Numerical analysis of coaxial dielectric barrier helium discharges: three-stage mode transitions and internal bullet propagation

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Dielectric barrier discharge (DBD) has recently attracted great interest for the generation of low-temperature (cold) atmospheric pressure plasma which could be applied in various fields, such as surface treatment,1 medicine,2–4 air-pollution control,5 and material synthesis with assimilation of carbon dioxide.6,7) DBD devices are composed of a planar or cylindrical dielectric and corresponding shaped electrode(s). The planar type generates plasma on and above the dielectric surface. The plasma jet type blows out a discharge. In the reactor type, discharge stays inside the device. All the device types can feed a chemically reactive species to the downstream region of the device.

In planar type DBD, discharge mode changes from streamer to glow depending on the applied voltage polarity.8) Control of the surface charge can align direction of movement of charged species in plasma collision with background gas and enhancing ionic winds.9) One-dimensional numerical simulations reported that parallel plate helium glow discharges were basically similar to low-pressure glow discharge in 100 kHz10) and 13.56 MHz11) and some mode changes occur with saw-tooth voltage.12)

Experimentally, it has been revealed that the plasma jets are a series of rapidly propagating (∼100 km s−1) luminous streamer discharges called plasma bullets13) and that the bullets are ring-shaped.14) The jet type discharges involve two streamer structures depending on the jet width15) and there are three distinct modes with increasing input power.16) The propagating bullets are guided by a He-air mixing layer with background atmospheric air.17) Photoionization determines the streamer propagation speed, but is not the reason for the propagation itself.18)

Discharges in reactor type DBD have been reached a consensus of glow-like in pure He and filament-like in pure Ar.19) As mixing NH3 and Ar, a transition of discharges from filament-like to glow-like was observed and this transition was explained by the Penning effect.20) The helium discharge appears nearly uniform, whereas strong emissions are observed in the region near the electrodes and dielectric surface.21) There are comprehensive parametric studies with respect to applied voltage and dielectric properties by one-dimensional numerical simulation.22,23) However, the nanosecond (ns)-scale dynamics of the reactor type discharges have not been elucidated yet and any reason for strong light emission near the electrodes remains unclear. One-dimensional analysis that assumes a uniform axial distribution is substantially limited in terms of revealing any details of the formation process of plasma inside the reactor type DBD. Therefore, multidimensional analysis is required to understand detailed plasma generation process in reactor type DBD.

This letter attempts to clarify the detailed plasma formation process in coaxial reactor type DBD on the nanosecond to microsecond timescale using numerical analysis. We will show that plasma generation process in the reactor type DBD is quite different from the planar DBD and also show that an insight of control of the plasma bullet. One of the reasons why multidimensional analysis of the entire device has not been performed so far is that the timescales of discharge phenomena and the cycle of applied voltage are significantly different. In particular, in order to analyze the plasma behavior in recently developed device driven by commercial power frequency (50 or 60 Hz)24) throughout an entire cycle, more than 1 × 109 iterations are required due to limitations in the numerical time step (∼1 × 10−12 s).

First of all, we have focused on the beginning phase of single discharge pulse. The voltage of alternating current (AC) with an amplitude of 10 kV under typical discharge duration (about 2 μs) changes sufficiently small 6 V (0.06% for 10 kV). This is valid that the applied voltage is regarded as square-pulse waveform of the discharged voltage. Although charge accumulation on the dielectric surface prior to the discharge pulse can affect under AC voltage waveform, the simulation of a single discharge pulse provides information on the complicated dynamics of mode transitions during a discharge.

A self-consistent, multi-species, multi-temperature plasma fluid model was used to analyze the formation process of plasma inside reactor type DBD. The model is composed of continuity equations for each species, an electron energy...
conservation equation, bulk (gas and dielectric) energy equation, and Poisson’s equation for self-consistent electric fields. The drift-diffusion model is used to calculate flux terms for all species, as described previously in detail.\textsuperscript{17,26} The coupled set of nonlinear governing equations was solved by a commercial plasma solver package.\textsuperscript{27} The chemistry model comprises six species of electron, helium ions, and metastables (E, He\textsuperscript{+}, He\textsubscript{2}\textsuperscript{+}, He\textsubscript{m}, He\textsubscript{2}\textsubscript{m}, and He). The reaction pathways and their reaction rate coefficients for non-electron impact reactions are obtained from a previous work.\textsuperscript{11} The rate coefficients for electron impact reactions are calculated using Boltzmann solver Bolsig+.\textsuperscript{28} Photoionization in discharges accompanied by streamers (bullets) has been noted to determine the streamer propagation speed, but is not essential for propagation itself.\textsuperscript{18,29} A recent model includes photoionization due to surrounding O\textsubscript{2} molecules photo-ionized by radiation arising from de-excitation of excited N\textsubscript{2} molecules.\textsuperscript{30} No model involves photoionization for noble gases. In our previous study without a photoionization model, the propagation speed of the argon streamer agreed well to within an order of magnitude with a number of experimental results (\textasciitilde mm/ns).\textsuperscript{26} Therefore, this does not assume any photoionization processes.

