Numerical simulations of stable, high-electron-density atmospheric pressure argon plasma under pin-to-plane electrode geometry: effects of applied voltage polarity

Yosuke Sato\textsuperscript{1,2}, Kenji Ishikawa\textsuperscript{2}, Takayoshi Tsutsumi\textsuperscript{2}, Akio Ui\textsuperscript{1}, Masato Akita\textsuperscript{1}, Shotaro Oka\textsuperscript{1} and Masaru Hori\textsuperscript{2}

\textsuperscript{1} Mechanical Systems Laboratory, Corporate Research and Development Center, Toshiba Corporation, 1 Komukai-Toshiba-Cho, Saiwai-ku, Kawasaki 212-8582, Japan
\textsuperscript{2} Nagoya University, Furo-cho, Chikusa, Nagoya, Aichi 464-8601, Japan

E-mail: yosuke7.sato@toshiba.co.jp

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Abstract

When applying high-voltage direct current to a pin-to-plane electrode geometry with a distance of 2 mm under atmospheric pressure in argon gas, electrical breakdown forms primary then secondary streamers. The polarity of the applied voltage affects this streamer-propagating phenomenon. Properties such as propagation speed, streamer head size, and plasma generation are parameterized at nanosecond scales by computational simulations of a self-consistent, multi-species, multi-temperature plasma fluid modeling approach. For positive polarity on the pin electrode, streamer-head propagation speeds up and streamer head size increases with increasing applied voltages. However, local electron density at the head decreases. For negative polarity, corona-like discharges form around the pin electrode under low applied voltages, and diffusive steamers form under high applied voltages. Secondary streamers re-propagate from the pin after primary streamer propagation, forming a plasma with a high electron density of \(10^{21} \text{ m}^{-3}\) for the positive polarity. We show that low-voltage operations with positive polarity are useful for stable high-electron-density discharges under atmospheric pressure argon.

Keywords: atmospheric pressure plasma, plasma simulation, argon streamer

(Some figures may appear in colour only in the online journal)

1. Introduction

Atmospheric pressure plasma has been utilized for a variety of applications, such as surface treatments [1, 2], gas decomposition [3, 4], ozone generation [5], and medicine [6, 7].Streamer discharge is fundamental for atmospheric pressure plasma phenomena, forming a propagation of filamentary ionization waves called a streamer head. This propagation can deliver a rich variety of reactive species, such as O atoms, N atoms, \(\cdot\)OH radicals, and \(\cdot\)NO radicals. However, such phenomena are incompletely understood, so we focus here on computational approaches toward clarification of spatiotemporal behaviors of streamer discharges, that occur in the order of nanoseconds.

To date, atmospheric pressure streamer discharges in air-related gases involving \(\text{N}_2\), \(\text{O}_2\), \(\text{N}_2/\text{O}_2\), and \(\text{N}_2/\text{O}_2/\text{H}_2\text{O}\) mixtures have been studied experimentally [7–14], analytically, and computationally [15–21]. Numerical simulation
studies have discussed the propagation of primary and secondary streamers mainly in air-related gases, causing generation of O, N, and OH radicals. There are few studies on streamer discharge in noble gases at atmospheric pressure, and while there are previous studies of positive streamers, which are generated by applying positive voltages, few consider negative streamers. In addition to low-pressure discharges, an understanding of noble gas discharges remains an important issue for providing broad knowledge over a wide range of atmospheric pressure plasma applications.

In this study, we investigated the fundamentals of atmospheric pressure discharges in noble gases, taking computational approaches to evaluate key parameters for propagation speeds and streamer head shapes. Plasma generation in atmospheric pressure argon discharges has been compared in cases of applied voltage polarity in a pin-to-plane electrode geometry. We found that a negative polarity of applied voltage to the pin electrode provided characteristic applied voltage in section 5. Prepared in cases of applied voltage polarity in a pin-to-plane atmospheric pressure discharge has been compared in cases of applied voltage polarity in section 4. The timescale associated with streamer propagation \( t_s \), which is generated by applying positive voltages, is too short for use as a gas dynamic response timescale. Therefore, the background temperature \( T_b \), where the mean free path of ions and neutral species (on the order of a micrometer) are much less than the characteristic scales of the geometry (on the order of a millimeter), and is widely used in studies of atmospheric pressure discharge.

