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Temporal modulation of plasma species in atmospheric dielectric barrier discharges

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The atmospheric pressure dielectric barrier discharge in helium is a pulsed discharge in nature and the moment of maximum species densities is almost consistent with peak discharge current density. In this paper, a one-dimensional fluid model is used to investigate the temporal structure of plasma species in an atmospheric He-N₂ dielectric barrier discharge (DBD). It is demonstrated that there exist microsecond delays of the moments of the maximum electron and ion densities from the peak of discharge current density. These time delays are caused by a competition between the electron impact and Penning ionizations, modulated by the N₂ level in the plasma-forming gas. Besides, significant electron wall losses lead to the DBD being more positively charged and, with a distinct temporal separation in the peak electron and cation densities, the plasma is characterized with repetitive bursts of net positive charges. The temporal details of ionic and reactive plasma species may provide a new idea for some biological processes. © 2014 AIP Publishing LLC.

I. INTRODUCTION

Dielectric barrier discharges, often known as DBDs, are a common route to nonequilibrium plasmas at elevated gas pressures. At atmospheric pressure, DBDs usually operate in a filamentary mode consisting of a large number of randomly occurring microdischarges (i.e., short-lived streamer filaments). Localized heat deposition of these plasma filaments and their lack of temporal repetitiveness restrict the application scope of atmospheric pressure DBDs, particularly for those that require controlled or even uniform treatment. However, technology advances over the past 20 yr or so have made it possible to realize spatially homogeneous and temporally repetitive DBDs at atmospheric pressure in specific parametric ranges of the gas mixture, the excitation frequency or a pulsed excitation. Homogeneous barrier discharges are of great interest for controlled, reproducible treatment of solid and liquid materials including living tissue and soft matters.

Homogeneous atmospheric pressure DBDs have been investigated both experimentally and numerically, and their current understanding is extensive particularly in gas mixtures dominated by noble gases (e.g., helium and argon). Reaction chemistry, central to almost all plasma applications, tends to be studied by means of its dependence upon time-averaged densities of plasma species and the relative chemical composition of the latter in the ionized gas as the discharge conditions are varied, even though it is known that different plasma species may reach their maximum concentrations at different times. This is appropriate for most existing applications in which differences in temporal profiles of plasma species have little consequence to the treatment efficacy of samples, for example, those with properties insensitive to short-term details of incoming plasma species such as packaging plastics, metallic tools, and contaminated environment. However, new and emerging applications of atmospheric pressure plasmas, such as those involving living systems and nanoscale systems, invite a revisit to the conventional strategy of relying on time-averaged densities of reactive plasma species.

N₂ impurity is known to strongly influence the level of ionization in helium DBDs and, to a lesser extent, the temporal features of the discharge current. For example, Golubovskii and co-worker showed that the impurities of N₂ in helium could influence the symmetry of the discharge current. Yuan and co-workers found that without the presence of N₂ impurities in the plasma chemistry model, the multipulse phenomena at lower frequencies cannot be simulated accurately since the pure helium chemistry model results in single broad pulse at the lower frequencies. Martens and co-workers have demonstrated how nitrogen may strongly alter the ionic composition of atmospheric pressure He plasma and indeed the temporal features of the discharge current. Urabe and co-workers found that the ionization frequency is maximized under the 0.25% condition of the N₂ impurity ratio. Of particular note is a recent report of an atmospheric He-N₂ DBD with N₂ fixed at 100 ppm, in which a delay of a few μs in the N₄⁺ concentration from other species was observed. Recently, there have been also several experimental...
works exploring the time-evolution of ionic species in an atmospheric pressure helium discharge \(^\text{20-22}\) and similar phenomenon is found in these studies. This observation was not explained in terms of plasma physics nor explored for its existence for other plasma species. The perhaps of greater interest is whether the phenomenon is a detail of DBD physics with no practical relevance or whether it may broaden how DBDs are used to treat biotic materials as well as smart materials, \(^\text{23}\) particularly those strongly sensitive to short-term temporal details of external stimuli. This observation was not explained in terms of plasma physics nor explored for its existence. A phenomenon is found in these studies. This observation was not explained in terms of plasma physics nor explored for its existence. However, it is worth noting that the abovementioned microsecond delay of the peak \(N_3^+\) density falls within the nanosecond-millisecond timescale of some biomolecular and cellular processes such as ligand binding, protein folding, and enzymatic catalysis. \(^\text{24-26}\)

