Uniform and stable plasma reactivity: Effects of nanosecond pulses and oxygen addition in atmospheric-pressure dielectric barrier discharges

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ABSTRACT

Uniform and stable reactivity of atmospheric pressure plasmas is a prerequisite for most applications in fields ranging from materials' surface processing, environment protection, to energy conversion. Dielectric barrier discharges (DBDs) are among the most promising plasmas to satisfy these requirements. However, the unpredictable and uncontrollable transitions between discharge modes, the limited understanding of the DBD ignition and extinction processes, and the complexity of plasma chemistries and reactions with admixture gases restrict their adoption in industry. Here, we report a practically relevant and elegant solution based on using customized nanosecond (ns) pulse excitation and precise addition of oxygen to an Ar flow. The effects of ns pulses and oxygen on the uniformity and reactivity of the DBD are investigated via quantifying the gap voltage U_g and the discharge current I_g from the current-voltage measurements and quantitative discharge imaging. The electron density, n_e , is estimated with U_g and I_g . With increasing U_g , more electron avalanches are ignited and overlap, which facilitate n_e , T_e , and discharge uniformity, while high U_g induces excessive electrons generated with high ionization rates, resulting in the distortion of the space electric field and reduced stability and uniformity. A small amount of added oxygen favors the production of electrons. Overdosed oxygen molecules capture electrons causing a drop in n_e and T_e and couple with the effect of the electrical field resulting in the filamentary discharges or complete plasma extinction. The mechanism of the effects of ns pulses and oxygen addition on the uniformity and reactivity of plasmas is based on the electrical measurements and discharge image analysis and is cross-validated by optical emission spectra measurements and the ratio of the Ar intensities' calculations as indicators of the variation in n_e and T_e . The results in this work contribute to the realization and controllability of uniform, stable, and reactive plasmas at atmospheric pressure.

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I. INTRODUCTION

Low pressure plasma processes can be precisely controlled and have desirable spatial uniformity and temporal stability, which are crucial for many applications, including atomic layer deposition and etching^{1,2} and micro-nano-pattern surface functional modifications.^{3,4} With the advantages of short processing time, low energy consumption, and without the need for vacuum equipment, atmospheric pressure plasmas have recently been actively investigated

and achieved inspiring results in materials processing,^{5,6} environment protection,^{7,8} energy conversion and storage,^{9–11} and other areas. However, from the industrial application point of view, it is necessary to achieve uniform plasmas with stable reactivity. At atmospheric pressure, the electron mean-free path is small (less than 1 mm), and in order to obtain sufficient ionization and discharge ignition energy, the applied electrical field between the electrodes needs to be high, which enhances the difficulty to maintain

the plasma uniformity and stability especially during industrial processes. Due to the dielectric layer(s) between the electrodes limiting the current and preventing arc formation, dielectric barrier discharges (DBDs) are among the most promising methods to provide uniform and stable plasmas.^{12,13}

Due to their short discharge duration and high breakdown voltage, pulsed voltage excited discharges are among the main choices to improve the plasma uniformity and reactivity.^{14–18} Nevertheless, in pulsed DBDs, because of the unclear understanding of the ignition and extinction processes, especially with reactive admixtures, the uniformity and stability are still uncontrollable. The two key problems are due to insufficient design of pulses to monitor the excitation electric field and the instability caused by the added reactive gases. Without a comprehensive understanding and solving of these problems, the discharge evolution in fast-timescales is unpredictable, which results in poor discharge stability and cannot meet the requirements for industrial processing.^{19,20}

For specific applications requiring unique plasma chemistry, reactive admixtures need to be added to the DBD which participate in the physicochemical processes and greatly enhance the chemical reactivity.^{21–24} The plasma chemistry is determined by the plasma chemical reaction rates, largely initiated by energetic electrons, which depend on the electron energy distribution function (EEDF). For pulsed DBDs, the breakdown voltage is high and the plasma excitation electric field changes dramatically during the breakdown. With the added reactive species, the plasma chemistry and the space charges distort the EEDF, initially shaped by the applied electric field, as well as the discharge uniformity and stability. Therefore, it is extremely challenging to control and monitor the plasma uniformity and reactivity with reactive admixtures.^{22,25–27}