2. Model description

We use a self-consistent, multi-species, multi-temperature plasma model, utilizing a commercial plasma solver to solve a coupled set of nonlinear governing equations \([22]\). A similar model is used in \([23]\) and \([24]\), so the model and governing equations are briefly described below.

2.1. Species continuity

The number densities of a species are obtained by solving the separate species continuity equations

\[
\frac{\partial n_k}{\partial t} + \nabla \cdot \Gamma_k = G_k, k = 1, \ldots, K_{\text{gas}} (k \neq k_d)
\]

for all charged and neutral gas species given by index \( k \), with the exception of a single dominant species given by index \( k_d \) (assumed to be ground-state neutral argon). In the above equation, \( n_k \) is the number density, \( \Gamma_k \) is the number density flux of species \( k \), and \( K_{\text{gas}} \) is the total number of gas species. The right side of the equation describes the volumetric source term of species \( G_k \) due to gas–phase chemical reactions. The number density of the dominant background is calculated using an ideal gas law constraint \( p = \sum n_k k_B T_k \), where \( p \) is the specified gas pressure and \( T_k \) is the species temperature.

2.2. Drift-diffusion approximation

The species number flux term \( \Gamma_k \) is evaluated using the drift-diffusion approximation

\[
\Gamma_k = n_k \vec{u}_k = \mu_k n_k \vec{E} - D_k \nabla n_k,
\]

where \( \mu_k \) is the species mobility, \( D_k \) is the species diffusion coefficient, and \( \vec{E} \) is the local electric field computed from the negative gradient of the electrostatic potential. This approximation is accurate at high pressures (atmospheric pressure or higher) and room temperature, where the mean free path of ions and neutral species (on the order of a micrometer) are much less than the characteristic scales of the geometry (on the order of a millimeter), and is widely used in studies of atmospheric pressure discharge.

2.3. Electron energy transport

Electron temperature \( T_e \) is determined by solving the electron energy conservation equation

\[
\frac{\partial e_e}{\partial t} + \nabla \cdot [(e_e + p_e) \vec{u}_e + \vec{q}_e] = e_e \vec{E} \cdot \vec{E} - e \sum_i i \Delta E'_i r_i - \frac{3}{2} k_B n_e \left( \frac{2 m_e}{m_b} \right) (T_e - T_g) \vec{\tau}_e,
\]

where the total electron energy \( e_e = \frac{3}{2} k_B T_e n_e \) electron pressure \( p_e \) follows the ideal gas law, and \( \vec{q}_e = -\kappa_e \nabla T_e \) is the electron thermal flux with electron thermal conductivity \( \kappa_e \). The right side of the electron energy equation includes three source terms: Joule heating, inelastic collisional heating, and elastic collisional heating. The electron unit charge is \( e \), \( \Delta E'_i \) is the inelastic collisional energy lost by an electron per collision event as described by the chemical reaction \( i \) (in units of eV), and \( r_i \) is the rate of progress of the chemical reaction. \( T_g \) is the heavy species (gas) temperature, \( m_e \) is the electron particle mass, \( m_b \) is the background species particle mass, and \( \vec{\tau}_e \) is the electron momentum-transfer collision frequency.

2.4. Heavy species temperature

The timescale associated with streamer propagation \( t_s \) is too short for use as a gas dynamic response timescale. Therefore, the background temperature \( T_g \) is kept at 300 K in all the simulations presented below.

2.5. Electrostatic potential

The self-consistent electrostatic potential \( \phi \) is calculated by Poisson’s equation

\[
\nabla^2 \phi = -\frac{e}{\varepsilon_0 \varepsilon_r} \sum_k Z_k n_k,
\]

where \( \varepsilon_0 \) is the permittivity of free space, \( \varepsilon_r \) is the relative permittivity of the material, and \( Z_k \) is the charge number of species \( k \).
2.6. Plasma chemistry and species transport model

The gas chemistry source terms $\dot{G}_k$ in the species continuity equation are evaluated using a finite-rate chemistry mechanism with a mass-action kinetics formulation. A pure argon plasma is considered and the following six species are modeled: electrons (E), argon ions ($\text{Ar}^{\pm}$), argon dimer ions ($\text{Ar}_2^{\pm}$), argon composite metastable species ($\text{Ar}^{\Xi}$), argon dimer metastable species ($\text{Ar}_2^{\Xi}$), and ground state argon atoms ($\text{Ar}$). Table 1 shows the plasma chemistry model. Reaction pathways for the pure argon reaction mechanism and reaction rate coefficients for non-electron impact reactions are obtained from a previous work [25]. Rate coefficients for electron impact reactions are calculated using an offline zero-dimensional Boltzmann solver BOLSIG+ [26] based on cross-sectional data from the literature, and are denoted as EEDF. For surface reaction mechanisms, all excited and charged species are assumed to be quenched with a unity-sticking coefficient.

