Influences of Gas Flowing on the Features of a Helium Radio-Frequency Atmospheric-Pressure Glow Discharge

Xiao-Fei Zhang[†], Zhi-Bin Wang^{†,§}, Qiu-Yue Nie, He-Ping Li^{*}, Cheng-Yu Bao

Department of Engineering Physics, Tsinghua University, Beijing 100084, P. R. China

[§] Present address: School of Electrical Engineering and Automation, Harbin Institute of Technology, Harbin 150001, P. R. China

^{*} Correspondent author: liheping@tsinghua.edu.cn (H.-P. Li), Fax: +86-10-6278-2816

[†] These two authors contributed equally to this paper.

ABSTRACT In this paper, a zoning model is employed to investigate the influences of gas flowing on the characteristics of the high-purity helium radio-frequency atmospheric-pressure glow discharge (RF APGD) plasma. The modeling results show that the influences of the gas flowing on the plasma features in the discharge region and the jet region are different. In the discharge region, the heavy-particle temperature decreases with the increase of helium flow rate, while the variations of the electron energy and the species concentrations are less than 0.5%. In the plasma jet region, the variation of the heavy-particle temperature shows a non-monotonous form with the gas flow rate, while the species concentrations become higher within a certain distance (e.g., smaller than 2 mm away from the plasma generator exit) at a higher helium flow rate. The modeling results are also qualitatively validated by comparing with the measured gas temperatures in both the discharge region and the plasma jet region.

KEYWORDS atmospheric-pressure plasma; radio-frequency glow discharge; gas flowing; non-equilibrium feature; modeling

1. Introduction

Radio frequency atmospheric pressure glow discharge (RF APGD) plasma sources produced using a pair of water-cooled bare-metallic electrodes have attracted much attention of the researchers due to their unique features, such as low breakdown/discharge voltages, low gas temperatures, high concentrations of chemically reactive species and convenient operations in open air, especially for the application in the biomedical fields^[1-3]. Although some numerical simulation results on the features in the discharge region of the RF APGD plasmas have been published, most of the authors focused on the chemical reaction processes, while the energy exchange process between the electrons and heavy particles in the plasmas were seldom considered (e.g., Refs. [4-10]). However, for the typical RF APGDs, the RF power input usually ranges from tens to hundreds Watts^[11, 12]; and thus, the gas temperature may be significantly higher than the room temperature resulted from the frequent collisions between the electrons and heavy particles in the plasma system. Therefore, the levels of the gas temperature and the chemically reactive species concentrations are the two key factors that should be considered carefully for the plasma treatment of the heat-sensitive materials, e.g., the living bio-materials or the precision instruments^[1-3].

On the other hand, different from the low pressure glow discharge plasmas, the gas flowing always exists in the RF APGD plasmas for forming a stable plasma jet at the downstream of the plasma generator, which may, to some extent, influence the spatial distributions of the gas temperatures and the concentrations of the chemically reactive species. Recently, Hemke and his co-workers studied the characteristics of the helium-oxygen RF APGD plasmas both in the discharge region and jet region operating in a pure helium atmosphere numerically using a two-dimensional transient computer code *nonPDPSIM*^[13]. In Ref. [13], a triangular unstructured mesh with approximately 10000 nodes was employed to obtain a complete picture of the dynamics of the RF APGD plasmas including the discharge region and the downstream effluent region. Due to the very small gap spacing between the electrodes and the large volume of the plasma jet region comparing with that of the discharge region in space, as well as the very high driving frequency of the power supply compared

with the characteristic time of the gas flowing, it is a very time-consuming job to simulate both the discharge region and the jet region simultaneously based on a serial computer code.

Therefore, the purpose of this paper is to study the influences of the gas flowing on the levels of the gas temperatures and the chemically reactive species concentrations both in the discharge region and the jet region in a high-purity helium RF APGD plasma system based on a zoning model; that is to say, the spatiotemporal evolutions of the plasma parameters, including the electron and heavy-species temperatures (T_e and T_h), the electrical potential (ϕ), and the species concentrations (n_i), in the discharge region are simulated using a one-dimensional (1-D) transient fluid model, while the quasi-steady spatial distributions of T_e , T_h , n_i and the flow fields of the plasma jet region are predicted using a two-dimensional (2-D) steady fluid model based on the calculated time-averaged parameter values in the discharge region as the inlet boundary conditions. The calculated results are also compared with the measured gas temperatures in these two regions.

