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Submicrosecond pulsed atmospheric glow discharges sustained without dielectric barriers at kilohertz frequencies

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In this letter, the authors report the experimental observation of a large-volume atmospheric glow discharge sustained without dielectric barriers at 1 kHz. This barrier-free mode of operation is made possible with a submicrosecond pulsed excitation instead of the usual sinusoidal excitation. Its current-voltage characteristics are shown to be very different from conventional atmospheric dielectric barrier discharges, and its generation mechanism is studied with nanosecond resolved optical emission spectroscopy. The pulsed barrier-free atmospheric plasma is shown to produce very intense atomic oxygen emission line at 777 nm, up to one magnitude more intensive than that of a comparable atmospheric dielectric barrier discharge. © 2006 American Institute of Physics. [DOI: 10.1063/1.2361274]

Atmospheric-pressure glow discharges (APGDs) offer a unique chamberless route to a wide range of highly desirable surface functionalities without wet chemistry and close to room temperature.¹ In the excitation frequency range of 1–300 kHz, dielectric insulation of electrodes has long been believed to be essential for APGD generation.² It is therefore of significant interest to study whether APGDs without dielectric barriers are fundamentally possible at kilohertz frequencies and, if so, whether barrier-free APGDs offer a different route to producing glow discharges at atmospheric pressure. From an application standpoint, it is sometimes desirable to dispense the use of the electrode-insulating dielectric layers that may become contaminated after continuous usage and so potentially lose their ability to stabilize APGD. So far, barrier-free APGD have been achieved largely outside the 1–300 kHz range, either at the main frequency using effectively a large ballast resistor of various forms^{3,4} or at megahertz frequencies^{5–7} both to restrict current growth. Within the 1–300 kHz range, the only reported barrier-free APGD is achieved by maintaining the sinusoidal applied voltage close to the gas breakdown voltage.⁸ In this letter, we report the experimental observation of a barrier-free APGD achieved with submicrosecond pulsed voltage instead of the sinusoidal excitation.

The submicrosecond pulsed barrier-free APGD employed two parallel stainless-steel electrodes with a surface area of 6 cm² and a separation distance of 0.5–1.5 cm. The two electrodes were naked without any dielectric barriers, and the electrode unit was housed within a Perspex box fed with a through flow of helium-oxygen mixture. The Perspex box was not airtight, and so oxygen and nitrogen were present. A home-made power source provided a train of unipolar voltage pulses of up to 4 kV at 0.01–20 kHz. Figure 1 shows an example of a homogeneous APGD in an electrode gap of 1 cm and sustained with submicrosecond voltage pulses at 1 kHz. The image in Fig. 1(a) was taken with an exposure time of 1/60 s, whereas the image in Fig. 1(b) was taken in 10 ns with an Andor iCCD system (DH720). These

clearly show a laterally uniform APGD without any streamers. The bottom electrode was the cathode and the bright thin layer near the cathode in Fig. 1(b) was the negative glow. Water cooling was not needed, and the electrode temperature measured with a thermocouple was found to be about 2 °C above room temperature. The generated plasma was stable and repetitive for many hours of continuous operation. These observations suggest that the pulsed barrier-free plasma was essentially a room-temperature, homogeneous, atmospheric glow discharge.

To understand the temporal behaviors of the submicrosecond pulsed APGD, we performed electrical measurements using a Tektronix oscilloscope (TDS 5054B) via wideband voltage and current probes. Figure 2(a) shows the current-voltage characteristics of a barrier-free pulsed APGD in a 0.5 cm electrode gap and with a He–O₂ gas flow [He: 5 SLM (SLM denotes standard liters per minute) and O₂: 5 SCCM (SCCM denotes cubic centimeter per minute at STP)]. The applied voltage was made of a train of unipolar 2 kV pulses repeated at 1 kHz. Each voltage pulse had ap-

FIG. 1. (Color online) Images of a submicrosecond pulsed atmospheric glow discharge in a barrier-free helium gap of 1 cm, taken with exposure times of (a) 1/60 s and (b) 10 ns.