Oxygen addition can produce a diverse range of reactive oxygen species that are used in film etching, volatile organic compounds (VOCs) oxidation, and biomedical treatments.^{28–30} A small amount of oxygen can promote Penning ionization and electron avalanches with inert gas discharges, and with the excessive oxygen added, the number of electron is reduced by forming O_2^- ions, which aggravate the uneven distribution of the electric field and easily form filaments in a discharge gap.^{20,31–33} For pulsed DBDs, in particular, due to the presence of a dielectric barrier, the plasma excitation electric field does not depend on the applied voltage but rather the voltage drop across the discharge gap, which cannot be measured directly.^{17,34} Owing to the difficulty of monitoring the voltage across the discharge gap and the sensitivity of reactive species on plasma chemistry, the uniformity, stability, and reactivity of pulsed DBDs are difficult to control by simply adjusting the process parameters. For the purpose of optimizing the plasma reactivity and uniformity of the oxygen containing pulsed DBD in applications, it is important to understand the discharge phenomena, which determine the behavior of the discharge.

In this work, a practical approach is proposed to investigate the effects of nanosecond (ns) pulses and precisely dosed oxygen on the pulsed DBD plasma uniformity and reactivity. The voltage on the discharge gap and discharge current are estimated from the measured applied voltage and current to explore the effects of the electric field and space charges on discharge uniformity quantified by time-integrated discharge imaging and analysis. Optical emission spectroscopy measurements are used as an indicator of plasma

reactivity, and the variation of T_e , and cross-validate the results obtained from the study of electrical characteristics. With the tuning of the applied electric field and space charges, the discharge uniformity and reactivity are optimized. The findings and mechanism in this work will help improve atmospheric pressure plasma uniformity and reactivity, ultimately advancing practical applications.

II. EXPERIMENTAL SETUP

The experimental setup is illustrated schematically in Fig. 1. It is composed of an ns pulse power supply, an electrical measurement system, an optical measurement system, a discharge reactor, and a gas supply system. The voltage of the ns pulse power supply (Xi'an Lingfeng source HV-2015) can be continuously adjusted from 0 to 15 kV at a pulse repetition frequency (PRF) from 0 to 100 kHz. The rising (t_r) and falling (t_f) times of the voltage are in the range of 50–500 ns, and the pulse width (t_w) time is in the range of 0–1 ms. During the experiment, the PRF is fixed at 5 kHz, both t_r and t_f are fixed at 50 ns and t_w is fixed at 1200 ns. The reactor is made of stainless-steel and can be pumped down to the vacuum below 2 Pa. The stainless-steel parallel plate electrode (50 mm in diameter and 5 mm in thickness) is placed in the center of the reactor with a quartz glass (80 mm in diameter and 2 mm in thickness) covering the ground plate. The gas gap can be adjusted from 0 to 40 mm and is fixed at 2 mm during the experiment.

The voltage and current are measured with a 1:1000 high-voltage probe (North Star Research Inc., PVM-5), a current probe (Pearson 6560), and an oscilloscope (Tektronix TDS 3052B). A reference capacitor (C_r) of 2.2 nF is inserted in series with the ground electrode. The voltage across C_r is measured using a 1:10 voltage probe (Tektronix P6139) to estimate the voltage on the dielectric barrier. The optical emission spectra emitted from the discharge are measured by a spectrometer (Ocean Optic HR4000CG, 250–900 nm range, 0.7 nm resolution) with 100 ms integrated time for 500 discharges. The discharge images are recorded by a digital camera (Canon EOS 6D) with 1/8 s exposure time. High purity Ar (99.999%) and O_2 (99.999%) and a mixture gas of 1% O_2 in Ar are used as discharge gases and are controlled by mass flow meters (Sevenstar D08-4F). The studied O_2 content ranges from 0.01% to 10%. The background impurities have no significant influence on the experimental results.

III. RESULTS AND DISCUSSIONS

A. Discharge uniformity of argon DBD

The uniformity of discharge is closely related with the plasma treatment effect, which is mainly determined by the space electric field in our studied system. The adjustment of the applied voltage can effectively influence the electric field in the gas gap in a DBD and the discharge uniformity. Figure 2 shows the discharge images of the Ar DBD at different applied voltages from 3.1 kV to 5 kV. The exposure time of the digital camera with 1/8 s is the integration result of 625 times discharge. The uniformity of discharge in this study is not a time-resolved result but an integrated effect, which is useful for plasma treatment in industrial applications. It can be observed that at 3.1 kV, the discharge is weak while the plasma

