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The back-diffusion effect of air on the discharge characteristics of atmospheric-pressure radio-frequency glow discharges using bare metal electrodes

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Abstract
Radio-frequency (RF), atmospheric-pressure glow discharge (APGD) plasmas using bare metal electrodes have promising prospects in the fields of plasma-aided etching, deposition, surface treatment, disinfection, sterilization, etc. In this paper, the discharge characteristics, including the breakdown voltage and the discharge voltage for sustaining a stable and uniform $\alpha$ mode discharge of the RF APGD plasmas are presented. The experiments are conducted by placing the home-made planar-type plasma generator in ambient and in a vacuum chamber, respectively, with helium as the primary plasma-forming gas. When the discharge processes occur in ambient, particularly for the lower plasma-working gas flow rates, the experimental measurements show that it is the back-diffusion effect of air in atmosphere, instead of the flow rate of the gas, that results in the obvious decrease in the breakdown voltage with increasing plasma-working gas flow rate. Further studies on the discharge characteristics, e.g. the luminous structures, the concentrations and distributions of chemically active species in plasmas, with different plasma-working gases or gas mixtures need to be conducted in future work.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
Non-thermal plasmas generated at different pressure levels, from low pressure to atmospheric pressure or even higher pressure levels, have been widely used in industrial, biomedical and military fields and even extended to national securities because of their unparalleled capabilities for production of chemically active species with low gas temperatures while maintaining high uniform reaction rates over relatively large areas [1–3]. For example, they can be used for plasma-aided etching of a variety of substrates, such as polyimide, silica, tantalum and tungsten, in the microelectronic industry [4, 5], for plasma-enhanced chemical vapour deposition of silicon nitride or silicon dioxide films [6–8], for decontamination of chemical and biological warfare (CBW) agents [9–14], for decontamination and decommissioning of radioactive and chemical waste [15], graffiti removal, car wash [16], and so on. Although low-pressure plasmas have found wide applications in the last few decades, there are some drawbacks...
for such kinds of plasma sources operating at reduced pressures related to the expensive and complicated vacuum system which results in high capital costs, the limitations imposed on the sizes of the treated objects, the complex robotic assemblies used to shuttle materials in and out of vacuum chamber, etc [1]. In recent years, different kinds of novel atmospheric-pressure glow discharge (APGD) plasma sources have been developed, such as the dielectric barrier discharge (DBD) plasma [9,12], the cold arc-plasma jet [17], the one atmosphere uniform glow discharge plasma (OAU/GDP) [18], the surface-wave discharge [19], the microhollow cathode discharge [20] and the radio-frequency atmospheric-pressure plasma jet [1–8,10,15,21–25]. Due to the removal of the expensive and complicated vacuum system at atmospheric pressure, the APGD plasmas show bright prospects of potentially replacing low-pressure discharge devices for some existing applications and of creating new applications in future [1–3].

Among different kinds of APGD plasma sources, the APGD plasmas using bare metal electrodes driven by radio-frequency (RF) power supply developed in recent years have attracted much attention of researchers globally [1–8,10,15,21–25]. Besides the preceding stated advantages compared with traditional low-pressure glow discharges due to the removal of the vacuum system, RF APGD plasma sources using bare metal electrodes also show outstanding features compared with atmospheric-pressure DBDs. Resulting from the elimination of the dielectric(s) covered on the electrodes or placed between electrodes in atmospheric-pressure DBDs, the breakdown voltage of the gas can be reduced significantly and a more homogeneous glow discharge can be produced in RF APGDs using bare metal electrodes [1]. With the foregoing advantages, it can be expected that the RF APGD plasma sources using bare metal electrodes would be utilized in a wider range, such as etching, deposition, decontamination of chemical and biological warfare agents, food safety [4,8,11,22,26].