2. Model descriptions

In this paper, the features of the high-purity helium RF APGD plasmas produced by a planar-type plasma generator, as shown in Fig. 1(a), are simulated. As indicated in Section 1, the modeling is divided into two parts corresponding to the discharge region and the jet region as shown in Fig. 1(b), **where the electrode gap spacing is fixed at 2.4 mm, while the height and length of the plasma jet region are 5.0 mm and 20.0 mm, respectively.**

2.1. Assumptions

In this study, the high-purity helium (with 0.5 ppm nitrogen as an impurity) is used as the plasma forming gas. Thus, seven species involved in thirteen chemical reactions as listed in Table 1^[7, 14-19] are considered in the modelling, including electrons (e), helium metastables (He^* , He_2^*), helium ions (He^+ , He_2^+), nitrogen molecules (N_2) and nitrogen molecular ions (N_2^+). The number density of helium atoms (He) is assumed to be constant and determined by the ideal-gas law since its number density is much higher than those of the other species. The major assumptions used in this study are as follows: (i) The variation of the gas pressure in the discharge region and the jet region is negligible, i.e., $p=1$ atm during the whole discharge

process. (ii) In the discharge region, all the metastable species and ions are quenched or neutralized at the electrode surfaces and return back to the inter-electrode space as stable neutral species; the drift-diffusion approximation is employed for calculating the number fluxes of the charged species. (iii) In the plasma jet region, the plasma flow is in a quasi-steady, incompressible and laminar regime; the values of the mass density, viscosity, specific heat at constant pressure and thermal conductivity of the plasmas are constant; and the electric field is negligible, and thus no drift process is considered for the charged particles.

2.2. Governing equations in the discharge region

Based on the proceeding assumptions, the governing equations in the discharge region include the species continuity equation, Poisson equation, electron and heavy-particle energy conservation equations.

(i) Species continuity equation:

$$\partial n_i / \partial t + \nabla \cdot \bar{\Gamma}_i = S_i \quad (1)$$

where n_i , $\bar{\Gamma}_i$ and S_i represent the number density, number flux and the homogeneous production/destruction rate of specie i , while t is the time. The drift-diffusion approximation is used for calculating the species number flux as

$$\bar{\Gamma}_i = \text{sgn}(q_i) \mu_i n_i \bar{E} - D_i \nabla n_i \quad (2)$$

where μ_i , D_i and q_i are the mobility, diffusion coefficient and the charge number of specie i , while ‘sgn’ is the signum function. **The values of μ_i and D_i , as well as the masses of different species (m_i) appearing in Equations (4) and (6), are listed in Table 2 with the cited references [5, 20, 21]. And it should be emphasized that all the values of μ_i and D_i are evaluated with the assumption that the species transport in a helium background gas at the room temperature under atmospheric pressure ($p=1$ atm).**

(ii) Poisson equation:

$$\nabla \cdot (\epsilon_0 \nabla \phi) + e \sum_i q_i n_i = 0 \quad (3)$$

where ϕ is the electric potential, ϵ_0 is the vacuum permittivity, and e is the elementary charge.

(iii) Electron energy conservation equation:

$$\partial(n_e \varepsilon) / \partial t + \nabla \cdot (5 / 3 n_e \bar{\Gamma}_e - 5 / 3 n_e D_e \nabla \varepsilon) = -e \bar{\Gamma}_e \cdot \bar{E} - \sum_i \Delta E_i^e K_i - 3 m_e / m_{He} n_e \bar{v}_e k_B (T_e - T_h) \quad (4)$$

where ε is the electron energy, ΔE_i^e and K_i are the energy loss during the inelastic collision process and the corresponding reaction rate **which are listed in Table 1**, m_e and m_{He} are the masses of electrons and helium atoms or ions, while k_B , T_e ($=[(2\varepsilon)/(3k_B)]$) and T_h are the Boltzmann constant, electron temperature and the background helium gas temperature (or heavy-particle temperature), respectively. The three terms on the right hand side of Equation (4) represent the Joule heating, energy transfer due to inelastic collisions and elastic collisions, respectively. The momentum transfer rate between electrons and the background helium ($\bar{v}_e$) is obtained from BOLSIG^[14, 15], which can be expressed as

$$\bar{v}_e = 4.9 \times 10^{11} + 1.2 \times 10^{12} \exp(-0.019\varepsilon) - 1.5 \times 10^{12} \exp(-0.41\varepsilon) [s^{-1}] \quad (5)$$