FIG. 1. The experimental setup of the ns pulsed DBD.

does not fill the whole space. The discharge is in the filamentary mode but with dim filaments. With the increase in the voltage to 3.2 kV and 3.25 kV, the discharge becomes stronger with the whole space becomes filled with plasma. Interestingly, the number of the observed filaments decreases with the increase in the applied voltage. When the applied voltage is further increased to 3.5 kV, it is difficult to distinguish the filaments from the discharge image. At 3.75 kV, 4 kV, 4.25 kV, and 4.5 kV, the discharge is very uniform, while the increased applied voltage just enhances the discharge intensity as evidenced by the higher brightness in the discharge images. With the applied voltage increased to 4.75 kV and

5 kV, there are bright thin filaments appearing in the plasma, which lowers the discharge uniformity.

Although the variation of the discharge can be seen from the observed discharge images, it is hard to compare the discharge uniformity for the discharge images visually. To analyze the discharge dynamic and spatial uniformity, researchers have employed fast Fourier transform, gray-level histogram, spatial correlation function, etc., as image processing methods.^{19,35,36} In our experiment, the discharge images are observed from the side with 1/8 s exposure time. Considering that the filaments bridge the high voltage plate electrode and the dielectric barrier, the middle horizontal line can

FIG. 2. Discharge images of the ns pulsed Ar DBD at different applied voltages (photo exposure time is 1/8 s).

be used to represent the plasma uniformity. Without considering the two-dimensional (2D) uniformity and the periodic pattern of the plasma in space, the image processing method can be simplified and the gray value standard deviation (GVSD) method has been used to quantitatively analyze the plasma uniformity. At first, the gray values of the pixels on the 40 mm long horizontal line in the middle of the discharge area are obtained by graying processing and divided the length of plasma to get the values of x_n . With the averaged value x_m of x_n , to standardize the value of the pixels, the zero-mean normalization is obtained from Eq. (1),

$$\bar{x}_n = \frac{x_n - x_m}{\nu}, \quad (1)$$

where $\bar{x}_n$ represents the zero-mean normalized pixel value and ν is the variance. For the evaluation of discharge uniformity, the gradient value of $\bar{x}_n'$ is obtained by differentiation of $\bar{x}_n$. The standard deviation of $\bar{x}_n'$, GVSD, is used to represent the uniformity of the discharge.

Figure 3 shows the GVSD of the discharge images of Fig. 2 at different applied voltages from 3.1 kV to 5 kV. It can be observed that the GVSD decreases from about 10 at 3.1 kV to about 1.3 at 3.5 kV, pointing to an improvement in plasma uniformity and is consistent with the observation from the discharge images. In the voltage range from 3.75 kV to 4.5 kV, the values of GVSD are below 1, representing discharges which are very uniform, which is also seen in the corresponding discharge images. Therefore, for this work, we define a uniform discharge for plasmas with a GVSD value below 1. With the appearance of bright filaments in discharge images at 4.75 kV and 5 kV, the values of GVSD increase to about 3.1 and 6.6, respectively. The values of GVSD are consistent with the variation of the discharge uniformity.

FIG. 3. GVSD of the ns pulsed Ar DBD at different applied voltages.

The effects of the applied voltage on the discharge uniformity can be explained as follows. When the applied voltage is low (3.1 kV in our experiment), the electric field strength is low, and the free electrons require a long distance in the gap to obtain sufficient energy to ionize the background gas molecules. It reduces the probability of the formation of electron avalanches. Without sufficiently overlapped electron avalanches channels, the discharge is weak and only fills the space between the electrodes partially with many individual dim filaments.

With the increase in the applied voltage, the electric field is strengthened, and the number of the electron avalanches is increased through more effective collisional ionization. The intensity of the discharge is increased, and with the overlapped electron avalanche channels, the discharge uniformity is improved. Within the voltage range from 3.75 kV to 4.5 kV, more electrons are generated, and collisional ionization occurs in the whole space resulting in a uniform discharge mode. When the electric field strength is increased further, it increases the reduced field strength E/n affecting the reaction rates and the Townsend ionization coefficient. A small overvoltage can greatly promote the production of electron avalanches. Therefore, the discharge turns into filamentary mode with bright filaments.