Although many papers have been published concerning the discharge characteristics of RF APGD plasmas using bare metal electrodes [2,21–25,27–30], there are some problems which need to be solved for speeding up the actual applications of such kinds of APGD plasmas in a variety of fields. For example, at the present time, only helium or argon can be employed as the primary plasma-working gas, to which a for example, at the present time, only helium or argon can be

In addition, studies concerning the influences of the plasma-working gas flow rate on the discharge characteristics in a RF APGD plasma using bare metal electrodes are also very limited [27]. In previously published papers, influences of the plasma-forming gas flow rate on the discharge characteristics in atmospheric-pressure DBDs have been investigated [32–35]. It was pointed out that the introduction of the gas flow removed the metastable quenchers (gaseous products from dielectrics and electrode surfaces), which was helpful for obtaining glow discharges [32,33]. Dong et al [35] indicated that an optimum flow rate existed corresponding to the lowest breakdown voltage for an atmospheric-pressure DBD plasma with two liquid electrodes. But in [36], experimental results for an atmospheric-pressure DBD plasma showed that the plasma-working gas flow rate had no influence on the establishment of the ionization equilibrium, but could influence the emission intensities of plasmas. Concerning the RF APGD plasma using bare metal electrodes, Wang et al [27] investigated the relationship between the gas flow rate and the reflected power, RF current and discharge voltage with a coaxial-type plasma torch. The results in [27] indicated that the RF current and discharge voltage did not vary with the gas flow rate monotonically, but there existed an optimum value of the gas flow rate which corresponded to the largest root-mean-square (rms) current, least rms voltage as well as the optimum reflected power for a helium RF APGD plasma using bare metal electrodes. But the experiments in [27] were carried out in ambient without considering the influences of air on the discharge processes. In this study, the back-diffusion effect of air on the discharge characteristics of RF APGD plasmas using bare metal electrodes is studied by placing the home-made planar-type plasma generator in ambient and in a vacuum chamber, respectively, while keeping other operation parameters, e.g. the gas pressure, the gap spacing between electrodes, etc. unchanged. The experimental results show that it is the back-diffusion effect, instead of the plasma-working gas flow rate, that influences the discharge characteristics of RF APGD plasmas using bare metal electrodes when the discharge processes occur in ambient.

2. Experimental setup

A schematic diagram of the experimental setup is shown in figure 1(a). The plasma generator, as shown in figure 1(b), is composed of two 5 × 8 cm² planar, water-cooled, bare copper electrodes, i.e. the RF (13.56 MHz) powered bottom electrode and the grounded top electrode. The PTFE spacers are used to seal the plasma generator on both sides (shaded in figure 1(b)) and adjust the distance between the electrodes. The plasma-forming gas (99.99% or better for helium and/or air from a compressor) is admitted into the plasma generator from the left side through a circular hole, flows through a cross-section-variable channel (gradually converged in the direction normal to the electrode surface and enlarged in the direction parallel to the electrode surface), ionized between electrodes and flows out of the generator from the right side forming a non-thermal plasma jet. For improving the uniformity of the flow field before the plasma-working gas enters into the discharge region, a 1 mm thick PTFE multi-hole sheet, as shown in figure 1(c), is located between the discharge region and the cold
gas entrance (figure 1(b)). The rms values of the current ($I_{\text{rms}}$) and voltage ($V_{\text{rms}}$) and the current–voltage phase difference ($\theta$) are measured using a current probe (Tektronix TCP202) and a high voltage probe (Tektronix P5100) and recorded on a digital oscilloscope (Tektronix TDS3034B). Thus, the RF power input can be expressed as $P_{\text{in}} = V_{\text{rms}} \cdot I_{\text{rms}} \cdot \cos \theta$. The discharge images are taken by a digital camera (Fujifilm S5500).

In this study, a vacuum chamber with a two-stage vacuum pump system is employed to examine the back-diffusion effect of air, i.e. the planar-type plasma generator can work either in ambient or in the vacuum chamber with similar operation conditions. When the plasma generator is placed in the vacuum chamber, pure helium atmosphere can be achieved, which eliminates the back-diffusion effect of air.

The light emission from the discharge region is measured when the discharge process occurs in ambient. As shown in figure 1(a), the collection optics for the emission measurement consists of a fibre, a monochromator, a photomultiplier tube (PMT) and a personal computer. One side of the fibre is located between the electrodes and at the exit of the plasma generator. The optical spectrometer is a 300 mm focal length monochromator (WDG30 made in China) equipped with 1200 lines mm$^{-1}$ gratings and coupled to the PMT. The system works with wavelengths ranging from 300 to 900 nm. The measured data are recorded using a personal computer.