Our study first investigates temporal characters of plasma particle concentrations of He-N\(_2\) DBDs and their modulation by the nitrogen admixture in terms of time delays in the moments of the maximum concentrations of charged and neutral plasma species from the instant of the peak discharge current density by means of a fluid modelling. Following a short introduction to the numerical method and details of relevant chemical reactions in Sec. II, the results of the study are presented and discussed in Sec. III for a wide N\(_2\) content from 0.1 to 10\,000 ppm so as to bring out a full scope of temporal modulation of plasma chemistry. Finally, our findings are summarised in Sec. IV.

II. COMPUTATIONAL MODEL

The present study considers an atmospheric pressure He-N\(_2\) DBD sustained at nominally 10\,kHz and a peak applied voltage of 1\,kV between two dielectrically insulated electrodes. The relative permittivity of the dielectric insulation layers is 9 and the thickness of each layer is 1 mm. The gas gap is fixed at 2 mm and the gas temperature at 300 K. The amount of nitrogen in the plasma-forming gas varies from 0.1 to 10\,000 ppm. A one-dimensional fluid model is employed here, following its successful simulation of atmospheric pressure helium plasmas. \(^\text{27-29}\) Details of the model are given in the literature, however a brief summary of its key features is provided here. The very high particle collision frequency at the atmospheric pressure allows the usage of drift and diffusion approximation. The governing equations of the fluid model are given below

\[
\frac{\partial n_i}{\partial t} + \nabla \cdot \Gamma_i = S_i, \quad \Gamma_i = \text{sgn}(q_i)n_i\mu_iE - D_i\nabla n_i, \quad (1)
\]

where \(n_i, \Gamma_i, S_i, q_i, \mu_i, \) and \(D_i\) represent, respectively, the particle density, flux, net generation rate, charge, mobility, and diffusion coefficients for plasma species \(i\). \(E\) is the electric field and \(\varepsilon = 1.5k_B T_e\) is the mean electron energy. \(\varepsilon_0\) is the vacuum permittivity, \(e\) is the elementary charge, and \(k_B\) is the Boltzmann constant. \(R_{ce}\) is the momentum transfer collisional rate between electrons and helium, \(T_e\) is the temperature of electron and \(T_{He}\) is the temperature of helium. \(\Delta E_i\) and \(R_i\) are the electron energy loss due to inelastic collision \(j\) and its corresponding reaction rate. Subscript \(e\) represents electron. Equations (1)–(3) are solved by using a time-dependent finite-element partial differential equation solver, COMSOL Multiphysics, and simulation results are post-processed with MATLAB. \(^\text{27-29}\)

For charged particle fluxes to the dielectrics, the following boundary conditions are used:

\[
\Gamma_e \cdot n = -\alpha_\text{e}\mu_\text{e}\cdot n\mu_\text{e} + 0.25v_{th,e}n_e - \gamma \sum_p \Gamma_{p,e}, \quad (4)
\]

\[
\Gamma_\pm \cdot n = \alpha_\pm\mu_\pm\cdot n\mu_\pm + 0.25v_{th,\pm}n_\pm, \quad (5)
\]

where \(\mathbf{n}\) is the normal vector pointing towards the dielectric, \(v_{th}\) is the thermal velocity and \(\gamma\) is the secondary emission coefficient. \(\gamma\) is set at 0.05 for helium ions and 0.001 for nitrogen ions. \(^\text{19}\) The switching function \(\alpha\) takes the value of 1 when the drift velocity is directed towards the electrodes and zero, otherwise

\[
\alpha = \begin{cases} 
1, & \text{sgn}(q_i)\mu_iE \cdot n > 0 \\
0, & \text{sgn}(q_i)\mu_iE \cdot n \leq 0.
\end{cases} \quad (6)
\]

