Comparative Study between He/O₂ and Ar/O₂ Plasma in Atmospheric-Pressure Glow Dielectric Barrier Discharge

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Abstract: The discharge characteristics between He/O₂ and Ar/O₂ plasmas generated by atmospheric-pressure DBD are compared by electrical measurements and numerical simulation. The He/O₂ plasma shows a glow mode characterized by high-density and low-temperature electrons, while the Ar/O₂ plasma exhibits a Townsend mode featured by moderate-density and high-temperature electrons with relatively-high plasma power density.

Keywords: DBD, He/O₂, Ar/O₂, electrical measurement, simulation, Glow, Townsend

1. Introduction

Over the decades, atmospheric-pressure dielectric barrier discharge (DBD) has been focused on their potential applications to surface treatment and material processing by substituting the low-pressure glow discharge. For the extensive use of DBD to meet industrial needs, it is essential to generate plasmas more efficiently and to determine operating conditions, like applying voltage and frequency, and discharge gas composition. One of solutions for providing economical DBD processing is to replace expensive helium, which has been widely used in atmospheric-pressure nonequilibrium plasma generation, with cheap discharge gases like argon.

The aim of this paper is to verify the possibility of substituting helium with argon through experimental and numerical work comparing the discharge characteristics and efficiencies between the He/O₂ and Ar/O₂ plasmas generated by the atmospheric DBD.

2. Experiments and numerical simulation

A parallel plate DBD reactor considered is composed of two planar electrodes, of which inner surfaces are covered with 1-mm-thick dielectric material (Al₂O₃) having a 3-mm discharge gap (dgap). The discharge power is supplied to the reactor with voltages of 4 ~ 7 kV at 20 kHz of a sinusoidal wave. The discharge gas is introduced inside it in the form of a mixture of plasma-forming gas (He or Ar) of 10 liter/min and O₂ additive of 0 ~ 2 volume-%.

Electrical measurements are made to find discharge voltages and currents. By assuming that the accumulated charges $Q_b$ in the barrier and plasma are equal to the stored charges $Q_x$ in a dummy capacitor, $C_x$ (800 pF), as shown in Fig. 1, the discharge voltage $V_b(t)$ between the barrier surfaces and the bulk electric field $E(t)$ in the discharge gap can be obtained as follows:

$$V_b(t) = V_p(t) + V_e(t)[(C_e - 2C_x)/C_e]$$

$$E(t) = V_e(t)/d_{gap}.$$  

From the calculated electric field $E$, the electron diffusion coefficient $D_e$, mobility $\mu_e$, and drift velocity $v_e$ can be estimated by solving Boltzmann equation for $f(E/n)$:\

$$D_e, \mu_e = f(E/n), \quad v_e(t) = \mu_e E(t).$$

The average electron density $n_e$ and electron temperature $T_e$ are estimated from the discharge current density $j(t)$ and Einstein’s relation:\

$$n_e(t) = j(t)/\mu_e E(t), \quad kT_e(t)/e = D_e(t)/\mu_e.$$  

In addition, from the measured discharge voltage and current, the electric power deposited in plasma ($P_p$) and barrier ($P_b$) can be derived as:

$$P_p = \frac{1}{l} \int V_p(t) \cdot j(t) dt,$$

$$P_b = \frac{1}{l} \int V_b(t) \cdot j(t) dt,$$

and the power delivery efficiency ($\eta$) is estimated as

$$\eta = P_p/(P_p + P_b) = P_p/P_r.$$  

For validation of the measured results and understanding of plasma characteristics in detail, a one-dimensional numerical code has been developed to solve continuity equations for species densities and Poisson's equation for electric field. Surface charge accumulation over the barriers is also included. As reaction processes in the DBD plasmas, 45 reactions among He/O₂-related 16 species and 33 reactions among Ar/O₂-related 11 species are assumed, respectively.

Fig. 1 Equivalent circuit for discharge voltage ($V_p$) and barrier voltage ($V_b$) estimations of a DBD reactor.
3. Results and discussion

Figs. 2 and 4 are the measured electrical characteristics and the estimated electron density and temperature in the He/O₂ and Ar/O₂ discharges, respectively. Figs. 3 and 5 are the calculated spatio-temporal density distributions of electron and helium meta-stable species He* in the DBD reactor for the two plasmas, respectively, during a single cycle of the applying voltage, \( V_a(t) = V_0 \sin(2\pi ft) \). Measured and estimated results are validated with simulation ones in fairly good agreement.

The unique characteristics of the He/O₂ DBD found in Fig. 2(a) exhibit a phase difference between the applying and discharge voltages as well as the presence of an “active period” during which the discharge occurs alternatively in rising and falling phases of the applying voltage due to the surface charge accumulation on the barrier surface. In response to the applying voltage cycle, the active periods appear repetitively and are synchronized with the variations of discharge current, electron density and temperature.

When the applying voltage \( V_0 \) are changed from 4 to 7 kV, the maximum values of discharge voltage and current rise from 1.5 to 2.2 kV and from 0.22 to 0.35 mA/cm², respectively. The maximum values of electron density and temperature increase with the applying voltage in ranges of \( 2.5 \times 10^8 \) to \( 5.4 \times 10^9 \) cm⁻³ and 3.9 to 4.5 eV, respectively. Especially, at the high voltage condition, the discharge shows a periodic enhanced current peak that is a typical current shape of the glow discharge mode at atmospheric-pressure [3].