3. Experimental results

3.1. Glow discharges with helium in ambient and in a vacuum chamber

In this study, the uniform glow discharges with helium can operate either in ambient or in a vacuum chamber with the same gas pressure (1.0 atm). The relationship between the breakdown voltage ($V_b$, the minimum voltage to ignite the discharge) and the gas flow rate ($Q_{\text{He}}$) with different gap spacings ($d$) is shown in figure 2. It can be seen from figure 2 that (1) for the discharge processes occurring in ambient, the breakdown voltage is sensitive to the helium flow rate when the flow rate is small (e.g. less than 5.0 slpm), while at larger helium flow rates, the corresponding breakdown voltages are nearly constant, (2) keeping the operation parameters (e.g. the gas pressure, the gap spacing between electrodes) unchanged, but placing the plasma generator in the vacuum chamber, the measured gas breakdown voltages are almost the same in a large helium flow rate range ($Q_{\text{He}} = 0.6 \sim 82.6$ slpm), with the averaged values of the breakdown voltages $131 \pm 3$ V and $192 \pm 5$ V for $d = 1.55$ and 2.48 mm, respectively, (3) when the helium flow rate is larger than 5.0 slpm, the measured breakdown voltages for the cases with the plasma generator placed in ambient are almost the same as those when the
plasma generator is located in the vacuum chamber with the same gap spacing and gas pressure. Thus, in our opinion, the reason for the large discrepancy between the breakdown voltages corresponding to the discharge processes in ambient and in the vacuum chamber for lower helium flow rates is the back-diffusion effect of air when the discharge processes occur in ambient.

The back-diffusion effect also exists in the stable α mode discharge plasmas. Keeping the gap spacing to be constant \( d = 1.55 \text{ mm or 2.48 mm} \) and locating the plasma generator in ambient and in the vacuum chamber, respectively, it can be seen from figure 3 that after ignition (1) for the cases with discharging in ambient, the discharge voltages \( V_d \), the voltage to maintain the discharge) obviously decrease with increasing helium flow rate at first and then tend to be constant after the flow rate reaches a larger value (\( \sim 10.0 \text{ slpm} \)) and the corresponding discharge voltages are 155±3 V and 205±4 V with the RF power input \( P_{in} = 65 \pm 2 \text{ W and 153} \pm 2 \text{ W for } d = 1.55 \text{ mm and 2.48 mm, respectively} \)

while for the cases with discharging in the vacuum chamber, the variations of the discharge voltages with the increase in the helium flow rate are much smaller (\( \sim 2.5\% \)), which correspond to the discharge voltages 150 ± 3 V and 200 ± 3 V with the power input \( P_{in} = 58 \pm 2 \text{ W and 122} \pm 3 \text{ W for } d = 1.55 \text{ mm and 2.48 mm, respectively} \)

For verifying the experimental observations and the possible explanations stated above, glow discharges with helium–air mixture as the plasma-forming gas operated in the vacuum chamber with \( d = 1.55 \text{ and 2.48 mm} \) are conducted in this section. The variations of the breakdown voltage with different concentrations of air \( \chi = Q_{air}/(Q_{tot} + Q_{he}) \) and with the constant helium flow rate \( Q_{he} = 15.0 \text{ slpm} \) at atmospheric pressure are presented in figure 4. Because the flow rate of helium is much larger than that of air, the total gas flow rate can be regarded as constant, i.e. \( Q_{tot} = Q_{he} + Q_{air} \approx Q_{he} \).

3.2. Glow discharges with helium–air mixtures in a vacuum chamber

For verifying the experimental observations and the possible explanations stated above, glow discharges with helium–air mixture as the plasma-forming gas operated in the vacuum chamber with \( d = 1.55 \text{ and 2.48 mm} \) are conducted in this section. The variations of the breakdown voltage with different concentrations of air \( \chi = Q_{air}/(Q_{tot} + Q_{he}) \) and with the constant helium flow rate \( Q_{he} = 15.0 \text{ slpm} \) at atmospheric pressure are presented in figure 4. Because the flow rate of helium is much larger than that of air, the total gas flow rate can be regarded as constant, i.e. \( Q_{tot} = Q_{he} + Q_{air} \approx Q_{he} \).

In figure 4, air is pre-mixed with helium and the helium–air mixture is admitted into the plasma generator to form a stable α mode discharge between the water-cooled bare copper electrodes under the applied RF electric field. With the increase in the air concentration, the breakdown voltage increases greatly and the γ mode discharge may appear when the value of \( \chi \) reaches an upper level, e.g. a γ mode discharge appears at \( \chi \approx 1.2\% \) for the case of \( d = 2.48 \text{ mm} \) as shown in figure 4.
Photographs of the discharges with different air concentrations operated in the vacuum chamber for $d = 1.55$ mm, $Q_{He} = 30.0$ slpm; (a) $Q_{Air} = 0.3$ slpm and (b) $Q_{Air} = 0.0$ slpm.

