on the plasma plume length and other discharge characteristics. These effects are systematically studied for both cases, namely the expansion of the APPJ into the open surrounding air and the guided plasma plume propagation through a relatively narrow channel within a glass tube attached to the nozzle.

A. Effect of the applied voltage

Figures 2(a)–2(h) show the photographs of the plasma sustained by different applied voltages V_a in the case when the glass tube is attached to the nozzle. When V_a is increased to about 5.5 kV, the plasma emerges from the nozzle. The length of the plasma L_{pla} increases linearly upon a further increase of the applied voltage. However, when V_a is increased to over ~8.0 kV, the increase in the plume length with the applied voltage becomes less steep, as can be observed in Fig. 3. Figure 3 also shows the length of the plasma plume for different V_a when no tube is attached to the nozzle. In this case, L_{pla} is much shorter (for the same applied voltage) compared to the case when the plasma propagates through the glass tube.

This observation can be attributed to the effect of diffusion of the surrounding air into the He gas stream. This dif-

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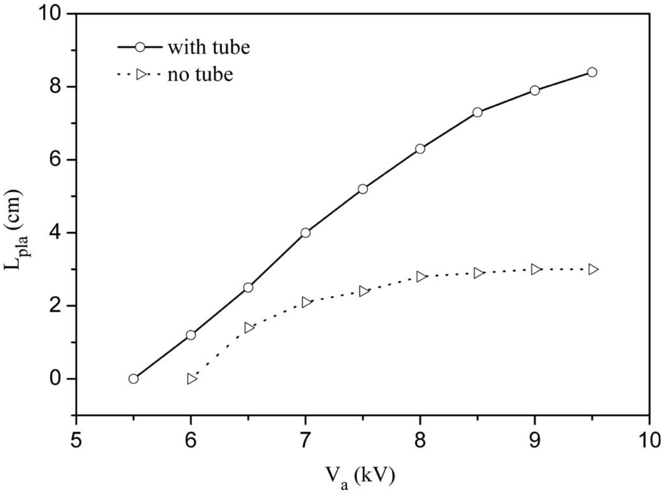

FIG. 3. The length of the plasma L_{pla} for different applied voltages. Other parameters are the same as in Fig. 2. Solid line: glass tube is attached to the nozzle, dot line: no glass tube attached.

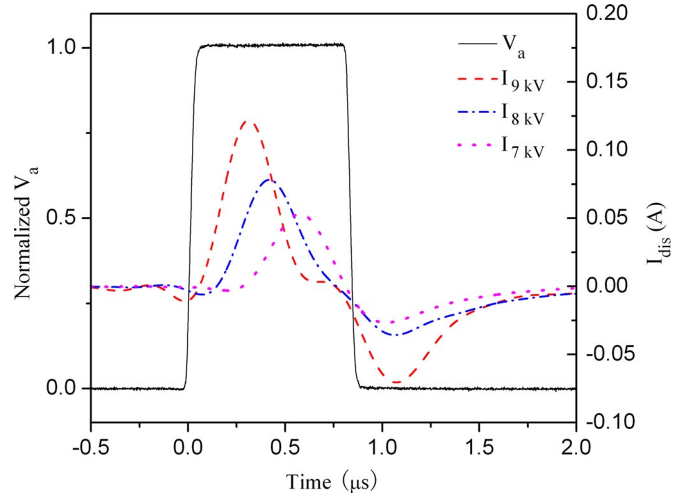

FIG. 4. (Color online) Temporal dynamics of the applied voltage and the actual discharge current I_{dis} for different values of V_a. Other parameters are the same as in Fig. 2. Since all the voltage waveforms have the same shape, their amplitudes are normalized.

fusion brings a significant amount of air molecules into the stream of He atoms and ions. The further into the open air the ionization front propagates, the more air molecules are encountered on the way. As a result, a significant fraction of the charge particles and excited species, such as He metastable states are lost through the interaction of the He gas/plasma stream with the surrounding air. This interaction involves the large lateral surfaces of the plume and becomes more intense as the APPJ elongates. Therefore, longer plasma plumes in the open air require higher voltages to sustain the discharge. When the glass tube is attached to the exit nozzle, the lateral surfaces of the plasma plume are effectively shielded from the surrounded air, which substantially reduces the charge particles losses and eventually makes it possible to achieve larger L_{pla} with the same applied voltages.

