Contrasting characteristics of pulsed and sinusoidal cold atmospheric plasma jets

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Pulsed excitation of cold atmospheric plasmas is commonly believed to offer valuable benefits compared to the mainstream sinusoidal excitation. However, direct comparison of pulsed and sinusoidal atmospheric plasmas remains few, if any, thus casting an uncertainty of whether pulsed excitation facilitates any significant advantage. In this letter, we report a comparison study of pulsed and sinusoidal cold atmospheric plasma jets through electrical characterization, gas temperature measurement, and optical detection of reactive plasma species. An example of pulsed excitation is shown to reduce the electrical energy consumption by a factor of 12 for producing the same amount of oxygen atoms. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2198100]

Atmospheric pressure glow discharges (APGD) offer the prospect of a future basic technology for low-temperature processing of gaseous and solid materials without the use of a vacuum chamber. For them to impact on the future industry as profoundly as the conventional vacuum plasmas have impacted on the microelectronics industry, they must simultaneously achieve both high plasma stability and efficient reaction chemistry.[1] This is highly challenging, since these two requirements are often mutually exclusive—the former favors the use of inert gases whereas the latter demands chemically reactive gases. One solution is to generate APGD in an inert gas and then flush it to a separate region of reactive gases for material processing, thus attaining high plasma stability and active reaction chemistry in spatially different regions where they are most needed. Often this is realized in a jetlike configuration.[2–11] The vast majority of APGD jets reported so far employ sinusoidal excitation, although it has been known that nonsinusoidal excitation, through voltage pulsing, for example, offers a useful freedom to further improve APGD performance.[12] Initial results from few reported studies of pulsed APGD jets[4,10,13] have shown their general similarity to and contrast with sinusoidal APGD jets. However, a direct comparison of pulsed and sinusoidal APGD jets remains elusive, thus casting an uncertainty of whether a pulsed APGD jet offers significant advantages. Given that pulsed APGD jets introduce additional system parameters including pulse width, pulse rise time, and duty cycle, the answer to the above question will also significantly influence ways with which pulsed APGD jets can be optimized and the APGD technology can be advanced. In this letter, we present an experimental study to directly compare electrical and optical characteristics of a pulsed APGD jet and its sinusoidal counterpart.

The APGD jet used for this study employed a dielectric tube of 6 cm long, one end of which was wrapped with a concentric copper belt of 1 cm wide as the powered electrode. A stainless steel plate, either dielectrically insulated or naked, was used as the ground electrode and placed at 3–5 cm away from the gas exit point of the dielectric tube. As shown in Fig. 1, the electrode configuration was essentially that of a dielectric-barrier discharge jet. Atmospheric helium gas was fed to flow at 5 slm (standard liters per minute) through the dielectric tube and ionized by a high voltage externally applied to the copper electrode. The ionized helium gas was flushed out of the dielectric tube into the surrounding ambient air, where the excited helium species transferred their energy to ground-state oxygen and nitrogen molecules there and created excited oxygen and nitrogen species such as atomic oxygen, OH, and excited N_2/N_2^+. The generated plasma jet was very stable and can be sustained for many hours. Its volumetric appearance near the