Relationships between the breakdown voltages and the helium flow rates at constant values of $\chi$ for the discharges operated in the vacuum chamber with $d = 1.55$ mm. In figure 4, the measurements are repeated three times, and the maximum standard deviation of the measured voltages is 7.6 V. The corresponding photographs of the discharges in the $\alpha$ mode with $Q_{Air} = 0.3$ slpm and 0.0 slpm and $d = 1.55$ mm are shown in figures 5(a) and (b) at the RF power input $P_{in} = 80$ W and 77 W and with discharge voltage $V_d = 227$ V and 145 V, respectively, which also shows that it is air that results in the significant change in the discharge characteristics, as well as the colours, of the plasmas.

The relationships between the breakdown voltages and the helium flow rates at constant values of $\chi$ are shown in figure 6 for the discharges operated in the vacuum chamber with $d = 1.55$ mm. In figure 6, the experiments are repeated three times for each case, and the maximum standard deviation of the measured breakdown voltages is 6.2 V. It can be seen from figure 6, that at the same value of $\chi$, the breakdown voltage of the gas is nearly constant which is independent of the total gas flow rate.

Since the gas breakdown voltage is unchanged for the given values of the air concentration ($\chi$) and the gap spacing ($d$) according to the experimental results presented in figure 6, the relationship between the air concentration ($\chi$) and the helium flow rate ($Q_{He}$) for the discharges operated in ambient can be derived from figures 2 and 4 for a given gap spacing by finding the values of $Q_{He}$ and $\chi$ corresponding to the same value of $V_d$ in figures 2 and 4, respectively, as shown in figure 7. It can be seen from figures 2, 3 and 7 that for the discharge processes in ambient: (1) although for the cases studied in this paper, the fraction of air back-diffused into helium is very small, i.e. $\chi$ is usually less than 1.0%, the back-diffusion effect of air on the gas discharge characteristics is significant, which leads to an obvious increase in the breakdown voltage, a factor about 2, as well as in the discharge voltage; (2) under the same helium flow rate, the air concentration in the discharge region for the case with larger gap spacing is a little higher than that for the case with smaller gap spacing.

### 3.3. Spectroscopic measurements of helium discharge in ambient air

The emission spectra of the $\alpha$ mode discharge with helium in ambient are measured using the optical spectrometer system as described in section 2. Since one of the major components of air is nitrogen, we chose the molecular lines from the second positive systems of $N_2$ ($C^3\Pi_u - B^3\Pi_g$) at 375.4 and 380.4 nm and from the first negative system of $N_2^+$ ($B^2\Sigma_u^+ - X^2\Sigma_g^+$) near 391.4 nm [37] to reveal the back diffusion effect of air on the purity of helium in the discharge region. The optical emission spectra in the range 372–396 nm are shown in figure 8 for different helium flow rates with $d = 1.55$ mm and $P_{in} = 120$ W. It can be seen from figure 8 that with the increase in the helium flow rate, the emission intensities of the lines 375.4, 380.4 and 391.4 nm obviously decrease, which indicates that the contents of air back-diffused into the discharge region decrease with increasing helium flow rate when the discharge process occurs in ambient.
4. Discussions

As discussed in section 1, although the experimental results presented by Wang et al [27] indicated that there was an optimum value of the gas flow rate which corresponded to the largest rms current, least rms voltage and the optimum reflected power for a helium RF APGD plasma using bare metal electrodes, the influence of air on the discharge process was not considered carefully since the coaxial-type plasma torch worked in ambient. In [2, 21] the discharge assembly was placed in a vacuum chamber in order to minimize the impurities in the discharge. As is known, for igniting a plasma, the breakdown voltage \( V_b \) depends on the electrode spacing \( d \) and the pressure \( P \) as follows \([1, 38]\):

\[
V_b = \frac{B(P \cdot d)}{\ln[A(P \cdot d)] - \ln[1 + 1/\gamma_{se}]}\]  