As can be seen in Fig. 3, for the case when no glass tube is attached to the nozzle, the plume length reaches about 3 cm when V_a is increased to 8 kV. Further increasing the applied voltage has no effect on the APPJ length. This observation will be interpreted in Sec. IV

Another interesting finding was that the current-voltage (I-V) characteristics of the APPJ are almost the same in the cases with and without the glass tube attached to the nozzle. Figure 4 shows the temporal dynamics of the applied voltage (solid curve) and the discharge current for different values of V_a. The current I_{dis} is the actual discharge current. As can be seen from Fig. 4, two distinctive (positive and negative) current pulses are related to two consecutive (primary and secondary) discharges, which is consistent with the results of other authors.[30,35] With the increase in V_a, both the positive (primary) and the negative (secondary) discharge currents are increased. It is instructive to note that the primary discharge current pulses I_{pri} appear at an early stage when the applied voltage is on the rise. Conversely, the secondary current pulses I_{sec} are always generated during the falling cycle of the voltage pulses. Therefore, the time lag between the commencement of the applied voltage pulse and the effective gas breakdown (the onset of the discharge current) becomes shorter and the discharge current increases significantly as the applied voltage becomes higher. Another interesting observation from Fig. 4 is that at lower V_a the secondary discharge begins almost immediately after the current in the primary discharge drops to zero. However, when V_a is increased to 9 kV, the secondary discharge is ignited approximately 200 ns after the primary discharge is extinguished.

For better understanding of the effect of the applied voltage on the length of the plasma, the peak value I_{peak} and the integrated total charge Q_{pri} of the primary discharge current have been measured for different V_a (Fig. 5). One can clearly notice that both the current peak value and the integrated total charge increase consistently with the applied voltage. This result and the relation between I_{peak}, Q_{pri}, and L_{pla} will be expanded on and interpreted in Sec. IV.

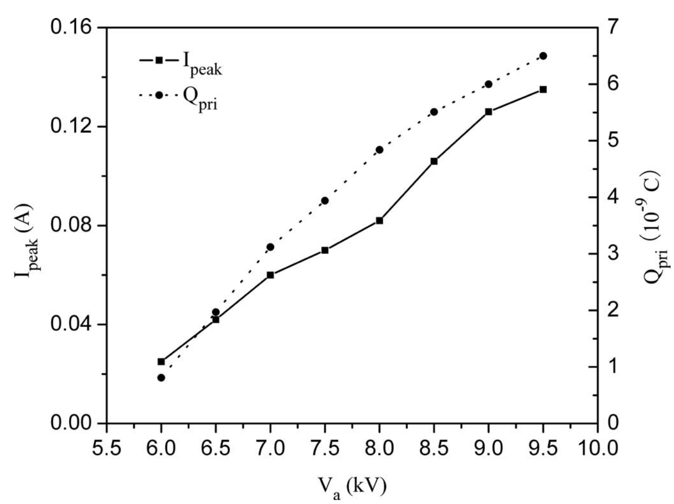

FIG. 5. The peak current I_{peak} (solid line) and the integrated total charges Q_{pri} (dot line) of the primary discharge current for different applied voltages. Other parameters are the same as in Fig. 2.

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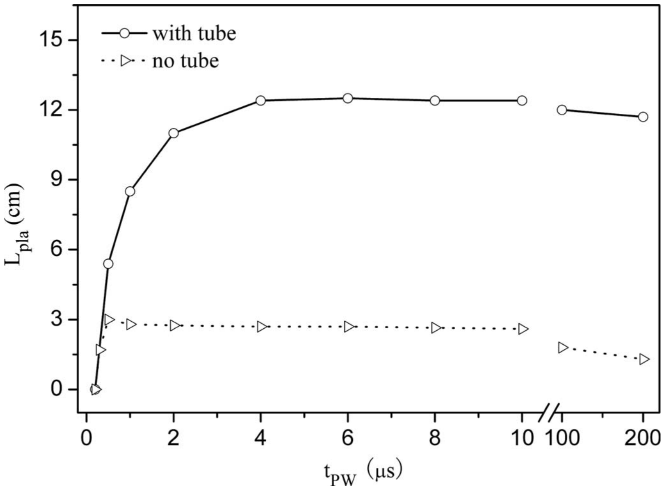

FIG. 6. The dependence of the length of the plasma L_{pla} on the pulse width t_{pw}. Applied voltage V_a=9 kV, pulse frequency f=4 kHz, and He flow rate q=0.5 1/min. Solid line: glass tube is attached to the nozzle, dot line: no glass tube attached.