Closure of the governing equations requires specification of transport properties, mobility $\mu_k$, and diffusion coefficient $D_k$ for all ions and neutral species, and the thermal conductivity of electrons $\kappa_e$. Mobility and diffusion coefficients for electrons are calculated offline using the BOLSIG + solver and tabulated as a function of the electron temperature $T_e$. Thermal conductivity is computed as $\kappa_e = \frac{2}{5} \kappa_b D_e n_e$. The mobility of argon ions is specified as a function of the local reduced electric field, as described in [25]. Diffusion coefficients are computed using Einstein’s relation. For neutral species, the transport properties are calculated using cross-sections.

Photoionization plays an important role in streamer propagation [13, 27]. In air, radiation from the de-excitation of $\text{N}_2$ molecules can photoionize surrounding $\text{O}_2$ molecules and enhance the streamer propagation speed [13, 23]. Further, Breden et al. [28] showed that the photoionization mechanism significantly impacts the speed of streamer propagation, but is not necessary for propagation [28]. This study does not consider photoionization because we focus on qualitative understanding of streamer propagation and plasma generation, and do not consider $\text{O}_2$ molecules, which are considered to be the main radiation receptors. When quantitatively comparing results with experimental data, the photoionization mechanism may be important.

2.7. Computational approach

The set of governing equations is solved with cell-centered finite volume discretization on an unstructured mesh framework, using a commercial plasma solver [22]. The special discretization of flux terms at the cell faces is evaluated using the Scharfetter–Gummel exponential scheme [29] for transport equations (species continuity and electron energy transport). A first-order backward Euler method is used for time-derivative terms. The electrostatic Poisson’s equation is solved using a semi-implicit approach [24].

2.8. Geometric configuration and conditions

We consider the simple pin-to-plane electrode geometry shown in figure 1. The inter-electrode distance between the pin and the plane electrode is fixed at 2 mm. The pin electrode form is a hyperboloid of revolution with radius 100 $\mu$m, corresponding to that used in previous works [11, 16]. The material for both electrodes is assumed to be copper.

A constant DC voltage is applied to the pin electrode, and the plane electrode is fixed to 0 V. Experimentally observed voltage waveforms in streamer discharge usually have a rise and fall time [11], but we used a constant voltage in this study because we found no data for argon streamer discharge at atmospheric pressure. Effects of voltage waveforms on discharges should be investigated in future.

Figure 2 shows the computational mesh and boundary conditions. The geometry is modeled as axisymmetric, and a hybrid mesh is used. A geometrically flexible unstructured mesh is placed near the tip of the pin electrode, and a structured mesh is placed in the region where the streamer discharge propagates. The minimum mesh size is 1 $\mu$m near the pin electrode, and the maximum mesh size is about 10 $\mu$m. The computational region in the radial direction is 3 mm. The total number of computational cells is about 440 000.

| Rxn | Reactions | Reaction rate coefficient$^a$ |
|-----|-----------|-----------------------------|
| G1  | E + Ar $\rightarrow$ E + Ar$^m$ | EEDF                        |
| G2  | E + Ar $\rightarrow$ 2E + Ar$^+$ | EEDF                        |
| G3  | E + Ar$^m$ $\rightarrow$ 2E + Ar$^+$ | EEDF                        |
| G4  | E + Ar$^m$ $\rightarrow$ E + Ar$^+$ | EEDF                        |
| G5  | E + Ar$^m$ $\rightarrow$ 2E + Ar$^+$ | EEDF                        |
| G6  | E + Ar$^m$ $\rightarrow$ E + 2Ar | EEDF                        |
| G7  | E + Ar$^+$ $\rightarrow$ Ar$^m$ | EEDF                        |
| G8  | 2E + Ar$^+$ $\rightarrow$ E + Ar$^m$ | EEDF                        |
| G9  | E + Ar$^m$ $\rightarrow$ Ar + Ar$^m$ | EEDF                        |
| G10 | 2Ar$^m$ $\rightarrow$ E + Ar + Ar$^+$ | [25]                        |
| G11 | Ar2$^m$ $\rightarrow$ 2Ar | [25]                        |
| G12 | 2Ar$^m$ $\rightarrow$ E + Ar$^+$ + 2Ar | [25]                        |
| G13 | Ar$^m$ + 2Ar $\rightarrow$ Ar2$^m$ + Ar | [25]                        |
| G14 | Ar$^+$ + 2Ar $\rightarrow$ Ar2$^+$ + Ar | [25]                        |

$^a$In units of cm$^3$ s$^{-1}$ for two-body reactions and cm$^6$ s$^{-1}$ for three-body reactions.