B. Discharge voltage and current estimation

From the discharge image observation and the GVSD analysis, it can be seen that the applied voltage has profound influence on the discharge uniformity by monitoring the electric field in space. However, for DBDs, the applied voltage U_t is composed of the voltage drop across the gap, U_g , and the voltage across the dielectric barrier, U_d . Correspondingly, the measured current I_t is divided into conduction current through discharge, I_g , and displacement current, I_d , for the capacitances in DBD.^{17,37} Since U_g determines the reduced electric field, E/n , in discharge space and I_g represents the space charges, the variation in U_g and I_g is crucial to reveal the mechanisms of the discharge uniformity and the ignition, development, and extinction process of a DBD.

To obtain U_g and I_g from the measured U_t and I_t , a reference capacitor, C_r , is inserted between the grounded electrode and the ground. The voltage on the reference capacitor U_r should be proportional to U_d since both voltages are across the capacitors. The equivalent electrical circuit is shown in Fig. 4. An amplification coefficient can be selected to estimate U_d by the measured U_r with the method reported in Ref. 17, which assumes that U_g should be near zero when the discharge is extinguished. Therefore, U_g can be obtained by subtracting the estimated U_d from U_t . U_b , U_d , and U_g at 3.5 kV and 5 kV are shown in Figs. 5(a) and 5(c). To extract I_g from I_t , I_d is obtained by pumping the reactor to vacuum with the voltage applied. Without the electrical discharge, the obtained current should be I_d . Then, I_g can be obtained by subtracting I_d from I_t . Figures 5(b) and 5(d) show I_b , I_d , and I_g at 3.5 kV and 5 kV, respectively.

It can be seen from Fig. 5 that the peak values of U_g are much smaller than those of U_b , which is a clear demonstration of the need of voltage separation. The peak moments of U_g , about 86 ns at 3.5 kV and 50 ns at 5 kV, are almost the starting times of I_g , about 85 ns at 3.5 kV and 55 ns at 5 kV. With the increase in U_b ,

FIG. 4. Equivalent electrical circuit of DBD.

the peak value of U_g increases. The peak moment of U_g also moves forward, which induces early breakdown and higher I_g . With the increase in E/n , the electron avalanches are accelerated, while the electron migration velocity is also increased. Therefore, it can be noticed that the rising time of the first peak of I_g decreases from about 35 ns (from 85 ns to 120 ns) at 3.5 kV applied voltage to about 23 ns (from 55 ns to 78 ns) at 5 kV applied voltage.

Figure 6 shows the peak values of U_g and I_g in the applied voltage ranging from 3.25 kV to 5 kV. It can be seen that the U_g increases from about 2.2 kV to 3.5 kV, and I_g is not linear to U_g but increases from about 0.5 A to 5.5 A, which grows much faster than U_g . It is because the increase in U_g results in the increase in E/n . The E is obtained by the breakdown voltage (peak value of U_g) dividing the gap distance. The plasma sheath can be formed in atmospheric pressure DBD as a few tens of μm thickness with a few hundreds of kV cm^{-1} electric field,³⁸ which affects the E/n in space. But the formation cathode sheath in positive pulsed DBD needs more than 10 ns,³⁹ and at the breakdown point, it is reasonable to estimate E/n without considering the sheath. With a higher E/n , the ionization rate is increased and more electrons are produced.^{17,40,41} The electron drift velocity is also increased, which also contributes to a higher I_g . With the promoted electron avalanches, the discharge changes from partial discharge to uniform discharge and then to filaments discharge. Therefore, the discharge uniformity is sensitive to the variation in the amplitude voltage.

Assuming uniform discharge, the electron density, n_e , can be estimated from the following equation:

$$\frac{I_g}{S} = en_e V_{\text{drift}}, \quad (2)$$

where S is the area of discharge, e is the elementary charge, and V_{drift} is the drift velocity of electron, which is defined in the following equation:

$$V_{\text{drift}} = \mu_e E, \quad (3)$$

where μ_e is the electron mobility. With the peak values of U_g on gap increasing from 2.15 kV to 3.61 kV, the E/n raised from 44 Td to 74 Td ($1 \text{ Td} = 10^{-17} \text{ V cm}^2$). The electron drift velocity can be obtained by resolving the Boltzmann equation with the BOLSIG+ code over the range of E/n .⁴² The n_e is estimated to change from $1.15 \times 10^{17} \text{ m}^{-3}$ to $2.76 \times 10^{17} \text{ m}^{-3}$ with U_t increases from 3.25 kV to 5 kV. The estimated n_e is based on the assumption of uniform discharge, and the values of n_e are more accurate for the voltage from 3.75 kV to 4.5 kV with uniform discharge mode. In the filamentary discharge conditions, n_e in filaments is higher and in space is lower than the estimated value.