B. Effect of the pulse width

The effect of the pulse width t_{pw} on the length of the plasma plume L_{pla} is quantified in Fig. 6. When the glass tube is attached to the nozzle, the plasma plume appears to be much longer. We recall that this happens mostly because of the effective shielding of the propagating He stream from the diffusion of the surrounding air.

From Fig. 6 one can observe that the length of the plasma plume is initially increased when the pulses become wider; this is the case for the situations with and without the glass tube. Without the extra tube attached, L_{pla} reaches a maximum length of about 3 cm when t_{pw} is increased to approximately 800 ns. On the other hand, when the glass tube is attached, L_{pla} reaches its maximum length of about 13 cm [see Fig. 1(b)] when the pulse width is increased to 4 μs. For both cases, after the plasma plumes reach their maximum length, a further increase in the pulse width exerts no noticeable effect on L_{pla}. If the pulse width is increased to more than 100 μs, one can notice a slight decrease of the plasma length with t_{pw} (Fig. 6).

For a better understanding of this phenomenon, Figs. 7(a) and 7(b) show the peak value I_{peak} and the integrated total charge Q_{pri} of the primary discharge current, respectively, versus the pulse width. Figure 7(a) clearly shows that when t_{pw} is increased from 200 ns to 10 μs, the peak value of the discharge current remains almost invariable. The discharge current does not change even in the range of pulse widths from 200 to 800 ns, where the plasma plume length is increased dramatically for both discharge configurations (Fig. 6). Consequently, it appears that the peak amplitude of the discharge current is not a significant factor affecting the APPJ length.

On the other hand, the integrated total charges of the primary discharge current also experience a dramatic increase [see Fig. 7(b) plotted for the discharge configuration with the glass tube attached] in the same range of pulse durations (t_{pw}=200–800 ns), very similar to the dependence L_{pla} (t_{pw}) depicted in Fig. 6. In the subsequent range of pulse widths, Q_{pri} is almost constant until t_{pw} reaches ~10 μs. Upon a further increase in t_{pw} to 200 μs, Q_{pri} starts to decrease. We emphasize that this trend is also very similar to the decrease of the length of the plasma plume in the same range of t_{pw}.

For relatively short pulse widths (t_{pw}=200–800 ns), the effect of t_{pw} on Q_{pri} can be explained as follows. In this case, there is no significant charge transfer because the applied voltage starts falling well before the primary discharge current drops to zero. However, if the pulse width is increased, the discharge current lasts longer. Accordingly, the total charge carried by the plasma plume becomes larger. We have also observed that 700–800 ns is the common range when the discharge current drops to zero (this is clearly seen in Figs. 4 and 8). This explains why a further increase in t_{pw} has no effect on the peak amplitude of the discharge current, or on the integrated charges of the primary discharge. However, the duration of the zero-current (no discharge) phase is affected by the pulse width.

When the pulse width reaches approximately 100 μs, which is comparable to the voltage off time, Q_{pri} undergoes a slight decrease. This observation requires further investigation.