For neutral species, the boundary conditions are

\[
\Gamma_i \cdot n = 0.25v_{th,n_i}, \quad (7)
\]

For simplicity, the mean electron energy at both gas-facing borders of the electrodes is fixed at 1\,eV. For the Poisson equation, one electrode is grounded and a sinusoidal voltage is applied to the other electrode. The gas-dielectric interface has the following boundary condition:

\[
\varepsilon_\text{dielectric}\cdot E \cdot n - \varepsilon_0 E \cdot n = \sigma, \quad (8)
\]

where \(\varepsilon_\text{dielectric}\) is the permittivity of dielectric and \(\sigma\) is the surface charge density on the dielectric. Similar to previous study, \(^\text{16}\) the model comprises 9 species (He, e, He\(^+\), He\(_2^+\), He\(_3^+\), N\(_2\), N\(_2^+\), and N\(_4^+\)) and 18 chemical reactions. The ions N\(_2^+\) and N\(_3^+\) and the neutral species N, N\(_a\), and N\(_{2a}\) are not taken into account for matters of simplicity. N\(_2^+\) and N\(_3^+\) are very quickly converted into N\(_2\)\(_2^+\) through N\(_2^+\) + N + He → N\(_2\)\(_2^+\) + He and N\(_3^+\) + N + N → N\(_2\)\(_2^+\) + N\(_2^+\) \(^\text{16}\) respectively. N\(_a\) and N\(_{2a}\) are of minor importance in the charged species composition of He-N\(_2\) discharge, as the N\(_2\) impurity level is very low in this study. The chemical reactions used in the model are obtained from most of the publications on He-N\(_2\) discharge. Overall, the species and chemical reactions considered in this paper are sufficient to provide an accurate representation of the He-N\(_2\) discharge chemistry based on the previous studies. \(^\text{14-17}\) The reaction rate coefficients for electron impact reactions as well as the transport coefficients of electron were calculated using an offline Boltzmann solver.
The remaining reaction rate coefficients were obtained from Ref. 16. The transport coefficients for other species are obtained from the literature.

The transport coefficients for other species are obtained from the literature.27,32

III. RESULTS AND DISCUSSIONS

A. Time delays

For all results represented here, the He-N2 DBD is operated at sufficiently low current densities such that there is one discharge event every half cycle of the applied voltage. Simulation results suggest that the pulse shape of the discharge current density remains little changed and its pulselength is less than 1 μs (more specifically Δt = 0.25–0.36 μs for [N2] = 10–100 ppm and Δt = 0.99 μs for [N2] = 1000 ppm, data not shown). The narrowness of the current pulse allows for the instant of the peak current density to be used as the reference point to which time delays of the maximum particle concentrations are calculated and considered.

Fig. 1 illustrates the space-averaged electron density as a function of time at four N2 content levels of [N2] = 10, 30, 100, and 1000 ppm. The zero point of the horizontal axis represents the moment of the maximum discharge current density. At 10 ppm, the electron density reaches its maximum value close to t = 0 and its curve has a singular peak hereafter known as PV1. This is consistent with the results of numerical studies reported in literature.33 Increase in the N2 content level leads to a reduction in PV1 and a rise of the tail of the electron density curve. At 30 ppm, the electron density is seen to possess two peaks with the first being narrow and the second being broad in pulse shape. The second peak, hereafter referred to as PV2, has the same magnitude as PV1 and lags the peak current density by Δt = 5.8 μs, almost 20 times as large as the pulse width of the current density at 30 ppm. As the nitrogen level is increased further, PV2 becomes increasingly dominant with concurrent reduction in the magnitude of PV1. Above 100 ppm as seen in the 1000 ppm case, the first peak is no longer distinguishable. The existence of two electron density peaks has not been reported before nor has the considerable time delay of the second electron density peak. Interestingly, such temporal features can be modulated by the N2 content.