(1)

where, \( A \) and \( B \) are constants found experimentally and \( \gamma_{se} \) is the secondary electron emission coefficient of the electrode. In this study, water-cooled copper electrodes are employed throughout the experiments. Therefore, it can be assumed that the value of \( \gamma_{se} \) is a constant. Although gas flowing can cause a pressure drop in the flow direction, the influence of the rather low flow rate in this study on the pressure distributions in the discharge region between electrodes is still negligible. For the steady, two-dimensional viscous flows between two infinitely long parallel plates, the relationship between the velocity component \( u \) in the flow direction \( x \) and the pressure gradient \( dP/dx \) can be expressed as \([39]\):

\[
u = -\frac{(d/2)^2}{2\mu} \frac{dP}{dx} \left[ 1 - \left(\frac{2y}{d}\right)^{2}\right], \]  

(2)

where \( P \) is the gas pressure, \( \mu \) is the viscosity of the feeding gas \( (1.99 \times 10^{-5} \text{ Pa s}) \) taken in this study for helium at room temperature \([40]\) and \( y \) is the distance away from the symmetric plane of the gap in the normal direction of the electrodes. According to equation (2), we can obtain the relationship \( u_{\text{max}} = 1.5\bar{u} \approx 3 \bar{u} \) approximately, where \( u_{\text{max}} \) and \( \bar{u} \) represent the maximum value and the averaged value of the velocity component \( u \). In this study, the maximum volumetric flow rate is 82.6 slpm; thus, the corresponding pressure drop along the flow direction is \( \Delta P \approx 55 Pa \) for the case with gap spacing \( d = 1.55 \text{ mm} \) and the length in the flow direction \( X = 50.0 \text{ mm} \), which is much smaller than the atmospheric pressure \( (10 Pa) \). Therefore, based on the foregoing discussions, the values of \( (P \cdot d) \) can be assumed constant throughout the experiments by varying the helium flow rate if the gap spacing is kept constant. Because the values of \( A \) and \( B \) are also unchanged for a certain kind of gas \([38]\), it can be concluded that for a certain kind of gas, the flow rate of the plasma-forming gas has no influence on the breakdown voltage provided that the operation conditions, including the gap spacing between the electrodes and the electrode materials, are kept constant during experiments, which is consistent with the experimental measurements presented in section 3.

Laimer et al [24] pointed out, on the one hand, that back diffusion was not a problem provided that the impurity level of the plasma-working gas was below 0.1\% even through the glow discharge occurred in ambient, while on the other hand, the reason which led to much higher values of the voltage needed for the igniting and sustaining the \( \alpha \) mode discharge in their experiments, compared with those presented by Park et al [2, 21], was due to the contamination of helium, most probably by air in the order of 1\%. Based on the preceding discussions in this section, in our opinion, the discrepancy between the measured breakdown voltages presented in [24] (with a gap spacing of 2.5 mm and the helium flow rate of 1.5 mmol s\(^{-1}\) (about 2 slpm)) and those in [2, 21] (with gap spacing varying from 1.6 to 3.2 mm operated in a vacuum chamber or operated in ambient with the helium flow rate of 50 slpm in [2] and with gap spacing varying from 1.0 to 9.7 mm operated in a vacuum chamber [21]) is very possibly due to the back-diffusion effect of air.

5. Conclusions

In the present paper, the back-diffusion effect of air on the discharge characteristics of RF APGD plasma using bare metal electrodes is studied by placing the homemade planar-type plasma generator in ambient and in the vacuum chamber, respectively, while keeping other operation parameters unchanged. The main conclusions are as follows.

(1) It is the back-diffusion effect of air, instead of the plasma-working gas flow rate, that influences the discharge characteristics (e.g. the breakdown voltage, the discharge voltage for sustaining the \( \alpha \) mode discharge plasma) of RF APGD plasmas using bare metal electrodes operated in ambient.

(2) When the glow discharge processes occur in ambient, the breakdown voltage decreases significantly with increasing helium flow rate at first and then tends to be constant when the flow rate is larger than 5.0 slpm for the cases studied in this paper.

(3) When the glow discharge processes occur in the vacuum chamber, the fluctuations of the breakdown voltages with the variations of the plasma-working gas flow rates are very small (within \( \sim 3.0\% \) in this study).

(4) Under the operation conditions employed in this study, the estimated maximum air concentration in the gas discharge region between the electrodes is about 1.0\% for a RF APGD plasma using bare metal electrodes operated in ambient with helium as the primary plasma-forming gas.

Although in this paper preliminary studies on the discharge characteristics of RF APGD plasmas using bare metal electrodes are conducted considering the back diffusion effect of air in ambient, further studies on the luminous structures and the concentrations and distributions of chemically active species in plasmas with different plasma-working gases or gas mixtures need to be conducted in future work.

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