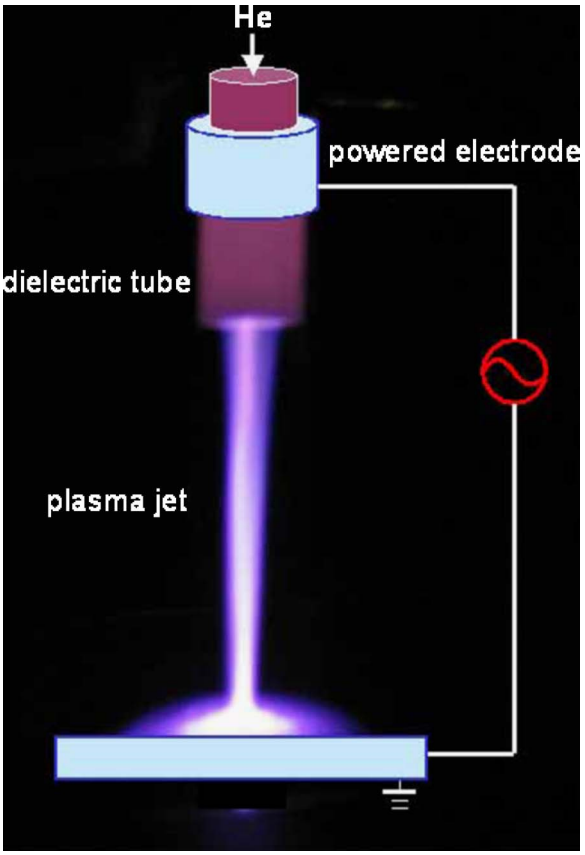

FIG. 1. (Color online) Image of a cold atmospheric plasma pen and a schematic of its electrode unit.

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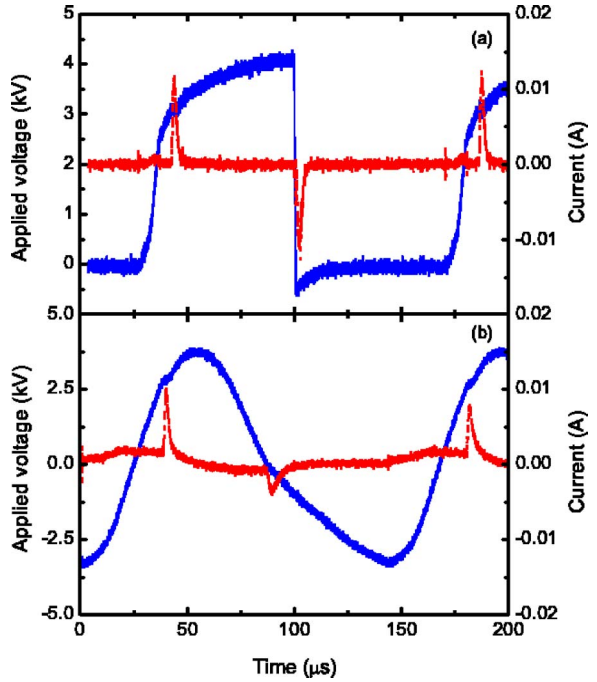

FIG. 2. (Color online) Applied voltage (blue) and discharge current (red) of the APGD jet of Fig. 1 with (a) pulsed and (b) sinusoidal excitations.

gas exit point seemed to depend largely on the applied voltage and the helium flow rate, relatively independent of the ground electrode. With dielectric insulation, the ground electrode was found to terminate the plasma jet with a spread-out plasma root as shown in Fig. 1. When the dielectric insulation was removed from the ground electrode, the plasma root was seen to converge into a small spot.

To facilitate a direct comparison of sinusoidal and pulsed excitations, we developed and constructed a sinusoidal voltage source and a pulsed power source both with a variable excitation frequency of 1–10 kHz but nominally at 7 kHz. For all results presented here, the sinusoidal source had a peak-to-peak voltage of 7.3 kV and the pulsed source delivered a unipolar pulse train having a peak voltage of 4 kV, thus both with similar voltage spans. To facilitate direct comparison, the pulse width of the pulsed source was fixed at 71 μs and this yielded a duty cycle of 49%. Parameters of the pulsed source were chosen to make its excitation voltage comparable with that of the sinusoidal source. Electrical measurements were made using a Tektronix oscilloscope (TDS 034B) via high voltage and current probes, whereas optical measurements were performed with an Andor DH720/Shamrock system at a grating of 2400 grooves/mm.

The discharge current and the applied voltage characteristics are shown in Fig. 2 for both pulsed and sinusoidal excitations. The pulsed voltage had a rise time of 30 μs and a fall time of 100 ns, providing a clear contrast with each other and also enabling easy comparison with the sinusoidal voltage that nominally required 35 μs to climb from zero to the next peak. Our pulsed voltage source was designed to have a rise time similar to one pulsed APGD jet study[4] and a fall time similar to the other,[13] thus enabling a comparison with these previous studies. With the pulsed excitation, two discharge current pulses were induced during one voltage pulse—one in the voltage-rising phase and the other in the voltage-falling phase. Although the pattern of two current pulses per one voltage pulse was not explicitly mentioned in previous studies of pulsed APGD jets,[4,13] it was observed with a pulsed APGD between parallel-plate electrodes.[14] It is expected that this temporal character is likely to be observable in most previously reported pulsed APGD jets.[4,13] As shown in Fig. 2(a), the peak current is 12.1 mA in the voltage-rising phase and 12.9 mA in the voltage-falling phase. The difference in the peak discharge current is insignificant, suggesting that the time scale of the voltage variation, 30 μs and 100 ns, respectively, was unlikely to be the only influencing factor. The pattern of the discharge current was highly periodic with the peak current being the same from one voltage pulse to another.