The variation in electrical parameters also influences the emission spectral characteristics of the Ar DBD. Figure 7 shows the emission spectra recorded at 3.5 kV and 5 kV. Figure 8 shows the variation of a group of Ar line intensities on the studied applied voltage ranging from 3.5 kV to 5 kV. It can be seen that the intensities of the Ar lines are increased with the increase in the applied voltage, which indicates the increase in n_e , and the distributions of the Ar line intensities are also changed for the change in the electron energy distribution, which affects the electron temperature, T_e , and plasma-chemical reaction efficiency. The emission intensity, I , of direct electron impact excitation Ar, can be given by¹⁷

$$I \propto C_\lambda A \tau [N] n_e k, \quad (4)$$

where C_λ , A , and τ are the spectral response of the optical measure system, the Einstein coefficient, and the effective lifetime (related with the quenching processes), respectively; $[N]$ is the number density of the ground state Ar; and k is the excitation rate. Considering the electron energy distribution follows, the Maxwellian function, T_e , can be determined by the ratio of the emission intensities with the corresponding excitation rates at specific E/n , as shown in the following equation:¹⁷

$$\frac{I_1}{I_2} = \frac{C_{\lambda 1} A_1 \tau_1 k_1}{C_{\lambda 2} A_2 \tau_2 k_2}. \quad (5)$$

Without the calibration of spectral response of the optical measure system and the absence of the related quenching rate coefficients and excitation rates, it is difficult to obtain accurate T_e values directly. However, the ratio of the emission intensities with different excited energies can provide the trend of T_e . The Ar I (696.5 nm) and Ar I (912.3 nm) lines are independent from other lines, and there is approximately a 0.4 eV difference in the excitation energies (13.3 eV and 12.9 eV, respectively). Therefore, the

FIG. 5. Discharge gap voltage separation and current separation at 3.5 kV (a) and (b) and 5 kV (c) and (d).

ratio of the emission intensities of Ar I (696.5 nm) and Ar I (912.3 nm) is selected to indicate the trend of T_e at different applied voltages, which is shown in Fig. 9.

From Fig. 9, it can be seen that with the increase in applied voltage, the ratio of the intensities of Ar I (696 nm) and Ar I (912 nm) is increased. Considering the excitation energy of Ar I (696 nm), 13.3 eV, is higher than that of Ar I (696 nm), 12.9 eV, it means more high energy electrons than low energy electrons are produced and T_e is increased. When the applied voltage reaches 4.25 kV, the ratio becomes saturated. It is because with the increase in the applied voltage and E/n , electrons gain more energy in space

and T_e is increased correspondingly. However, with the further increase in the applied voltage, more electrons are generated with higher ionization rate, and there are also strong electrical field distortions in space, which affects the further acceleration of electrons and forms filaments. T_e of the electrons in whole space becomes saturated.

C. Effect of oxygen addition on discharge uniformity

Ar is an inert gas, and there is a lack of plasma chemical activity in pure Ar discharges. O_2 is added in Ar DBD to increase the

FIG. 6. The peak values of U_g and I_g with varying applied voltages.

plasma oxidative ability for practical applications. The discharge characteristics after oxygen addition are investigated. Figure 10 shows the discharge images of Ar DBD at 4 kV with different O_2 contents. It can be seen that the addition of O_2 affects the discharge uniformity profoundly. At a small O_2 content below 0.1%, the discharge shows uniformity. With a higher O_2 content, the discharge becomes non-uniform and nearly distinguished.

To analyze the effect of O_2 addition on the plasma uniformity of Ar DBD quantitatively, the GVSDs of the Ar DBD at 4 kV, 4.25 kV, and 4.5 kV with different O_2 contents are calculated and presented in Fig. 11. It can be seen that at a lower O_2 contents, the Ar DBD at 4 kV, 4.25 kV, and 4.5 kV remain in a uniform mode with GVSD values below 1. When the O_2 content increases, the GVSD values peak indicates poor discharge uniformity. This behavior is because O_2 is an electronegative gas, where electrons can attach and form O_2^- ions. The electron attachments and O_2^- ions in space would distort the space electric field and promote the formation of filaments, which results in the rising of GVSD and

non-uniform discharge,

where M is either Ar or O_2 molecule. With the further increase in O_2 content, the number of free electrons decreases, and there are no enough electron avalanches formed in space. The discharge fills the space partially with many individual dim filaments and the breakdown voltage increases correspondingly. It is also noted that the GVSDs of 4.5 kV and 4.25 kV show more clear peaks than the GVSD of 4 kV. It is because that the E/n of 4.5 kV and 4.25 kV is higher than that of 4 kV. With the addition of O_2 , the filaments and the electrical field distortion are more evident and appear at a lower O_2 content. Therefore, there is a higher peak in the GVSD