Most plasma species in the discharge plasma are directly or indirectly linked to electrons. Therefore, the time delay of the maximum electron density from the peak current density should be reflected in the concentrations of neutral and other charged plasma species. The time delays of space-averaged concentrations of charged particles and helium metastables from the instant of the maximum current density are shown in Fig. 2 as a function of the N2 content. It is evident that the time lags for nitrogen cations and electrons are significant, with the maximum delay at, respectively, ΔN2+ = 12 μs, ΔN4+ = 11.1 μs, and Δe = 5.8 μs. Consistent with previous reports,16 our simulation shows that, when compared in space- and time-averaged concentrations, the nitrogen cations become dominant when the N2 level is above 1 ppm (data not shown). For [N2] = 1–30 ppm, N2+ ions are found to dominate, with a time lag of 1.9–12 μs from the peak current density. Xiong et al.20 also found that the peak moment of excited N2+ (B, v′ = 0) by optical emission spectroscopy is delayed by about tens of ns compared with that of discharge current in a helium plasma jet. It is apparent that the time delay of excited N2+ (B, v′ = 0) is much smaller than that of N2+ in this paper. While Oh et al.21 and McKay et al.22 found that the time delays for N2+ and O2+ are a few μs by a mass/energy analyzer HPR60. On the other hand, the electron density has a negligible time delay for 0–30 ppm. So the electrons reach their maximum space-averaged concentration earlier than the cations and the duration of 1.9–12 μs, for 1–30 ppm, represents a timescale when the electrical neutrality of the plasma may not be maintained.

Above 30 ppm in N2 impurity, N4+ becomes the dominant cations when compared in terms of space- and time-averaged concentrations (data not shown). The delay of the instant of its maximum concentration is seen to decrease monotonically from ΔN4+ = 8 μs at [N2] = 30 ppm. The instant of the maximum electron density is seen in Fig. 2 to jump to Δe = 5.8 μs at [N2] = 30 ppm, and then to undergo a similar monotonic decay to the N4+ density. However, the

FIG. 1. Space-averaged electron density as a function of time at different N2 levels.

FIG. 2. Time lags of the peak space-averaged particle densities from the peak current density as a function of the N2 content.
N$_2^+$ density has a greater decay rate between 30 and 300 ppm, and its time delay becomes similar to that of electrons after [N$_2$] = 300 ppm. In other words, the relative time delay between electron and cation concentrations may disappear around 300 ppm. The above discussion suggests that the nitrogen content in the plasma-forming gas may be adjusted to tune the temporal profile of the net-charge density and hence the electrical neutrality of the DBD. It is worth mentioning also that helium ions have a time delay much less than 0.3 $\mu$s (not shown in Fig. 2).

The point of [N$_2$] = 30 ppm appears to be a separation of two regions. In the first region (indicated by “I” in Fig. 2) below 30 ppm, the time delay for electrons is no more than 0.1 $\mu$s and almost constant. Above 30 ppm in the second region (indicated by “II” in Fig. 2), the time delay of the instant of the peak electron density has a step increase to $\Delta t_e = 5.8$ $\mu$s, much greater than the current density pulse of 0.30–0.36 $\mu$s for [N$_2$] = 30–100 ppm. The monotonic decay with [N$_2$] in the instant of the peak electron density after 30 ppm is also seen in the trends for He$_2^*$, N$_2^+$, and N$_4^+$ species. For [N$_2$] above 300 ppm, the time difference for N$_2^+$ and He$_2^*$ is not more than 0.3 $\mu$s. Above 5000 ppm, the time delays for all species are not more than 0.3 $\mu$s. To gain insights into possible mechanisms responsible for the time delays, space-averaged electron generation, and loss rates are shown in Fig. 3 for three N$_2$ content levels at 10, 100, and 1000 ppm. Identical to that in Fig. 1, the time zero point is the moment of the maximum current density and the vertical dashed line corresponds to the moment of the maximum space-averaged electron density. The electron density reaches its maximum when the electron loss rate exceeds the electron generation rate. The main route for electron loss is the wall loss at the electrodes, and their recombination with ions is comparably negligible. In other words, the electron loss remains relatively independent of the nitrogen content in the plasma-forming gas. By contrast, electron generation is dominated by the electron impact ionization and the Penning ionization with the relative importance of the latter determined by the N$_2$ level. Penning ionization includes self-Penning ionization of helium metastables and ionization of N$_2$ by helium metastables. Our simulation indicates that for [N$_2$] > 10 ppm the rate of self-Penning ionization is negligible compared to that of Penning ionization of nitrogen molecules. As our discussion of Figs. 1 and 2 is largely for 10–10000 ppm, self-Penning ionization will be ignored hereafter and the term of Penning ionization used specifically for the ionization of N$_2$ by He metastables.