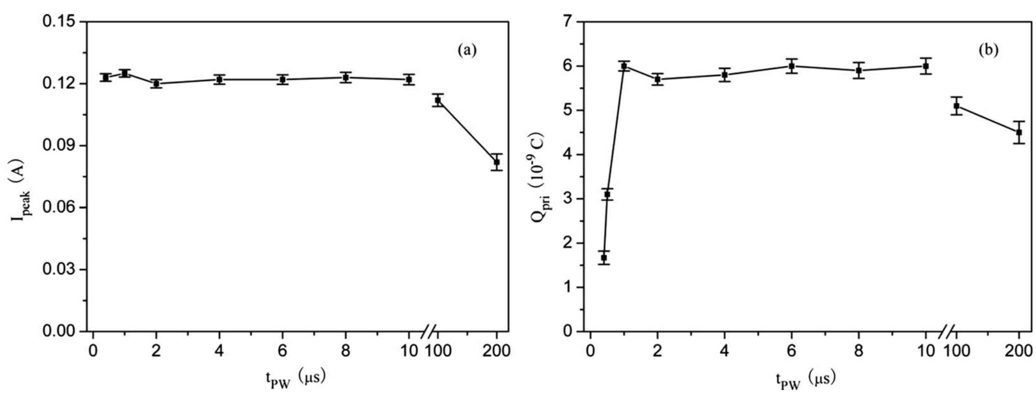

FIG. 7. (a) Peak current I_{peak} and (b) integrated total charges Q_{pri} of the primary discharge current vs the pulse width. Applied voltage V_a=9 kV, pulse frequency f=4 kHz, and He flow rate q=0.5 1/min.

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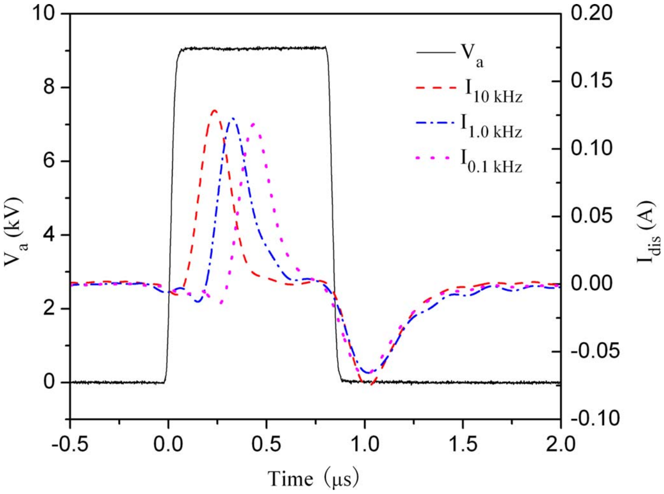

FIG. 8. (Color online) Same as in Fig. 4 for different pulse frequencies f. Applied voltage V_a=9 kV, pulse width t_{pw}=800 ns, and He flow rate q=0.5 l/min.

C. Effect of the pulse frequency

The effect of the pulse frequency (repetition rate) f on the length of the plasma L_{pla} has also been studied. The pulse frequency was varied from 0.1 to 10 kHz. The variation of this parameter did not affect the APPJ length noticeably. Figure 8 shows the temporal dynamics of the applied voltage and the discharge current for the pulse frequencies of 0.1, 1.0, and 10 kHz. One can notice a slight increase in the peak current value when f is increased. It is worthwhile to stress that the primary discharge current appears much earlier when the pulse frequency is higher. This may be related to the accumulation of excited species, such as He$(2\,^1S_1)$, N_2W $^3\Delta_u$, $a\,^1\Pi_g$, and W $^1\Delta_u$, which have lifetimes between 0.1 and 10 ms, which are comparable to the duration of the pulse off cycle. When the pulse frequency is increased, the excitation of such species becomes more effective. Consequently, larger amounts of these species are involved in the gas breakdown and discharge maintenance processes, which ultimately lead to noticeably earlier detection of the discharge current (Fig. 8).

D. Effects of the gas flow rate and the diameter of the nozzle

The flow rate of the working gas q and the diameter of the nozzle D are the other two important parameters that may affect the length of the plasma. For the case without the glass tube attached to the nozzle, with V_a=9 kV, f=4 kHz, and t_{pw}=800 ns, when the nozzle diameter D=0.8 mm, L_{pla} reaches a maximum of about 3 cm as the flow rate of the He gas is increased to 0.5 l/min. A further increase in the flow rate exerts no obvious effect on the length of the plasma plume. The latter becomes slightly shorter when the flow rate is more than ~3 l/min. This is most likely due to the effect of the turbulence of the gas flow. When the nozzle diameter of about 2.4 mm is used, at the flow rate of 0.5 l/min, L_{pla} is approximately 5 cm. The plasma plume reaches a maximum length of about 7 cm at a flow rate of about 2.5 l/min.