FIG. 7. The emission spectra of Ar DBD at 3.5 kV and 5 kV.

FIG. 8. The selected Ar emission lines intensities at different applied voltages.

FIG. 9. The ratio of emission intensities of Ar I (696 nm) and Ar I (912 nm) at different applied voltages.

values for the higher voltages than that of lower voltages at a lower O_2 content.

To further investigate the effect of O_2 addition on the electrical characteristics of the Ar DBD, U_g and I_g are calculated at 4.5 kV with 0.1%–10% O_2 content, which are shown in Fig. 12. n_e is also estimated with Eq. (2) and the separated U_g and I_g with uniform discharge assumption. It can be seen that with a small amount O_2 addition (from 0.1% to 0.4%), U_g decreases slightly, which corresponds to the filament discharge stage. This means the filament channels bridge the electrodes and release the electric power. n_e is

FIG. 11. GVSD of the Ar DBD at 4 kV, 4.25 kV, and 4.5 kV with different O_2 contents.

also increased from 0.1% to 0.4% O_2 addition, which shows the promotion of the production of free electrons with small amount O_2 addition especially with U_g decreased in the meantime. With more O_2 added, to about 1%, U_g maintains at a high level for the less conductivity of the filament channels for the O_2^- ions generation with the attachment of electrons on O_2 . With the O_2 content increasing from 1% to 4%, more charged particles of O_2^- fill the space and a larger proportion of the applied voltage applied across the dielectric barrier other than the gas gap, which induces the slight decrease in U_g . But n_e is still maintaining in high level at

FIG. 10. Discharge images of the Ar DBD at 4 kV with different O_2 contents (photo exposure time is 1/8 s).

FIG. 12. The peak values of U_g and I_g and the estimated n_e with different O_2 contents.

about $2.5 \times 10^{-17} \text{ m}^{-3}$. With further O_2 addition, the discharge goes toward extinguishment with lack of free electrons and drop of U_g . Then, I_g and n_e drop significantly. It is noticed from the curves of I_g and n_e that the dropped proportion of I_g is larger than that of n_e , which indicates the drop of mobility and drift velocity. The reason is the more O_2^- ions generation.⁴³ It is worth noting that the estimated n_e is more accurate in the uniform discharge mode with O_2 content below 1% and for higher O_2 content DBDs with filamentary discharge conditions, n_e in filaments is higher and in space is lower than the estimated value.

The addition of O_2 also affects the emission spectra of the Ar DBD as shown in Fig. 13 with 0.1% and 1% oxygen contents. Figure 14 shows the variation of a group of Ar line intensities on the studied O_2 content ranging from 0.1% to 10%. It can be seen

from those figures that the emission intensities of the Ar lines are decreased significantly with the increase in the O_2 content, which indicates the decrease in n_e . Compared with the variation of I_g in Fig. 12, the emission intensities of Ar lines in Fig. 14 drop almost 80%, but I_g remains almost constant for oxygen contents from 0.1% to about 4%. This observation suggests that the electrons are attached with O_2 to form O_2^- ions and the drop of the electron energy. When the O_2 content is increased further, the discharge becomes weak, and the emission intensities of Ar lines and I_g decrease. The distributions of the Ar line intensities are also changed, which represent the change in T_e . The ratio of the intensities of Ar I (696.5 nm) and Ar I (912.3 nm) is also used to indicate the variation of T_e with different O_2 contents, which is shown in Fig. 15.

FIG. 13. The emission spectra of Ar DBD at 4.5 kV with 0.1% and 1% O_2 contents.

FIG. 14. The selected Ar emission lines intensities at different O₂ contents.