Figure 1. Geometry.
The boundary conditions are for a flux boundary condition at the pin electrode and a symmetric boundary at the plane electrode and outer boundaries. Previous studies [11, 16] reported that simulations with flux boundary conditions at the plane electrode tend to be unstable when secondary electron emission is considered, so we used symmetric boundary conditions at the plane electrode without secondary electron emission.

The background gas pressure is set at 1 atm, temperature is set at 300 K, and density follows the ideal gas law. Initial densities for all other species, including electrons and charged species, are set at $10^{9}$ m$^{-3}$. This condition is set that initial background (seed) electrons will ignite discharges if the electric field is strong enough to accelerate the electrons near the tip of the pin electrode and streamer head. Neutral gas flow is assumed to be stagnant, and the initial conditions are set uniformly throughout the computational region. The time step is set at $10^{-12}$.

3. Positive polarity for the pin electrode

This section presents simulation results for the propagation of primary and secondary streamers and generated plasma properties when a positive polarity voltage is applied to a pin electrode.

3.1. Positive primary streamer

When applying a positive high voltage at the pin electrode, the electric potential distribution distorts around the tip of the pin electrode, generating a strong electric field and propagating streamer discharge along a directed strong field towards the plane electrode. See [11, 30] for a detailed description of the mechanisms for generation and propagation of streamer discharge.

Figure 3 shows contour maps for simulated distributions of the reduced electric field and the electron number density. Atmospheric pressure discharge studies generally use the reduced electric field $E/N$ to discuss the electric field. Distributions of other variables, namely the electric potential and electron temperature, are shown in the appendix. Figures 3(a) and (b) show distributions at the start of streamer propagation, before arriving at the plane electrode. A filamentary peak of the reduced electric field can be seen. This peak, called the streamer head or the ionization wave front, increases the electron temperature (mean electron energy). Near the streamer head, highly energetic electron impact reactions generate electrons, ions, and radicals. The streamer head propagates towards the plane electrode. (See appendix A1 for the $+5$ kV condition.) Increased electron number density can be seen behind the streamer-passing region. This region is called the streamer channel and is in a plasma state. At 1.4 ns, the streamer head arrives at the plane electrode. Effective propagation speed between the electrodes is approximately $1.4 \text{ mm ns}^{-1} (1.4 \times 10^6 \text{ ms}^{-1})$.

The streamer head expands in size until it reaches a position of 1 mm and reduces slightly when it reaches the plane electrode surface. There is insufficient time to propagate at nearly constant size due to the small 2 mm gap, so the streamer head appears expanded and contracted. Note that previous works [11, 16] reported that under longer electrode gap conditions, the streamer head grew in the first propagation stage, became nearly constant in size at the middle of the gap, then reduced in size before arriving at the plane electrode.
Spatiotemporal behavior of a primary streamer in atmospheric pressure argon discharge when positive DC voltage of +10 kV is applied to the pin electrode. A peak in the reduced electric field (streamer head) and a high electron number density region (streamer channel) can be observed.

Figure 3. Spatiotemporal behavior of a primary streamer in atmospheric pressure argon discharge when positive DC voltage of +10 kV is applied to the pin electrode. Left is the reduced electric field and right is the electron number density for (a) 0.3 ns and (b) 1.3 ns. A peak in the reduced electric field (streamer head) and a high electron number density region (streamer channel) can be observed.

3.2. Positive secondary streamer

In streamer discharge, a high reduced-electric-field region arises from the pin electrode before and after the primary streamer arrives at the plane electrode. This is known as a secondary streamer. As shown in previous studies [11, 16], secondary streamers arise due to redistribution of space charge after a primary streamer arrives at a plane electrode. These studies showed that different species are generated in primary and secondary streamers, so it is equally important to investigate the properties of both. Regarding streamer discharge in air, comparisons between primary and secondary streamers have shown differences in generation efficiency of O, N, and OH radicals, which are thought to be important species for applications [8, 12, 15, 19].