IV. DISCUSSION

To generate a plasma plume in the surrounding air, a He gas stream with a low concentration of air is required. Without the glass tube attached to the nozzle, for a given nozzle diameter, the maximum length L_{He} of the He gas stream is determined by the gas flow dynamics and the diffusion of the surrounding air into the plasma plume. When the applied voltage is relatively low, the plasma length L_{pla} is smaller than L_{He}. With the increase in V_a, the plasma length increases. However, when L_{pla} reaches L_{He}, any further increase in the applied voltage has no noticeable effect on the length of the plasma plume. This effect can be clearly seen in Fig. 3.

In part, this observation can be attributed to the adverse effect of diffusion of the ambient air into the He stream. Under these conditions, the concentration of air molecules inside the volume occupied by the plasma plume and in front of its ionization front reaches a critical value when the applied voltage is sufficient to maintain the discharge but is no longer sufficient to ensure further propagation of the ionization front into the surrounding air. An increase in the applied voltage from 8 to 9.5 kV does not lead to any further expansion of the plasma plume into the surrounding air. This excess voltage is most likely required for additional excitation of the molecules of the air inside the APPJ; this argument is supported by a slight increase in the plume luminosity when the voltage is increased.

When the adverse effects of the diffusion of the surrounding gas are eliminated by guiding the plasma plume into the channel of the glass tube, significantly longer [up to 13 cm as can be seen in Fig. 1(b)] plasma can be generated for the same applied voltage amplitudes. In this way, the plasma plume can be made almost three times larger at V_a=9 kV as can be seen in Fig. 3.

As mentioned in Sec. I, the plasma plume of our interest can be touched with a bare hand without any adverse effects (i.e., electric shocks). This has been explained by noting that the potential at the exit of the nozzle is only a few hundreds volts, which is much smaller than the applied voltage required to sustain the plasma. The gas temperature in the plasma plume can be estimated using common methods of optical emission spectroscopy.[44–46]

Furthermore, studies on the dynamics of similar plasma jet devices show that the APPJ contains a bulletlike plasma clump, which is isolated from the HV electrode. According to the results presented in Figs. 3 and 5, the effect of the diffusion of the surrounding air into the He stream can be eliminated by attaching a glass tube to the nozzle. Under these conditions, an increase of the applied voltage V_a leads to larger values of I_{peak} and Q_{pri} and eventually, to longer plasma plumes. Consequently, the increase in I_{peak} and/or Q_{pri} is directly related to the increase in L_{pla}. We also stress that when the pulse width is increased from 200 to 800 ns both Q_{pri} and L_{pla} increase dramatically whereas the peak current shows no significant changes (Figs. 6 and 7).

Therefore, this remarkable correlation may imply that the propagation of the plasma bullet is directly related to Q_{pri} rather than I_{peak}. This result is also consistent with the

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photoionization-based plasma bullet model.[38] The most essential point of this model is that a plasma volume with sufficient charges is required in order to sustain the propagation of the ionization front.

When no tube is attached to the nozzle, the diffusion of the surrounding air into the He stream plays a crucial role in determining the length of the plasma plume. This makes it very difficult to interpret the relationships between the length of the plasma plume L_{pla} and various parameters including the applied voltage, pulse width, peak current, and total charge of the primary discharge. On the other hand, when the glass tube is attached to the nozzle, the experimental results can be clearly explained. Further modeling and experimental research on the properties of the plasma plumes propagating in the open air are required to explain the intricate behavior of the APPJs of our interest.

V. CONCLUSION

In summary, the effects of different discharge parameters on the length of the room-temperature APPJ have been investigated. It is found that the applied voltage, the pulse width, and the diameter of the nozzle exert significant effects on the length of the plasma plume. By confining the plasma jet inside a channel of a glass tube, the effect of the diffusion of the surrounding air into the He stream was eliminated. It was found that the integrated total charge Q_{pri}, rather than the peak current I_{peak} of the primary discharge, is directly related to the propagation of the plasma plume. These results are directly related to the development of advanced biomedical and materials technologies based on flexible, cheap, and environmentally friendly APPJs. Future work will be concentrated on understanding the interactions between the plasma plume and the surrounding gas as well as exploring the ultimate physical limits of the sustainable lengths of this technologically important class of nonequilibrium plasma discharges.

ACKNOWLEDGMENTS

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