From Fig. 15, it can be seen that with the increase in O₂ content from 0.1% to 10%, the ratio of the intensities of Ar I (696 nm) and Ar I (912 nm) is decreased. For the threshold, the energy of the upper state of Ar I (696.5 nm) is higher than that of Ar I (912.3 nm), indicating that fewer high energy electrons are produced and T_e is decreased. This may result from the reduction of the electron mean free path with the O₂ addition, which limits the energy the electrons gain from the electric field.

The addition of O₂ can increase the plasma chemical activity by producing a range of reactive species such as O, O₃, etc. With the observation of the emission spectra of DBD, the effect of O₂

FIG. 15. The ratio of emission intensities of Ar I (696 nm) and Ar I (912 nm) at different O₂ contents.

FIG. 16. O I (777 nm) emission line intensities at different O₂ contents.

content on the O emission can be obtained. Figure 16 shows the emission spectra of O I (777.4 nm) at 4.5 kV with different oxygen contents. It can be observed that the O I (777.4 nm) emission line appears when the O₂ is added to the Ar DBD. But O I (777.4 nm) is very weak and overlapped with the nearby Ar emission. It is because the production of the upper state O (3p⁵P) of O I (777.4 nm) need higher energy than the surrounding Ar emission lines, which transmit from the upper 4p states with excitation energy around 13 eV. The production of O ground state atom from reactions (8) and (9) needs 5.1 eV and 3.6 eV, respectively,⁴⁴ and the excitation of O (3p⁵P) needs 10.7 eV. Considering the small amount of O₂ molecular and O atom, the O (3p⁵P) is mainly produced by direct electron impact dissociation excitation, which needs 15.8 eV or 14.3 eV. Therefore, the O emission is much weaker than the Ar emissions,

For the weak and overlapped O emission line, it is hard to analyze the effect of O₂ content on the O I (777.4 nm) emission intensity quantitatively. However, from Fig. 16, the tendency of the O emission at different O₂ contents can be seen, qualitatively. The addition of O₂ promotes the O I (777.4 nm) emission at first and the O I (777.4 nm) emission line intensity shows a maximum at 0.4% O₂ content. With the O₂ content further increased, the O I (777.4 nm) emission line intensity drops. It is because that the upper state O (3p⁵P) of O I (777.4 nm) is produced by direct electron impact dissociation excitation. With the increasing O₂ content, the O I (777.4 nm) emission intensity increases correspondingly. But with more O₂ addition, n_e and T_e decrease. Although there is an increase in the number density of O₂, the electrons with energies higher than the threshold for the dissociation excitation reaction

are greatly reduced, which results in the decrease in the emission intensity of the O emission.

IV. CONCLUSIONS

In this paper, an Ar DBD is driven by an ns pulsed power supply, and the influence of the pulsed voltage on the discharge uniformity was investigated using optical and electrical measurements. Using the GVSD quantitative analysis, it is found the DBD changes from a non-uniform discharge to uniform discharge (values of GVSD below 1), and then non-uniform discharges with bright filaments. The addition of O₂ causes a filament discharge with high E/n and quenches the discharge leading to higher GVSD values. The separation of the applied voltage and measured current shows that the voltage across the gap, U_g , and the discharge current, I_g , increase with the increases in the applied voltage. I_g increases faster than U_g for a higher E/n favoring ionization processes and electron migration. The discharge uniformity is sensitive to the variation in the voltage. With increasing O₂ content, U_g shows a decrease, but I_g and n_e maintain high values for O₂ content ranging from 0.1% to 4%. n_e shows a maximum value of $2.7 \times 10^{-17} \text{ m}^{-3}$ at 0.4% O₂ content. When the O₂ content exceeds 4%, a larger proportion of the applied voltage applies to the dielectric barrier rather than the gas gap, and I_g also drops significantly. The larger drop of I_g than n_e indicates the drop of drift velocity and more O₂⁺ ions generation. The emission intensities of Ar lines increase with increasing applied voltage and decrease with the increasing of O₂ content. The ratio of the emission intensities of Ar I (696.5 nm) and Ar I (912.3 nm) can be used to indicate the electron temperature, T_e , of the Ar DBD, which decreases with increasing O₂ content, while it becomes higher with the voltage rising from 3.1 kV to 4.25 kV and then becomes saturated. The O I (777.4 nm) emission intensity shows a maximum at 0.4% O₂ content. The experimental results contribute to the improvement of discharge uniformity and plasma chemical reactivity, which are important for a diverse range of practical applications.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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