This study focuses on secondary streamers arising at about 1.4 ns, when the primary streamer head arrives at the plane electrode. Figure 5 shows spatiotemporal behavior of the electron production rate to discuss secondary streamer generation. In primary streamer propagation (1.2 ns and 1.3 ns in figure 5), a filamentary peak of the electron production rate is observed at the streamer head. The primary streamer arrives at the plane electrode at timings ranging from 1.4 ns to 1.5 ns. A region with high electron production rates arises from the pin electrode. This is a secondary streamer, and additional species are produced in this region. A similar high-value region from the plane electrode can also be observed. This is known as a return stroke, and it disappears relatively quickly.

Note that the numerical simulation diverges after around 0.1 ns when the secondary streamer arises, due to the application of a constant voltage to the pin electrode during the entire simulation period without a rising and falling waveform. As observed in actual situations, the streamer connects a path between the pin and plane electrodes to form a highly conducting channel across the entire gap. This corresponds to observations of arcing (sparking) in previous simulation studies of pin-to-pin geometry [33]. This may represent a complicated mathematical formulation for nonequilibrium plasma phenomena. Such complex transition phenomena, in terms of applied voltage waveforms and heating of neutral gases, are beyond the scope of this study.

3.3. Distribution of species in positive streamers

Figure 6 shows obtained plasma properties of species number densities and electron temperatures for primary and secondary streamers in atmospheric pressure argon. The distributions are at about 1 mm (1.0 ns) for primary steamers and after secondary streamer occurrence (1.5 ns) for secondary steamers. In figure 6(a), increased species number densities in streamer channels and increased electron temperatures at the streamer is induced by the approach of the plasma to the plane. Electron number density increases near the peak of the reduced electric field by electron impact reactions caused by high-energy electrons. Under these conditions, electron densities ranging from $10^{20}$ to $10^{21}$ m$^{-3}$ are obtained and remain constant along the streamer channel.
Figure 4. Reduced electric field and electron number density of a primary streamer between electrodes in atmospheric pressure argon discharges when a positive DC voltage of +10 kV is applied. The primary streamer head propagates from right to left.

The increased electron temperature in front of the streamer head is due to a large reduced electric field induced by the small gap. In the primary streamer, the dominant species in the streamer channel are Ar$^m$, E, and Ar$^+$, followed by Ar$_2^+$ and Ar$_2^m$. Note that E and Ar$^+$ appear to overlap, but Ar$_2^+$ is less than 1% of Ar$^+$ and the plasma is electrically neutral. Electron number density in the secondary streamer increases to twice that in the primary streamer (see figure 6(c)) because the secondary streamer generates additional electrons.
Species composition in the plasma is unchanged between the primary and secondary streamers in atmospheric pressure argon discharge. On the other hand, there is a noticeable difference in electron temperatures between the streamers. In the primary streamer, the electron temperature was about 20 000 K within the streamer channel and about 81 000 K near the primary streamer head. In the secondary streamer, the electron temperature increased to 53 000 K in the streamer channel. This noticeable change suggests that plasma composition in the streamer channel greatly changes between the primary and secondary streamer when molecular or electronegative gases are mixed with argon gas, even if argon is the main component.

4. Negative polarity for the pin electrode

The polarity of an applied voltage is an important control parameter for realizing required plasma properties. A previous study [34] reported that a streamer head in air at atmospheric pressure diffuses to a greater extent when the applied voltage is negative, compared to when it is positive. In a plasma jet case, it is reported that a negative primary streamer was not clearly observed, despite the discharge (plasma bullet) propagating [12, 35, 36]. However, there are fewer studies of negative streamers than positive. In particular, regarding streamer discharge in noble gases such as argon at atmospheric pressure, there are few studies of discrepancies in basic discharge types depending on the polarity of the applied voltage. The generation efficiency of metastable species leading to a Penning effect may change. This section presents simulation results for a negative voltage applied to the pin electrode. The other settings are identical to the positive voltage case described in the previous section.