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FIG. 4. (Color online) Nanosecond resolved optical signatures of three excited plasma species together with the current (green) and dissipated power (red) traces.

the same arrangement of an Andor Shamrock spectroscopy system at 1 ms exposure time. Both spectra exhibit spectral signatures of a very similar group of plasma species, for example, the dominant helium line at 706 nm, the nitrogen ion line in 391–393 nm and the nitrogen line at 337 nm, the OH (hydroxide) line at 309 nm, and the atomic oxygen line at 777 nm. Helium and oxygen lines were stronger in the pulsed APGD, and nitrogen lines were stronger in the atmospheric DBD. Oxygen atoms are useful as they are capable of modifying surface properties and inactivating bacteria.^{13–15} Figure 3 indicates that the atomic oxygen line in the pulsed APGD was about 3.7 times of that in the atmospheric DBD. This efficient production of oxygen atoms was found to improve further at larger applied voltages as shown in the insert in Fig. 3(a). As the applied voltage increased to 2.5 kV, the intensity of the 777 nm line became about one magnitude higher in the pulsed APGD than that in the atmospheric DBD. Hence the pulsed barrier-free APGD offers an energy-efficient route to the production of chemically reactive plasma species, such as atomic oxygen. Compared to sinusoidal barrier-free APGD,⁸ pulsed barrier-free APGD is also much more versatile, operated over a wider voltage range and at higher currents.

To further our understanding of the pulsed APGD, optical signatures of three excited plasma species were plotted in Fig. 4 together with the discharge current and dissipated power, all as a function of time on a nanosecond scale. Since the N_2^+ emission in the 391–393 nm range is the most intensive among ionic species in Fig. 3(a), its absence from the first 300 ns in Fig. 4 suggests the absence of significant amount of ions and hence electrons in the electrode gap. This is consistent with the fact that the current pulse became significant only after 300 ns even though the voltage pulse had persisted for the first 300 ns [see Fig. 2(a)]. Emission in 391–393 nm is known to be resulted from $N_2^+(B^2\Sigma_u^+, v_b=0)$ transition to $N_2^+(X^2\Sigma_u^+, v_x=0)$, and the former is populated through the Penning reaction and the charge transfer from He_2^+ ions. The temporal profile of the 391–393 nm line therefore reflects the evolution of the helium metastables and molecular ions.¹⁶ Similarly, the helium line at 706 nm indicates the presence of either energetic electrons or He_2^+ ions

and low energy electrons.¹⁶ Again the absence of a strong 706 nm line before 300 ns supports the suggestion that there were insufficient electrons in the electrode gap to sustain a large current during the first 300 ns of the voltage pulse and that the discharge was at most a Townsend discharge.

It is worth noting that the temporal profiles of all three plasma species are similar. The instants at which the three emission lines reached their peaks were, however, different, and their fall times were also markedly different. For example, raw data suggest that the fall times of the 337 and 706 nm lines were 130 and 150 ns, respectively. The ratio of their intensity integral over time in Fig. 4 was about 2, and compared well with the ratio of their time-accumulated intensity in Fig. 3. Difference in the temporal profiles of the three excited plasma species in Fig. 4 was probably related to their different life spans. It is also worth noting that their temporal profiles fit better with the power profile than the current profile. Given that the applied voltage tailed off during the current pulse in Fig. 2(a), it is possible that the average electron energy decreased in the latter phase of the current phase and the profile of the dissipated power offers a more accurate indication of the dynamics of energetic electrons. This suggests that the profiles of excited plasma species may be influenced by both their lifetimes and the dynamics of energetic electrons.

We have established that submicrosecond voltage pulses can be efficiently used to produce stable and homogenous APGD without dielectric barriers at kilohertz frequencies. Temporal behaviors of such barrier-free APGD were shown to be different from both sinusoidally excited and pulsed atmospheric DBDs. In addition, it has been demonstrated that they are a low-power source of high flux oxygen atoms.

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