Fig. 3 shows that the self-Penning ionization has a very low rate for all nitrogen contents considered. It is seen in Fig. 3(a) that the electron impact ionization is dominant at [N$_2$] = 10 ppm that is too small for the Penning ionization to significantly contribute to electron generation. Shortly after $t = 0$, the rate of electron impact ionization starts to decline and the electron loss rate exceeds its generation rate at point PV1* (or $t = 0.03$ $\mu$s), resulting in the maximum space-averaged electron density PV1 in Fig. 1.

With N$_2$ content increased to 100 ppm, the Penning ionization of N$_2$ becomes stronger and its duration becomes longer (data not shown), leading to an increasingly more significant contribution. These combine to result in the second peak of the electron density (i.e., PV2) in Fig. 1. At [N$_2$] = 100 ppm in region II (see Fig. 2), Fig. 3(b) shows that the electron generation rate due to the Penning ionization of N$_2$ is comparable to that of the electron impact ionization for $t < 0.2$ $\mu$s but becomes dominant after $t = 0.2$ $\mu$s. As both helium metastables and nitrogen molecules persist for a long period of time, Penning ionization last for a long period of time, leading to a very large time delay for the peak electron density at 100 ppm. Further increase of N$_2$ content to 1000 ppm in region II, the Penning ionization is so strong that PV1* is no longer present in Fig. 3(b) and so the electron density has no PV1 above 100 ppm in Fig. 3. It is worth noting that the Penning ionization is dominant at
In both regions with $[N_2]$ spanning for 10–10,000 ppm, the standpoint of He metastables and their production and loss. He$^*_2$ for $[N_2]$ shows little dependence on $N_2$ impurity (data not shown). The little time delay of He$^*$ and the enhanced reduction of He$^*_2$. Therefore, time delay for He$^*_2$ is negligible as shown in Fig. 2.

It is worth supporting the above discussion from the standpoint of He metastables and their production and loss. In both regions with $[N_2]$ spanning for 10–10,000 ppm, $e + He \rightarrow e + He^*$ is responsible for the generation of He$^*$ and this reaction mainly occurs during the gas breakdown process. Time delay for He$^*$ from the instant of the peak current density is negligible for all $N_2$ levels. For $[N_2] < 1$ ppm, the Penning ionization is too weak to contribute to any sizeable reduction of He$^*_2$. Therefore, time delay for He$^*_2$ shows little dependence on $N_2$ impurity (data not shown). With increasing $[N_2]$, Penning ionization becomes progressively dominant in the reduction of He$^*_2$, leading to an increasingly early onset of its peak concentration. In general, the time delay for He$^*_2$ is negligible for $[N_2] \geq 100$ ppm. The little time delay of He$^*$ and the enhanced reduction of He$^*_2$ for $[N_2] \geq 100$ ppm contribute to the reduced time delay of the peak electron density above 100 ppm (see Figs. 1 and 3).

B. Ionic burst and tail

It is now clear that the competition between the electron impact and the Penning ionizations results in a temporal concentration imbalance of electrons and ions with a microsecond delay in the He–$N_2$ DBD. It is conceivable that this will be observable also in atmospheric pressure DBD in other gas mixtures, since production of many reactive plasma species is closely related to the electron density and the electron energy.29 To see this, Fig. 4 shows space-averaged densities of electrons and ions for $[N_2] = 30$ ppm. Helium ions and electrons are seen to reach their peak densities at a delayed time similar to the time delay of the peak electron density above 100 ppm (see Figs. 1 and 3).