With the sinusoidal excitation, the discharge pattern was seen in Fig. 2(b) to have one discharge current pulse per half cycle of the applied voltage and the current pulses occurred always at a voltage-rising phase. This is similar to that in APGD sustained between two parallel-plate electrodes.[15] The discharge events were periodic, though the peak current appeared to vary from one cycle of the applied voltage to another. When the applied voltage was positive, the peak discharge current varied between 7.9 and 12.0 mA. When the applied voltage was negative, the peak discharge current became smaller. The difference in the peak discharge current is, however, not uncommon among parallel-plate APGD.[15] In general, the discharge current achieved with pulsed APGD jet was higher than that in sinusoidal APGD jet but the difference was not significant. The averaged power was 74 mW in the pulsed APGD jet and 370 mW in the sinusoidal APGD jet, representing a superior energy efficiency by a factor of 5 with the pulsed excitation for the same peak discharge current. This is significantly higher than a theoretical evaluation of a sinusoidal APGD and a comparable pulsed APGD both sustained between parallel-plate electrodes.[12] The plasma power dissipated in our pulsed APGD jet was more than one order of magnitude lower than that found in previous studies of pulsed APGD jets.[10,13]

Gas temperature was estimated using an optical spectroscopy technique[16] as shown in Fig. 3. By comparing measured and simulated optical emission of OH radicals around 309 nm with a spectral resolution of 0.3 nm, gas temperature was found to be between 290 and 350 K but closer to 290 K in both the pulsed and sinusoidal APGD jets. Essentially the difference in the measured gas temperature was insufficient to differentiate the pulsed and sinusoidal APGD jets, possibly because both plasma jets had their gas temperature either at or very close to room temperature. The dielectric tube was thermally safe to handle even after several hours of plasma operation, with both the sinusoidal and pulsed excitations. This is similar to the observation of Ref. 13.

Optical emission spectroscopy was also used to detect various excited plasma species as shown in Fig. 4. In general, atomic oxygen, OH, and various nitrogen species were observed and the general emission spectra were similar for the pulsed and sinusoidal APGD jets. In the spectral range of 200–300 nm, there were very weak emission lines with their magnitudes at most a couple of percent of the nitrogen emission line at 337 nm. Therefore UV photons were not a major plasma species in the two APGD jets studied. Additional spectroscopic measurements with greater gratings were performed (not shown) and the findings confirmed the above conclusion. It is worth mentioning that UV emission was found to be similarly weak in other studies of pulsed APGD jets.[10,13] This suggests that the APGD jets considered in this

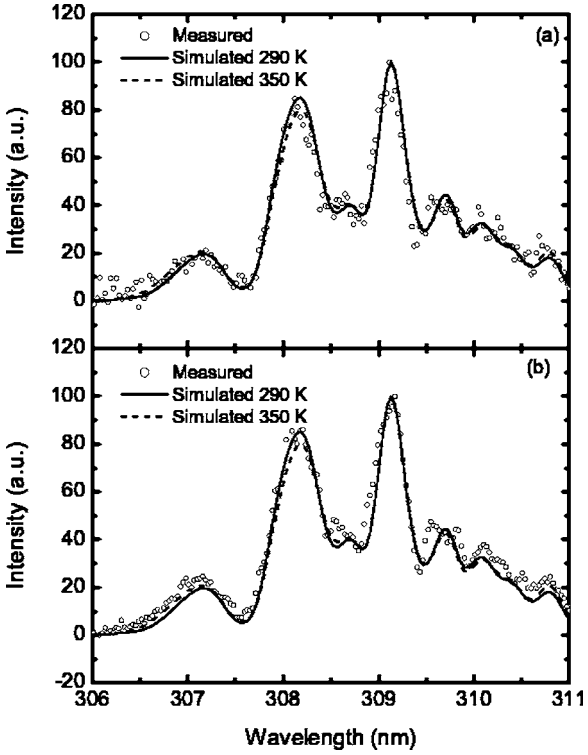

FIG. 3. Gas temperature estimated from measured and simulated optical emission of OH radicals around 309 nm with a spectral resolution of 0.3 nm for the APGD jet with (a) pulsed and (b) sinusoidal excitations of Fig. 2.