The simulation results in Fig. 3 apparently show the glow characteristics for the spatio-temporal density distributions of electron and He* within the discharge gap according to a cycle of the sinusoidal applying voltage variations. As the applying voltage increases during a \( 0-0.5\pi \) phase, electrons are rapidly accelerated and are multiplied from the anode barrier (\( y = 0.0 \)) to the cathode barrier (\( x = 0.3 \)), and a cathode fall region is created near the cathode barrier with higher He* density during the active period. After the applying voltage reaches its highest value, the electric field between the two barriers decreases during the \( 0.5\pi-1.0\pi \) phase, and the densities of electron and He* are also lowered in the whole discharge region. From the \( 1.0\pi-2.0\pi \) phase, the roles of cathode and anode are exchanged with a reversed applying voltage, and the discharge characteristics repeat the similar behavior to the previous ones described for the \( 0-1.0\pi \) phase.

At low O₂ additive (0.1%), large electric field distortions are found with increased electron density near the cathode barrier region. But at high O₂ addition (1.0 %), the overall densities of electron and He* and the discharge current decrease with decreasing \( n_e \) and \( T_e \) due to the quenching of He* by the additive O₂ that plays important role in supplying sufficient electrons and sustaining glow discharge characteristics.
On the other hand, in the measurements for the Ar/O\(_2\) discharge shown in Fig. 4, the discharge voltage exhibits higher values (2.0 ~ 2.6 kV) compared to the He/O\(_2\) discharge. The variation range of maximum electron density is 5.8x10\(^8\) ~ 7.4x10\(^8\) cm\(^{-3}\) smaller than that for the He/O\(_2\) discharge, but the maximum electron temperature range of 7.26 ~ 7.92 eV doubles that for the He/O\(_2\) discharge. In contrast to the He/O\(_2\) discharge with rapid changes of discharge voltage and current, the Ar/O\(_2\) plasma makes moderate changes in voltage and current variations that are featured in the Townsend-mode discharge.

In the spatio-temporal distributions in Fig. 5, the electron density exhibits moderate increase from the cathode barrier to the anode barrier in the Ar/O\(_2\) discharge region, and its overall values are smaller than those of the He/O\(_2\) discharge. The density distribution of argon meta-stable species Ar\(^*\) and the electric field intensity also show a similar trend of monotonous changes to the electron density variations. From these results, it is concluded that a Townsend mode characterizes the Ar/O\(_2\) DBD. When more O\(_2\) additive is added, the argon meta-stable species Ar\(^*\), which is responsible for providing additional electrons to the discharge just like He\(^*\) in the He/O\(_2\) plasma, is quenched by the O\(_2\) additive, and accordingly electron and Ar\(^*\) densities and electric field intensity decrease.

Fig. 6 represents the comparative plot of average electron temperature versus density between the He/O\(_2\) and Ar/O\(_2\) discharges. Both the plasmas show that \(T_e\) is proportion to \(n_e\) which increases with elevating applying voltage or reducing O\(_2\) additive. The He/O\(_2\) discharge turns out to have the high-density (2.4x10\(^8\) ~ 2.9x10\(^9\) cm\(^{-3}\)) and low-temperature (3.04 ~ 3.95 eV) electrons, while the Ar/O\(_2\) discharge does the moderate-density (1.6x10\(^8\) ~ 1.0x10\(^9\) cm\(^{-3}\)) and high-temperature (6.14 ~ 7.81 eV).

Fig. 7 shows the comparison of the total (\(P_t\)) and plasma (\(P_p\)) power densities between the He/O\(_2\) and Ar/O\(_2\) plasmas according to the variations of applying voltage and oxygen additive fraction. In both cases, \(P_t\) and \(P_p\)
increase with $V_a$, and $P_p$ also becomes larger when more O$_2$ is added. At the same level of applying voltage to both discharges, $P_t$ for them are almost comparable, but the Ar/O$_2$ plasma exhibits a bit higher $P_p$ than the He/O$_2$ plasma ($0.07 < P_p < 0.23$ W/cm$^2$ for He/O$_2$, $0.11 < P_p < 0.31$ W/cm$^2$ for Ar/O$_2$). The high plasma power density in the Ar/O$_2$ discharge is attributed to the high $V_p$, and therefore the Ar/O$_2$ plasma yields higher power delivery efficiencies up to 0.67 while the He/O$_2$ efficiency ranges from 0.32 to 0.53, as appeared in Fig. 8. Even though the Ar/O$_2$ plasma has the high power delivery efficiency, it accompanies an excessive rise (over 375 K) in plasma gas temperature by Joule heating loss.

4. Conclusion

To compare the distinct plasma characteristics between the He/O$_2$ and Ar/O$_2$ DBDs, electrical measurements and numerical simulation are carried out. The He/O$_2$ plasma shows a stable glow discharge mode with high electron density and low electron temperature, while the Ar/O$_2$ plasma exhibits a Townsend discharge mode with moderate electron density and high electron temperature.

As a substitute for expensive helium used in the atmospheric DBD, the use of argon is affordable for atmospheric-pressure plasma generation with a high plasma power density and an enhanced power delivery efficiency even though excessive gas heating is accompanied. Further comparative study is necessary to understand not only the power efficiency but also the generation efficiency of reactive species produced in the discharge for their practical applications.

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References