(iv) Heavy-particle energy conservation equation:

$$\nabla \cdot (-\lambda_h \nabla T_h) = 1/t_{rf} \int_0^{t_{rf}} \left[\sum_i q_i \bar{\Gamma}_i \cdot \bar{E} + 3 m_e / m_{He} n_e \bar{v}_e k_B (T_e - T_h) \right] dt - Q / (60 \times 22.4 \times V) \cdot c_p (T_h - T_r) \cdot 1.5 \cdot f_p \quad (6)$$

where Q stands for the helium flow rate, V is the volume of the discharge region, the specific heat and thermal conductivity of **helium** are c_p [$=20.786$ J/(mol·K)]^[22] and λ_h [$=0.1513$ W/(m·K)]^[23], while t_{rf} and T_r are the RF cycle and the room temperature ($T_r=300$ K in this study), respectively. It is known from the fluid dynamics that the velocity distribution inside a rectangular duct is in a parabolic form if the flow is in a fully-developed regime; and thus, we have $f_p = (x-1.2)^2 / 1.44$. The last term on the right hand side of Equation (6) represents the influences of the gas flowing on the gas temperature distributions in the discharge region.

2.3. Governing equations in the plasma jet region

In the plasma jet region, the 2-D quasi-steady behavior of the plasmas is simulated by solving the following species continuity equation, mass, momentum, electron and heavy-particle energy conservation equations.

(i) Species continuity equation:

$$\nabla \cdot \bar{\Gamma}_i = S_i; \quad \bar{\Gamma}_i = n_i \bar{v} - D_i \nabla n_i \quad (7)$$

(ii) Mass conservation equation:

$$\nabla \cdot (\rho \vec{v}) = 0 \quad (8)$$

(iii) Momentum conservation equation:

$$\nabla \cdot (\rho \vec{v} \vec{v}) = \nabla \cdot \vec{P} \quad (9)$$

(iv) Electron energy conservation equation:

$$\nabla \cdot (5/3 \bar{\epsilon}_e - 5/3 n_e D_e \nabla \epsilon) = - \sum_i \Delta E_i^e K_i - 3 m_e / m_{He} n_e \bar{v}_e k_B (T_e - T_h) \quad (10)$$

(v) Heavy-particle energy conservation equation:

$$\nabla \cdot (\rho c_p \vec{v} T_h) = \nabla \cdot (\lambda_h \nabla T_h) + 3 m_e / m_{He} n_e \bar{v}_e k_B (T_e - T_h) \quad (11)$$

In Equations (7)~(11), ρ is the mass density of the helium plasma jet with the value of 0.18 kg/m^3 , $\vec{v}$ is the mass-averaged velocity of the plasma flow, $\vec{P}$ is the pressure tensor, while the other variables represent the same meanings as those appearing in Equations (1)~(6).

2.4. Initial and/or boundary conditions

For the modeling of the discharge region, both the initial and boundary conditions are required. It is assumed that the electron/ion number density of the working-gas is $1 \times 10^9 \text{ m}^{-3}$ before breakdown due to the cosmic ray radiation^[24]. The initial number densities of the helium metastable species are set to be zero. The initial number density of the nitrogen molecules is determined with the fixed concentration of 0.5 ppm as impurity in the high-purity helium gas. The electron energy and the gas temperature are assumed to be 1 eV and 300 K as their initial values. The flux boundary conditions are imposed for all the species number densities at the electrode surfaces. For electrons, the flux at the electrode surface is determined as a sum of the kinetically limited Maxwellian flux and the secondary electron emission flux. For ions, they are assumed to be mobility limited at the boundaries. And the number fluxes for metastable species are determined as the kinetically limited Maxwellian flux; while the number fluxes for the stable neutral species are the sum of the corresponding ion/metastable fluxes. For the electric potential, the values of zero and sinusoidal function with $f=13.56 \text{ MHz}$ as the frequency are specified on the electrode surfaces. The electron energy flux at the boundary is given as a sum of the kinetically limited energy flux and the energy of the secondary electrons, while the heavy-particle temperature equals the

temperature of the solid electrode surfaces (300 K). **The exact expressions for the boundary conditions in the discharge region are listed in Table 3, where γ_i is the secondary electron emission coefficient of species i ($\gamma_i=0.25$ for the helium ions and metastables; while $\gamma_i=0.005$ for the nitrogen ions^[7, 20]), and ε_r (=5 eV) is the secondary electron energy^[25].**