Figure 1 shows a schematic of the reactor type DBD device used in this study. Pure helium flows through a dielectric tube (with a relative dielectric constant of 10) of 4 mm inner diameter, 0.8 mm thickness, and 100 mm length. For ease of viewing, the z direction is displayed at 1/5 scale. Two copper electrodes are placed around the dielectric tube and voltage is applied to the upper electrode while the lower electrode is earthed. Commercial power frequency (50 or 60 Hz) is assumed for the applied voltage and modeled as 15 kV constant voltage. The computational region and computational mesh are also shown in Fig. 1. Simulations are conducted in two-dimensional axisymmetric geometry. The entire computational mesh is a constructed mesh and the number of cells is 20 000 cells. This resolution is adequate for capturing qualitative discharge behavior. This configuration reproduces plasma jets, but the bullets do not jump out from the dielectric tube because the distance between the electrodes and exit is longer than general plasma jets (up to 20 mm). In the previous study,\textsuperscript{13} no bullets were produced by a similar configuration. The plasma discharge stays inside the tube. Therefore, the computational domain is set inside the device including the tube.

Notably, the gas flow is not considered in the simulation because of the highly disparate timescales of the plasma discharge phenomena and the fluid flow in typical conditions of several slm (standard litter per minute) gas flow rate in reactor type DBD. To model propagation of the bullets outside the tube, the mole fractions of helium and air outside the tube are estimated as the background concentration.\textsuperscript{11,14} Initial and boundary condition for the gas and dielectric temperature is set at 300 K. Temperature rise induced by the discharge under several microseconds was less than 0.1 K and had negligible effects on the discharge dynamics.

When a voltage is applied, a high electric field is induced between the electrodes and electron acceleration (heating) results in an increase in the electron temperature, and the electron impact reactions generate chemically reacted species. As shown in Fig. 2, the plasma in the device dynamically changes on the nanosecond timescale. Three discharge modes are observed. The distributions of reduced electric field and electron number density in the gas region are shown. Results are described together with the time history of the electron number density and electron temperature at the center between the electrodes and near the dielectric surface above the lower electrode, as shown in Fig. 3.

Firstly, an electric field of about 30 Td is induced between electrodes by the applied voltage, and an increase in the electron number density is observed (first stage). This discharge mode is the Townsend-glow type, which is nearly uniform between the electrodes, and the electron temperature is almost constant at 40 000 K. Previous studies report that

![Fig. 1.](image-url)
helium DBD tends to show glow-like luminance.\textsuperscript{19,21} Our results qualitatively agree these reports because the Townsend and glow discharge shows similar near-uniform light emission.

When the electron number density reaches about $1 \times 10^{18}$ m$^{-3}$ (150 ns), electric field wavefronts form at the edge of the plasma region, and the discharge mode transitions to the axially-propagating streamer (bullet) type (second stage). As the discharge passes, the electron number density further increases to $1 \times 10^{19}$ m$^{-3}$. The electron temperature decreases at the center between the electrodes in this stage, because this position is behind the streamer head and the electric field decreases. This high electric field region corresponds to the ionization wavefront or streamer head, and this is driven by a local electric field induced by a local space charge from the density difference between positive and negative charged particles. The wavefront starts to propagate when the local space charge reaches $1 \times 10^{19}$ m$^{-3}$, which is the same order of magnitude as bulk plasma density. This streamer type discharge spreads in the radial direction while propagating in the axial direction, and when it reaches the vicinity of the electrode, the propagation stops and the edge reaches the dielectric surface. After that, a gradual change is observed between the electrodes.