study are likely to enable applications through their reactive plasma species. Interestingly, optical emission intensities of the pulsed APGD jet were greater even though its power consumption was lower. Table I lists optical emission intensities of OH radicals at 309 nm, atomic oxygen at 616 and 777 nm, metastable helium at 706 nm, excited N_2 at 337 nm, and excited N_2^+ at 391 nm, measured with a fixed optical setup with the same alignment and distance between the spectrometer and the plasma jet. As shown in Table I, the most significant difference lies with the optical emission intensity of the atomic oxygen line at 777 nm—the emission intensity from the pulsed jet is 2.5 times greater than that from the sinusoidal jet. While the optical intensity at 616 nm was about 13% less in the pulsed jet than that in the sinusoidal jet, most oxygen atoms were produced via the channel associated with the 777 nm line. Therefore the pulsed jet was capable of producing more oxygen atoms, largely via the 777 nm channel. The emission intensity of the OH line was very similar in the two plasma jets, whereas the production of all other listed plasma species was more abundant with the pulsed plasma jet. This is both significant and desirable, as it offers an unambiguous evidence of the advantage of pulsed APGD jets as a more efficient producer of reactive plasma species.

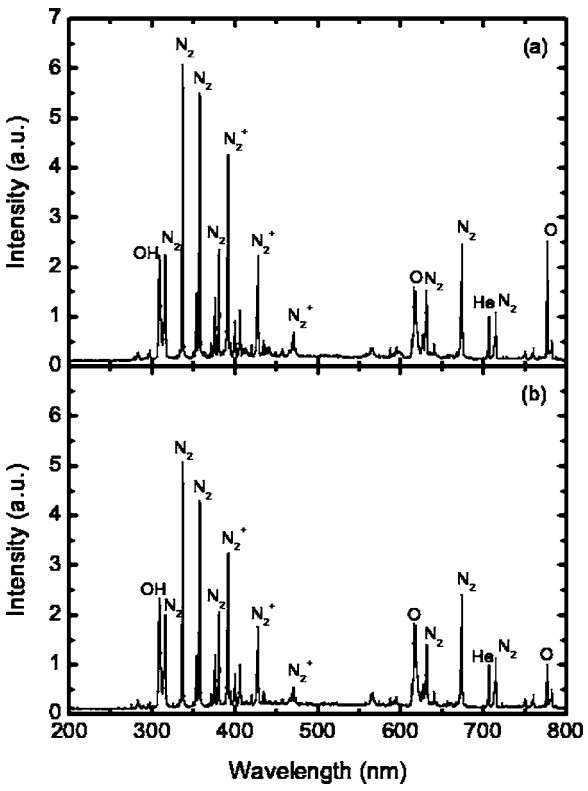

FIG. 4. Optical emission from 200 to 800 nm of the APGD jet with (a) pulsed and (b) sinusoidal excitations of Fig. 2.

TABLE I. Optical emission intensities of several plasma species.

Species	OH	N_2	N_2	O	He	O
Wavelength (nm)	309	337	391	616	706	777
Pulsed	2.22	6.09	4.26	1.59	1	2.52
Sinusoidal	2.33	5.09	3.24	1.83	1	1
Intensity ratio I_p/I_{sin}	0.97	1.20	1.31	0.87	1	2.52

We have already established from Fig. 2 that the pulsed APGD jet consumed lower electrical power by a factor of 5. To produce the same amount of oxygen atoms, the energy consumption of the pulsed APGD jet is therefore a factor of 12 ($\approx 5 \times 2.52$) lower than that of the sinusoidal APGD jet. It is conceivable that this advantage and others can be further enhanced by optimizing all pulsing parameters including pulse width, rise time, and duty cycle. In turn, these are likely to result in more superior application performance.

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