For the simulation of the plasma jet, the boundary conditions corresponding to the 2-D calculation domain [ABCDEA in Fig. 1(b)] are as follows: **(i)** At the inlet of the plasma jet (AB), the boundary conditions for the species number density, the electron and heavy-particle temperatures are specified from the calculated time-averaged values of the 1-D, transient modeling in the discharge region; a parabolic form of the velocity component in the y direction is assumed **as $v=v_{\max} (x-1.2)^2 / 1.44$, where the value of $v_{\max}$ is determined by the gas flow rate, while** a zero velocity component in the x direction **is used, i.e., $u=0$.** **(ii)** **The solid walls (BC and CD) are treated as the cold surfaces, i.e., $u=v=0$, $T_h=T_e=300$,** $n_i=0$ for e, He^+ , He_2^+ , N_2^+ , He^* and He_2^* , **while $n_{\text{N}_2}=0.5 \times 10^{-6} \times p / (k_B T_h)$.** **(iii)** Along the line AE, the symmetry condition is used for all the variables, **i.e., $\partial\varphi/\partial x=0$ ($\varphi=n_i, T_e, T_h, v$) and $u=0$.** **(iv)** **The one-way boundary conditions are employed at the downstream open boundary (DE), i.e., $\partial\varphi/\partial x=0$ ($\varphi=n_i, T_e, T_h, u, v$).**

3. Results and discussions

In this paper, a zoning model is employed to study the characteristics of the helium RF APGD plasma system by solving the governing equations based on the SIMPLE algorithm^[26] as follows: Firstly, the governing equations (1), (3), (4) and (6) are solved simultaneously with the initial conditions and the boundary conditions as listed in Table 3; and then, the governing equations (7)~(11) are solved simultaneously with the calculated time-averaged spatial distributions of the electron temperature (T_e), heavy-particle temperature (T_h) and the species concentrations (n_i) imposed on the inlet boundary (AB) and the other boundary conditions as discussed in Section 2.4.

For checking the grid independence, the 600 and 1199 non-uniform 1-D grids with the finer meshes near the electrodes are used for the simulation of the discharge region; while the $52(x) \times 52(y)$ and $104(x) \times 104(y)$ uniform meshes are employed for the plasma jet region. The simulation using the two sets of meshes are conducted for the cases of $I_d=16$ mA/cm², $Q=0$ slpm in the discharge region and $I_d=16$ mA/cm², $Q=10$ slpm in the jet region. The calculated relative discrepancies of the electron number density at the mid-plane between the electrodes and of that at 1.0-mm away from the plasma generator exit along the line AE in Fig. 1(b) are both less than 3%. Thus, for obtaining the solutions with enough spatial resolution, and simultaneously, with a high calculation efficiency, the 600 non-uniform mesh for the discharge region and the $52(x) \times 52(y)$ uniform mesh for the jet region are used in the following study.

3.1. Discharge region

With no consideration of the gas flowing ($Q=0$ slpm, here ‘slpm’ means standard liters per minute), the calculated distributions of the electron energy and the heavy-particle temperature between the electrodes are shown in Fig. 2 for the case with the discharge current density of $I_d=16$ mA/cm² and the electrode gap spacing of $L=2.4$ mm. Figure 2 shows that the distribution of the heavy-particle temperature (T_h) has a parabolic shape with a maximum value of 371 K at the mid-plane between the electrodes; while the peak value of the electron energy ($\varepsilon=5.84$ eV) exists near the electrode sheath edge where a large gradient of ε also exists. Correspondingly, the time-averaged spatial distributions of the species number densities for the electrons and He^* , He_2^* , He^+ , He_2^+ , and N_2^+ between the electrodes are presented in Fig. 3. It is seen from Fig. 3 that: (i) The time-averaged concentration distributions of all the preceding chemically reactive species are symmetric about the mid-plane between the electrodes. (ii) The major chemically reactive species in the discharge region are electrons, He_2^+ and He_2^* . (iii) The spatial distribution patterns of the concentrations for different species are very different. The numbers of the peak values for the concentrations of different species are shown to be one (e and N_2^+), two (He^* and He_2^*) or four (He^+ and He_2^+). This result is qualitatively consistent with that reported in Ref. [6].

With the increase of the helium flow rate, the variations of the electron energy (ε) and the heavy-particle temperature (T_h) at the mid-plane between the electrodes are shown in Fig. 4. The modeling results show that the influence of the gas flowing on the electron temperatures is very small (less than 0.5%); while the maximum value of T_h decreases with the increase of the helium flow rate resulting from the stronger convective heat transfer process at a higher gas flow rate. The maximum values of T_h are 367, 363, 360, 357, 354 and 351 K with the gas flow rates of $Q=5, 10, 15, 20, 25$ and 30 slpm, respectively. In this study, the rotational temperature, which is very close to T_h , is also measured based on the measurement of the optical emission spectrum in the discharge region for the helium RF APGD plasmas using a multi-channel spectroscopic diagnostic system. As illustrated in Fig. 5, the nitrogen first negative system of N_2^+ ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$), transitions (0, 0) at 391.4 nm in the spectrum is used in this study. Then, the value of T_h is determined by fitting the experimental curve with the simulated one created by Lifbase^[27]. As shown in Figs. 4 and 5, the calculated T_h (363 K) is very close to the experimentally derived value (361 K) for the case of $Q=10$ slpm. In addition, the modeling results show that the spatiotemporally averaged electron temperature (T_e) is ~ 33400 K, while the heavy-particle temperature (T_h) is ~ 360 K in the discharge region for the case of $I_d=16$ mA/cm² and $L=2.4$ mm. Therefore, we can conclude that the RF APGDs significantly deviate from the local thermodynamic equilibrium (LTE) state with $T_e/T_h \approx 93$.