![Fig. 4. Space-averaged densities of electrons ($\bigtriangledown^-$), He$^+_2$ ($\square$), N$^+_{2}$ ($\triangle$) and N$^+_{2}$ ($\bigcirc$) as a function of time at $[N_2] = 30$ ppm.](image)

 neutrality is broken down right from gas breakdown and well before the nitrogen ions become more abundant than the helium ions. This leads to a train of cation bursts, once every half cycle of the voltage excitation. Breakdown of electrical neutrality is known to be possible in atmospheric pressure plasmas.28,34

Fig. 5 shows the temporal profiles of space-averaged electron, ion, and net charge densities. It is evident that the net charge is always positive and so charges in the plasma are characterized by a narrow net positive-charge burst followed by a tail also of net positive charges. The early surge of He$^+_2$ ions appears to cause the breakdown of the electrical neutrality already and the late surge of nitrogen ions with a larger time- and space-averaged density seems to play a secondary role of maintaining the charge imbalance until the next gas discharge event. Yet nitrogen ions are directly related to production of reactive plasma species and so their role goes beyond contributing to electrical neutrality. For example, important reactive oxygen and nitrogen species such as O, O$_2^-$, and O$_3$ may be produced from N$_2^+$ + O$^-$ → O + N + N, N$_2^+$ + O$_3^-$ → O$_3$ + N + N, and N$_2^+$ + NO$^-$ → NO + N$_2$. This suggests that reactive plasma species may reach their peak densities at a delayed time similar to the moment of the peak nitrogen ion density when O$_2$ admixture is included in the plasma-forming gas. This is supported by a recent study of radio-frequency plasma in atmospheric He–O$_2$ mixture in which electron fluxes are shown to reach an inserted sample before ions.29,30 Two characters of the He–$N_2$ DBD are therefore emerging—first a sequence of net positive charge bursts during gas breakdown and second an abundant production of cations and reactive plasma species delayed by a microsecond scale.

Temporal details of ionic and reactive plasma species may impact on some biological processes. For instance, the membrane channel conductance of neuronal cells is known to depend upon how ions are accumulated and released from the cell membrane.35 Many cell functions including the cell-cell communication and nerve growth are induced by the initiation of the action potential and hence the latter is a key trigger. The axon hillock of a neuron cell has a resting
voltage of about $-70 \text{ mV}$ and the cell membrane is therefore normally polarized. The negative sign of the membrane voltage indicates that the inside of the membrane is more negatively charged. To initiate the action potential, the membrane needs to be depolarized by increasing the membrane voltage to around $-55 \text{ mV}$. This is done usually by injecting extra sodium cations into the cell via several biological mechanisms. Therefore, it is possible to trigger the action potential of neuron cells using the ionic burst of He-N$_2$ DBD and provide a new idea for some biological processes. Of course, a great deal of work, both in physics and biology, needs to be undertaken before the feasibility is translated into practical therapies.

**IV. CONCLUSIONS**

A dielectric barrier discharge (DBD) in He-N$_2$ mixture is studied using a fluid model for temporal characteristics of its chemical species with respect to those of its electric current. It is shown that there exist time delays between the moments of the maximum discharge current density and the maximum space-averaged particle density. The time delays are different with different species, and they change as the volume percentage of N$_2$ increases. Over the N$_2$ range up to 10 000 ppm, the time delays for electron, N$_2^+$, N$_4^+$, and He$_2^*$ are found to depend very strongly on the N$_2$ content largely due to the competition between the electron impact and Penning ionizations. By contrast, the time delay of the peak He$_2^*$ density to the peak discharge current varies little with increasing N$_2$ content. Significant electron wall losses lead to the DBD being more positively charged and, with a distinct temporal separation in the peak electron and cation densities, the plasma is characterized with repetitive bursts of net positive charges. The distinct temporal separation in the production of electrons and ionic species stems from different onset times of the competing electron-impact and Penning ionizations, and it is conceivable that a competition of similar or other ionization mechanisms in other plasma systems may lead to pulsed and temporally separated production of charged particles too. The temporal details of ionic and reactive plasma species may provide a new idea for some biological processes.

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