4.1. Negative primary streamer

Figure 7 shows spatiotemporal distributions of the reduced electric field and electron number density when a negative voltage of \(-10\) kV is applied to the pin electrode. (See appendix A2 for the \(-5\) kV condition.) As in the case where a positive voltage is applied, there is a filamentary peak of the reduced electric field and an increase in electron number density. Unlike the positive streamer, however, in the negative streamer a distorted (sharp) streamer head propagates from the pin to the plane electrode. The negative primary streamer head arrives at the plane electrode at 0.6 ns, and the effective propagation speed between the electrodes is estimated to be about 3.3 mm ns\(^{-1}\) (3.3 \(\times\) 10\(^6\) ms\(^{-1}\)). The distorted shape makes it difficult to evaluate the size of the streamer head, but it appears smaller than that in the positive case.

Figure 8 shows several temporal distributions of the reduced electric field and electron number density between the electrodes. The reduced electric field in the negative streamer was a nearly constant value of about 900 Td during propagation. An increase of the reduced electric field is observed before arriving at the plane electrode as for the positive streamer, but this increase is smaller. There is a remarkable peak in the reduced electric field at the streamer head, due to the sharp, small streamer head shape. Electron number density generated by passing of the streamer head is in the order of
Figure 6. Simulated species number density and electron temperature as a function of position when a positive DC voltage of +10 kV is applied for (a) primary and (b) secondary streamers, and (c) differences in electron number density between streamers. Plasma species composition is identical between primary and secondary streamers, but electron temperature is noticeably higher in the secondary streamer.

1.0 $\times$ 10$^{21}$ m$^{-3}$ and higher than that of the positive streamer (1.0 $\times$ 10$^{20}$ m$^{-3}$), except near the pin electrode in the positive streamer (1.0 $\times$ 10$^{21}$ m$^{-3}$).

4.2. Negative secondary streamer

Figure 9 shows the spatiotemporal behavior of electron production rates, to discuss the generation of secondary streamers when a negative DC voltage of −10 kV is applied. As figures 9(a) and (b) show, peak electron production rates are observed at the primary streamer head, as in the case of a positive 10 kV streamer, however the shape differs from that shown in figure 5. At about 0.6 ns (figure 9(c)), the negative primary streamer arrives at the plane electrode. Unlike the positive streamer, no high-electron-production region from the pin electrode was observed; only a return-stroke-like distribution was observed in negative streamer.

4.3. Distribution of species in negative streamers

The following describes the plasma properties generated by negative streamers. Figure 10 shows species number density over time and electron temperatures at a 1 mm position between the electrodes for primary (0.3 ns) and secondary (0.7 ns) streamers when a negative voltage of −10 kV was applied. The dominant species in plasma generated by negative streamers were Ar$^+$, E, and Ar$^+$, followed by Ar$^{2+}$ and Ar$^{2+}$. This tendency is identical to the case of positive streamer plasma, but the electron number density increased by ten. As figure 10(a) shows, this increase corresponds to the increased electron temperature (mean energy) to 200 000 K due to the larger reduced electric field at the negative primary streamer head. Further, the sharper streamer head compared to a positive streamer makes the peak in the reduced electric field remarkable and local. Electron temperatures in the negative streamer channel are about 35 000–50 000 K, compared with about 20 000 K in the positive streamer channel.

In the negative secondary streamer (figure 10(b)), there is a small difference in species number density compared to the primary one, but the electron temperature increased to 50 000 K. Figure 10(c) shows the discrepancy between the primary and secondary streamer. Near the pin electrode, the increase in the electron number density is small due to nonexistence of the secondary streamer, but a larger increase is observed near the plane electrode because of the return stroke, as figure 9 shows.
Figure 7. Spatiotemporal behavior of a primary streamer in atmospheric pressure argon discharge when negative DC voltage of $-10\,\text{kV}$ is applied to the pin electrode. Left is the reduced electric field and right is the electron number density for (a) 0.2 ns and (b) 0.5 ns. A distorted (sharp) streamer head can be observed, unlike the positive streamer.

5. Discussion

5.1. Relations between applied voltage, streamer head size, and propagation speed in positive streamers

Figure 11 shows simulated properties of the positive streamer head. The size (diameter) of the streamer head was estimated from the radial peak position of the reduced electric field when the primary streamer head passed at a position of about 1 mm. Propagation speed was estimated from the time the streamer head arrived at the plane electrode considering a 2 mm gap. As figures 11(a) and (b) show, the diameter and propagation speed of the streamer head increase with the applied voltage, while saturation is observed about the diameter at the high-voltage region. From these two figures, we can read the relation between size and propagation speed in the positive primary streamer head. Propagation speed and diameter are nearly proportional when the diameter is relatively small, and propagation speed greatly increases when the diameter becomes relatively large.