The variations of the species number densities at the mid-plane between the electrodes with different gas flow rates for the case of $I_d=16$ mA/cm² and $L=2.4$ mm are presented in Fig. 6. Although the major energy exchange occurs through the electron-heavy particle elastic collision process and the heavy-particle temperature decreases from 367 to 351 K with increasing the helium flow rate up to 30 slpm, the relative change of the term $3m_e/m_{He} n_e \bar{v}_e k_B (T_e - T_h)$ itself in the electron and heavy-particle energy conservation equations is very small since the value of T_e is much larger than that of T_h . Thus, on one hand, the variation of the electron temperature is very small with the increase of the helium flow rate as shown in Fig. 4. On the other hand, since the reaction rates for most of the chemical reaction

pathways are constant or depend on the electron energy as listed in Table 1, the species number densities mainly depend on T_e , instead of T_h , which leads to the quasi-constant values of the species number densities at the mid-plane between the electrodes under different helium flow rates, as shown in Fig. 6.

3.2. Plasma jet region

In the plasma jet region, two dielectric solid sheets are used to prohibit the engulfment of the surrounding air into the plasma core region. Such a method, on one hand, simplifies the interactions between the plasma jet and the ambient air in the simulation; and on the other hand, it is helpful to control, to some extent, the gas temperature, the types and concentrations of the chemically reactive species in actual applications^[28].

The calculated isotherms for the gas temperature and the streamlines of the plasma jet in the x - y plane is shown Fig. 7 for the case of $I_d=16$ mA/cm², $L=2.4$ mm and $Q=10$ slpm. It can be seen that the gas temperature decreases gradually with the increase of the distance from the generator outlet. There exists a vortex at the corner formed by the solid sheet and the frontal surface of the generator nozzle, and a fully developed flow regime appears with $y>10$ mm as shown in Fig. 7(b).

Under the same operation condition as that in Fig. 7, the modeling results in Fig. 8 show that although all the species number densities decrease along the flow direction, their decay rates are different. In some applications (e.g., genome mutation of the microbes using the RF APGD plasma jet^[29, 30]), the treated materials are usually located within 2 mm away from the plasma generator outlet. Thus, we can see that the major chemically reactive species are electrons, He_2^* and N_2^+ whose concentrations are on the order of 10^{15} m⁻³, which qualitatively agree with the results of Schütze et al^[31]. In addition, since the species of He_2^+ decays through the reaction pathway R_{12} ($\text{He}_2^+ + \text{N}_2 \rightarrow \text{N}_2^+ + \text{He}_2^*$), while simultaneously, the species N_2^+ forms through the same chemical reaction, the decay rate of He_2^+ is large than that of N_2^+ . The variations of the number densities for the electrons, He_2^+ , N_2^+ , He^* and He_2^* with the helium flow rate at $y=2$ mm in the mid-plane between the electrodes are

provided in Fig. 9. It shows that all of the preceding species concentrations increase with increasing the helium flow rates, while the increment rates of the species number densities decrease.

The variations of the calculated electron and heavy-particle temperatures of the plasma jet with the helium flow rate at 2 mm away from the generator outlet in the symmetric plane are shown in Fig. 10. It is seen that the electron temperature is a little bit higher than the gas temperature under all of the helium flow rates; and there exists a non-monotonic trend for the gas temperature with the helium flow rate. In this study, we also use a mercury thermometer to measure the gas temperature at 2 mm downstream of the plasma generator outlet. The measured area-averaged gas temperatures of the plasma jet (**about** 4 mm² based on the geometrical size of the thermometer head) are also presented in Fig. 10 under the same operation parameters. We can see that although the flow rates corresponding to the calculated and the measured maximum gas temperatures are somewhat different, the simulated and the measured variation trends of the gas temperatures with the helium flow rate are the same. On the other hand, **a maximum relative discrepancy of 8.3% between** the calculated and the measured gas temperatures still exists in the present study. One possible reason is the low spatial resolution of the measurement using the mercury thermometer; another possible reason may be related to the mixing of surrounding air into the plasma jet since it operates in an open environment. We will conduct deeper studies in our future work.