No increase in electron density near the dielectric surface between the first and second stages can be seen from Figs. 3(a)–3(c). In the third stage, the discharge mode further changes to a surface discharge that propagates near the dielectric surface. The surface discharge increases the electron number density near the dielectric surface as it propagates, and stops when it spreads to a slightly wider range than the electrodes. As this surface discharge passes, the accumulated charge density on the dielectric surface increases to about $9 \times 10^{-4}$ C m$^{-2}$, and the reduced electric field instantaneously increases to about 350 Td. The electron heating induced by this strong electric field causes the electron number density near the dielectric to increase to $1 \times 10^{20}$ m$^{-3}$, which is higher than that between the electrodes.

Through these three stages that occur within approximately 270 ns, a distribution with high electron number density is obtained in the DBD device between the electrodes and near the dielectric on the electrodes. Secondary ionization by He$^*$ and He$_2^*$ continues for longer than 10 $\mu$s. The fact that the inside of the tube is basically Townsend-glow-like and the plasma density is high near the dielectric where the electrodes are arranged qualitatively corresponds to the visualization results so far.\textsuperscript{19,20} The strong light emission in the vicinity of the electrode is confirmed to be due to surface discharge. Plasma in reactor type is formed by different discharge modes at the center between the electrodes and near the dielectric surface. This plasma generation process is quite different from that of planar DBD. In planar type DBD, one mode basically corresponds to one discharge pulse. On the other hand, three discharge modes are confirmed to occur in one pulse in the reactor type DBD. We have performed additional parametric studies (not shown) about important parameters. The basic distributions and the three modes do not change by the applied voltage, gap between the electrodes, dielectric tube properties (thickness and dielectric constant), and inner diameter of the dielectric tube.

Whether the streamer shoots out or stays inside can be determined in the second streamer formation step. In the plasma jet type, discharge becomes a bullet while passing through the surface discharge inside the tube when the voltage is applied and propagates to the outside of the tube. In the reactor type, discharge starts from a relatively gentle Townsend-glow-like discharge between the electrodes and settles to a surface discharge near the electrodes. Unlike the
Fig. 3. (Color online) Time history of electron number density (solid line) and electron temperature (dashed line). (a) Long timescale (all stages), (b) first stage, (c) second stage, and (d) third stage. Blue represents values at the center between the electrodes and orange represents values near the dielectric surface on the lower (earthed) electrode.
plasma jet, no bullets shoot out of the tube, and instead the bullet is actually formed between the Townsend-glow-like and surface discharges and travels a short distance of less than 6 mm. Our parametric study (not shown) confirmed that the bullet did not shoot out in all the cases, although the size and propagation properties changed with conditions. The strong electric field of about 350 Td induced by a combination of the ground electrode and the charge accumulation, and the dielectric surface promotes the transition to the surface discharge, and the distribution spreads slightly beyond the electrodes. These results indicate that arrangements of electrodes (shape of electrodes and position of 0 V electrode) and distance between the electrodes and the tube exit is important to shoot out the bullet. In fact, the distance is less than 2 cm and 0 V is set as target or surrounding air (virtual ground) in typical plasma jet devices.

In summary, we analyzed the formation process of helium plasma in the reactor type DBD using a plasma fluid model. Results indicate that plasma forms in the reactor type DBD through three discharge modes. Firstly, a Townsend-glow-like discharge is generated between the electrodes, and when the space charge due to the difference in positive and negative charged species densities reaches the same level as that of the bulk plasma, an electric field wavefront is formed and a streamer- (bullet-) like discharge develops in the axial direction. Finally, when the electric wavefront reaches the dielectric surface, it transitions to a surface discharge emphasized by the accumulated charge, and the discharge propagates to the full electrodes. At this time, the surface charge density is about $9 \times 10^{-4}$ C m$^{-2}$. Compared with $1 \times 10^{19}$ m$^{-3}$ between the electrodes, the electron number density near the electrodes is as high as $1 \times 10^{20}$ m$^{-3}$, which confirms that the strong emission in this region is due to the surface discharge. In the reactor type DBD, the development of the bullet is confirmed as in the case of the plasma jet, but it is trapped by the strong electric field (momentary about 350 Td) due to the electric field induced by a combination of the ground electrode and the accumulated charge, and the propagation distance is short at 6 mm and it immediately transitions to a surface discharge. This suggests the possibility of controlling the plasma bullet not only by the distance between the electrode and the tube outlet, but also by the electrode structure (arrangement).

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