Naidis discussed the relation between streamer velocity and diameter, analytically obtaining a relation between the reduced electric field, streamer propagation speed, and streamer head diameter [19]. He showed that propagation speed is basically proportional to diameter and deviates at high-speed regions, and many experimental results support this tendency. Naidis also showed the existence of a minimum sub-millimeter size for positive streamers. Our results also suggest a minimum size of about 0.1 mm, corresponding to zero streamer velocity. This qualitatively supports the validity of our model and results.

5.2. Dependence of electron number density on applied voltage in positive streamers

Figure 12 shows electron number density in a streamer channel when it passes a position of 1 mm as a function of applied voltage. With increasing applied voltage, a decrease in electron number density was observed to 10 kV, followed by an increase at 12 kV. This tendency is somewhat surprising, as we expected the increased size and propagation speed of the
Figure 9. Spatiotemporal behavior of electron production rate in atmospheric pressure argon discharge when a negative DC voltage of $-10 \text{kV}$ is applied to the pin electrode. Secondary streamer from the pin electrode is not observed and only return stroke is observed.

Figure 10. Simulated species number density and electron temperature as a function of position in (a) primary and (b) secondary streamers and (c) the difference in electron number density between the streamers when $-10 \text{kV}$ is applied.
of electron movement between positive and negative voltages. In positive streamers, electrons gather toward the pin electrode and central axis. In contrast, electrons spread away from the pin electrode and central axis in negative streamers. This spread leads to a diffuse shape of the negative streamer and to a corona-like discharge, with lower electron number densities observed under low-voltage conditions.

5.4. Comparison with streamers in air plasma

The following compares argon and air streamers when positive voltage is applied. One remarkable difference from air streamers is a lower decrease of electron number density in streamer channels. As figure 4 shows, electron number density is unchanged between times under propagation. This is due to the low rate of electron loss reactions in argon and noble gas discharges, because electron attachment reactions to form negative ions do not exist. A previous study [39] similarly showed that electron number density does not decrease behind the streamer head in streamer discharge generated within an atmospheric pressure helium plasma jet. Another study of differences between N\(_2\) and O\(_2\) streamer discharge showed that a decreased electron number density is observed in an O\(_2\) streamer channel and that the decrease is negligible for N\(_2\) [40]. Therefore, a small decrease of electron number density in a streamer channel can be considered as a feature of discharges in electropositive gases. The very short propagation time of 1.4 ns may emphasize this tendency in our conditions.

Regarding the reduced electric field at the streamer head, values are of the same order of magnitude between argon and air streamers. This may be because deviation in space charge is mainly due to fast-moving electrons and mass differences among ions are very small, compared to mass differences between electrons and ions.

As shown in previous studies [15, 16], dominant species are unchanged between primary and secondary streamers in argon discharge, but differ in air discharges. In atmospheric pressure argon streamers, electron generation is mainly due to electron impact reactions, which increase with increasing electron temperature. Therefore, increased electron temperature in

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5.3. Change of discharge mode in negative streamers

As figures 7 and A2 show, we observed a remarkable change in discharge mode between high and low voltages when the voltage polarity was negative. Under high-voltage conditions, a negative streamer with a diffusive shape propagated among the electrodes. This tendency qualitatively agrees with previous studies in air with negative applied voltage [34, 38]. This diffusiveness is likely induced by differences in the direction
an argon secondary streamer simply increases electron number density in the streamer channel.

It is also worth pointing out the similarities and differences between obtained streamer speed and plasma bullet speed in a plasma jet. Reuter et al measured the velocity of a pure argon plasma bullet, which was about 0.07 mm ns\(^{-1}\) \((7 \times 10^5 \text{ ms}^{-1})\), slower than our obtained results. The typical speed of the plasma bullet and streamer is respectively known as being from \(10^3\) to \(10^5 \text{ ms}^{-1}\) and \(10^6 \text{ ms}^{-1}\) [13, 14]. These results support previous results and also qualitatively our results.

5.5. Polarity-dependent streamer phenomena

When positive voltage is applied to the pin electrode, a primary streamer propagates toward the plane electrode, followed by a secondary streamer from the pin electrode when the primary streamer arrives at the plane electrode. The streamer head diameter correlates with propagation speed. Both the streamer head size and the propagation speed increase with voltage applied to the pin electrodes, reaching 1.4 mm and 1.5 mm ns\(^{-1}\) at 10 kV. On the other hand, the electron number density at the primary streamer head decreases with increased applied voltage, due to acceleration of the propagation speed reducing chemical reactions. Additional electrons are generated by propagation of the secondary streamer. The electron number density is twice that of the primary streamer in the 10 kV case.