4. Concluding remarks

The influences of the gas flowing on the characteristics of the helium RF APGD plasmas are studied numerically under the constant discharge current density of $I_d=16$ mA/cm² and gap spacing of $L=2.4$ mm. And the qualitative comparisons with the experimental measurements are also conducted. The main conclusions are as follows:

(1) In the discharge region, the calculated time-averaged parameters, including the electron and heavy-particle temperatures (T_e and T_h) and the species concentrations (n_i) are symmetric about the mid-plane between the electrodes; a higher gas flow rate leads to the decrease of T_h , while its influence on the electron energy (ε) and the species concentrations are negligible; and a significant deviation from the LTE state exists with $T_e/T_h \approx 93$.

(2) In the plasma jet region, the gas temperature (T_h) varies non-monotonously with the helium flow rate; while the higher species concentrations may be obtained within a certain distance away from the plasma generator outlet (e.g., $y \leq 2$ mm in this study) at a higher helium flow rate.

In **this study**, since the numerical modeling of the discharge region is based on the 1-D fluid model in the present study, the flowing effect of the plasma working gas is only taken into account with an effective source term added into the heavy-species energy conservation equation. This means that the convective effects of the gas flowing on the species number density distributions in the discharge region are not included in the present model. **In addition, with the employment of the zoning model, the coupling between the discharge region and the plasma jet region is neglected as an approximation.** So, it is indispensable to conduct the 2-D transient modeling of the **complete** discharge-jet region to reveal the influences of the gas flowing on the **transient and non-equilibrium** characteristics of the RF APGD **plasma sources** in the future work by developing the parallel computer codes.

Acknowledgment

This work has been supported by the National Natural Science Foundation of China (No. 10972119), the JST CREST of Japan, and Tsinghua University Initiative Scientific Research Program (2011Z01019).

References

- [1] G. E. Morfill, M. G. Kong, J. L. Zimmermann, Focus on plasma medicine, New J. Phys., 11 (2009) 115011.
- [2] M. G. Kong, G. Kroesen, G. Morfill, T. Nosenko, T. Shimizu, J. van Dijk and J. L. Zimmermann, Plasma medicine: an introductory review, New J. Phys., 11 (2009) 115012.
- [3] H.-P. Li, Z.-B. Wang, G. Nan, P.-S. Le, H. Wu, Y. Lu, L.-Y. Wang, C. Zhang, C.-Y. Bao, X.-H. Xing, Studies on the physical characteristics of the radio-frequency atmospheric-pressure glow discharge plasmas for the genome mutation of *methylosinus trichosporium*, IEEE Trans. Plasma Sci., 40 (2012) 2853-2860.
- [4] X. H. Yuan, L. L. Raja, Role of trace impurities in large-volume noble gas atmospheric-pressure glow discharges, Appl. Phys. Lett., 81 (2002) 814-816.
- [5] X. H. Yuan, L. L. Raja, Computational study of capacitively coupled high-pressure glow discharges in helium, IEEE Trans. Plasma Sci., 31 (2003) 495-503.
- [6] J. J. Shi, M. G. Kong, Mode characteristics of radio-frequency atmospheric glow discharges, IEEE Trans. Plasma Sci., 33 (2005) 624-630.
- [7] Y. Sakiyama, D. B. Graves, Nonthermal atmospheric rf plasma in one-dimensional spherical coordinates: Asymmetric sheath structure and the discharge mechanism, J. Appl. Phys., 101 (2007) 073306.
- [8] N. Balcon, G. J. M. Hagelaar, J. P. Boeuf, Numerical model of an argon atmospheric pressure rf discharge, IEEE Trans. Plasma Sci., 36 (2008) 2782-2787.
- [9] A.-J. Yang, X.-H. Wang, M.-Z. Rong, D.-X. Liu, F. Iza, M. G. Kong, 1-D fluid model of atmospheric-pressure rf He+O₂ cold plasmas: Parametric study and critical evaluation, Phys. Plasmas, 18 (2011) 113503.
- [10] Z.-B. Wang, P.-S. Le, N. Ge, Q.-Y. Nie, H.-P. Li, C.-Y. Bao, One-dimensional modeling on the asymmetric features of a radio-frequency atmospheric helium glow discharge produced using a co-axial-type plasma generator, Plasma Chem. Plasma Process., 32 (2012) 859-874.

- [11] J. Park, I. Henins, H. W. Herrmann, G. S. Selwyn, R. F. Hicks, Discharge phenomena of an atmospheric pressure radio-frequency capacitive plasma source, *J. Appl. Phys.*, 89 (2001) 20-28.
- [12] H.-P. Li, W.-T. Sun, H.-B. Wang, G. Li, C.-Y. Bao, Electrical features of radio-frequency, atmospheric-pressure, bare-metallic-electrode glow discharges, *Plasma Chem. Plasma Process.*, 27 (2007) 529-545.
- [13] T. Hemke, A. Wollny, M. Gebhardt, R. P. Brinkmann, T. Mussenbrock, Spatially resolved simulation of a radio-frequency driven micro-atmospheric pressure plasma jet and its effluent, *J. Phys. D: Appl. Phys.*, 44 (2011) 285206.
- [14] G. J. M. Hagelaar, L. C. Pitchford, Solving the Boltzmann equation to obtain electron transport coefficients and rate coefficients for fluid models, *Plasma Sources Sci. Technol.*, 14 (2005) 722-733.
- [15] W. Morgan, J. Boeuf, L. Pitchford, BOLSIG Boltzmann Solver (freeware) (1996) Kinema Software, USA-Toulouse, France. <http://www.siglokinema.com/bolsig.htm>.
- [16] R. Deloche, P. Monchicourt, M. Cheret and F. Lambert, High-pressure helium afterglow at room temperature, *Phys. Rev. A.*, 13 (1976) 1140-1176.
- [17] A. V. Phelps, Absorption studies of helium metastable atoms and molecules, *Phys. Rev.*, 99 (1955) 1307-1313.
- [18] Y. B. Golubovskii, V. A. Maiorov, J. Behnke and J. F. Behnke, Modelling of the homogeneous barrier discharge in helium at atmospheric pressure, *J Phys. D: Appl. Phys.*, 36 (2003) 39-49.
- [19] W. J. M. Brok, M. D. Bowden, J. van Dijk, J. J. A. M. van der Mullen, G. M. W. Kroesen, Numerical description of discharge characteristics of the plasma needle, *J Appl. Phys.*, 98 (2005) 013302.
- [20] **Y. P. Raizer, Gas Discharge Physics, Springer-Verlag, Berlin, 1991.**
- [21] **H. W. Ellis, R. Y. Pai, E. W. McDaniel, E. A. Mason L. A. Viehland, Transport properties of gaseous ions over a wide energy range, Atomic Data Nucl. Data Tables, 17 (1976) 177-210.**

- [22] W. M. Haynes, **CRC Handbook of Chemistry and Physics, 93rd Edition, CRC Press, 2012.**
- [23] G. F. Hewitt, G. L. Shires, T. R. Bott, **Process Heat Transfer, Behel House, 2000.**
- [24] H. Nishiyama, K. Tsuru, H. Shimizu, K. Katagiri, H. Takana, Y. Nakano, **Activated air plasma flow generated by pulsed arc discharge for combustion enhancement, Int. J. Heat Mass Trans., 52 (2009) 1778-1785.**
- [25] H. D. Hagstrum, **Auger ejection of electrons from tungsten by noble gas ions, Physical Review, 96 (1954) 325-335.**
- [26] S. V. Patankar, **Numerical Heat Transfer and Fluid Flow, Hemisphere Publishing Corporation, 1980.**
- [27] Q.-C. Guo, J.-L. Zhang, L.-Y. Liu, D.-Z. Wang, Characterization on the temperature of radio frequency argon capacitive discharge mode transition at atmospheric pressure, *Acta Phys. Sin.*, 60 (2011) 025207.
- [28] G. Li, P.-S. Le, H.-P. Li, C.-Y. Bao, Effects of the shielding cylinder and substrate on the characteristics of an argon radio-frequency atmospheric glow discharge plasma jet, *J. Appl. Phys.*, 107 (2010) 103304.
- [29] L.-Y. Wang, Z.-L. Huang, G. Li, H.-X. Zhao, X.-H. Xing, W.-T. Sun, H.-P. Li., Z.-X. Gou, C.-Y. Bao, Novel mutation breeding method for *Streptomyces avermitilis* using an atmospheric pressure glow discharge plasma, *J. Appl. Microbiol.*, 108 (2010) 851-858.
- [30] G. Li, H.-P. Li, L.-Y. Wang, S. Wang, H.-X. Zhao, W.-T. Sun, X.-H. Xing, C.-Y. Bao, Genetic effects of radio-frequency, atmospheric-pressure glow discharges with helium, *Appl. Phys. Lett.*, 92 (2008) 221504.
- [31] A. Schütze, J. Y. Jeong, S. E. Babayan, J. Park, G. S. Selwyn, R. F. Hicks, The atmospheric-pressure plasma jet: a review and comparison to other plasma sources, *IEEE Trans. Plasma Sci.*, 26 (1998) 1685-1694.