When negative voltage is applied to the pin electrode, different discharge types are observed at high and low voltages. With high negative voltage, the primary streamer propagates to the plane electrode, as with positive voltage, but the shape is more diffuse and propagation speed is faster, with no observed secondary streamer. At low negative voltage, the discharge seemed to be a corona type rather than a streamer, and low-density plasma is generated around the pin electrode. Dominant species generated by discharges are \(\text{Ar}^m, E, \text{ and Ar}^+\), followed by \(\text{Ar}_2^+\) and \(\text{Ar}_3^m\). This tendency was unchanged with the polarity of the applied voltage and with differences in the first and secondary streamers.

Differences of streamer head shape were observed between positive and negative cases. In the positive case, the streamer head is clearly seen; in contrast, in the negative case, the streamer head becomes sharp and small. This tendency agrees with previously obtained experimental results, which report that the negative streamer head and plasma bullet are diffusive or unclear [12, 34, 35].

5.6. Candidate choice of voltage polarity for low-voltage operations

The definition of appropriate plasma properties depends on the application and objects to be exposed to the plasma. Generally, safety and an electric source costs call for low-voltage operations that maintain a high plasma density. A positive voltage may be appropriate for argon atmospheric pressure discharge, to avoid the unintended changes in discharge type observed under negative voltage. Simulations in this study were conducted under ideal, nonperturbed conditions, while actual situations will include uncertain factors such as disturbances in background temperature or flow, mechanical vibrations, and voltage fluctuations of electric sources. Further, generation of a secondary streamer is preferable to increase plasma density.

From the perspective of low-voltage operations, therefore, positive polarity of voltage applied to the pin electrode is appropriate, as it provides relatively high-density plasma, stable streamer propagation, and secondary streamer generation. Simulation-based studies are also useful for developing atmospheric pressure plasma applications.

6. Conclusions

We studied streamer propagations in atmospheric pressure argon streamer discharge by a self-consistent, multi-species, multi-temperature plasma fluid modeling. We conducted a parametric study for positive and negative polarity of the applied voltage under a pin-to-plane geometry with a gap of 2 mm. The obtained knowledge was as follows. First, a positive primary streamer propagates in plasma with high electron density, then the secondary streamer further increases plasma density, generating abundant metastable species. We obtained a relation between streamer head diameter and its propagation speed, for instance, 1.4 mm and 1.5 mm ns\(^{-1}\) at 10 kV. Second, a negative streamer diffusely propagates in the inter-electrode gap under high-voltage conditions. We observed a corona-type discharge at low negative voltage. Since the generated species maintained both positive and negative polarities, a stable high density of \(10^{20}\) to \(10^{22} \text{ m}^{-3}\) under a positive low-voltage application has advantages for developing argon-based atmospheric pressure plasma applications.

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Appendix A1. Positive primary streamer at low voltages

Figure A1 shows simulated distributions of the reduced electric field and the electron number density when a positive DC voltage of +5 kV was applied to the pin electrode. The sizes of the streamer head and channel and the propagation speed decreased. The dependencies of positive streamer properties on applied voltage values are described below in the discussion. Similar streamer propagation was observed at 12 and 4 kV, but no discharge or plasma generation was observed below 3 kV.
Figure A1. Spatiotemporal behavior of the primary streamer in atmospheric pressure argon discharges under positive DC voltage of +5 kV.

Appendix A2. Negative primary streamers at low voltages

Figure A2 shows spatiotemporal distributions of the reduced electric field and electron number density when a negative voltage of −5 kV was applied to the pin electrode. There are remarkable differences in the −10 kV distributions, which show a corona-type discharge rather than a streamer discharge. No filamentary peak of the streamer head was observed, and the plasma region filled the space between the electrodes at a time scale in the order of 10 ns. There is a decrease in electron number density from $10^{17}$ to $10^{16} \text{m}^{-3}$, with the exception of $10^{21} \text{m}^{-3}$ near the electrode. The discharges were streamer types at −12 and −10 kV and corona types at −5 and −4 kV.

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ORCID iDs

Yosuke Sato  https://orcid.org/0000-0002-2772-2332
Kenji Ishikawa  https://orcid.org/0000-0002-8288-6620
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