Table 1

Elementary reactions, the corresponding rate constant, and the energy loss due to the inelastic collisions[§].

No.	Reaction	Rate constant (m, molecules, s)	ΔE_i^e (eV)	Refs.
R1	$e + \text{He} \rightarrow \text{He}^* + e$	$1.3 \times 10^{-14} \varepsilon^{-1.7} \exp(-6.0 \times 10 / \varepsilon)$	19.8	7,14,15
R2	$e + \text{He} \rightarrow \text{He}^+ + 2e$	$2.1 \times 10^{-7} \varepsilon^{-3.8} \exp(-1.3 \times 10^2 / \varepsilon)$	24.6	7,14,15
R3	$e + \text{He}^* \rightarrow \text{He}^+ + 2e$	$7.1 \times 10^{-12} \varepsilon^{-0.9} \exp(-1.7 \times 10 / \varepsilon)$	4.8	7,14,15
R4	$\text{He}^* + 2\text{He} \rightarrow \text{He}_2^* + \text{He}$	2.5×10^{-46}	/	16,17
R5	$\text{He}^+ + 2\text{He} \rightarrow \text{He}_2^+ + \text{He}$	1.1×10^{-43}	/	7,18
R6	$\text{He}_2^* + M \rightarrow 2\text{He} + M$	1.0×10^4	/	7,19
R7	$2\text{He}^* \rightarrow \text{He}_2^+ + e$	1.5×10^{-15}	-17.4	16,18
R8	$2\text{He}_2^* \rightarrow \text{He}_2^+ + 2\text{He} + e$	1.5×10^{-15}	-13.7	16,18
R9	$e + \text{He}_2^+ \rightarrow \text{He}^* + \text{He}$	$8.9 \times 10^{-15} (T_e / T_h)^{-1.5}$	ε	7,18
R10	$\text{He}^* + \text{N}_2 \rightarrow \text{N}_2^+ + \text{He} + e$	5.0×10^{-17}	-4.2	7,18
R11	$\text{He}_2^* + \text{N}_2 \rightarrow \text{N}_2^+ + 2\text{He} + e$	3.0×10^{-17}	-2.5	7,18
R12	$\text{He}_2^+ + \text{N}_2 \rightarrow \text{N}_2^+ + \text{He}_2^*$	1.4×10^{-15}	/	7,18
R13	$\text{N}_2^+ + e \rightarrow \text{N}_2$	$4.8 \times 10^{-13} (T_e / T_h)^{-0.5}$	ε	7,19

[§] In this table, ε is in the unit of eV, while T_e and T_h are in the unit of Kelvin; the symbol M stands for an arbitrary heavy collision partner.

Table 2

Drift coefficients, diffusivities and masses of different species in a high-purity helium plasma.

Species	$D_i (\text{m}^2 \cdot \text{s}^{-1})$	$\mu_i (\text{m}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1})$	$m_i (\text{kg})$	Refs.
e	$9.98 \times 10^{-6} \times T_e$	1.132×10^{-1}	9.1094×10^{-31}	20
He [*]	4.116×10^{-4}	/	6.6445×10^{-27}	5
He ₂ [*]	2.029×10^{-4}	/	1.3289×10^{-26}	5
He ⁺	5.026×10^{-5}	1.482×10^{-3}	6.6445×10^{-27}	5,21
He ₂ ⁺	8.148×10^{-5}	2.403×10^{-3}	1.3289×10^{-26}	5,21
N ₂ ⁺	1.015×10^{-4}	2.993×10^{-3}	4.6508×10^{-26}	5,21
N ₂	1.075×10^{-4}	/	4.6508×10^{-26}	5

Table 3

Boundary conditions for the modeling of the discharge region[§].

Parameter	Equation
Electrons (e)	$\vec{\Gamma}_e \cdot \vec{n} = \frac{1}{4} n_e \sqrt{\frac{8k_B T_e}{\pi m_e}} - \sum_i \gamma_i (\vec{\Gamma}_i \cdot \vec{n})$
Ions (He⁺, He₂⁺, N₂⁺)	$\vec{\Gamma}_{ion} \cdot \vec{n} = \alpha_s \mu_{ion} n_{ion} \vec{E} , \quad \alpha_s = \begin{cases} 1 & (\vec{E} \cdot \vec{n} > 0) \\ 0 & (\vec{E} \cdot \vec{n} \leq 0) \end{cases}$
Metastable species (He[*], He₂[*])	$\vec{\Gamma}^* \cdot \vec{n} = \frac{1}{4} n^* \sqrt{\frac{8k_B T_h}{\pi m^*}}$
Nitrogen molecules (N₂)	$\vec{\Gamma}_{N_2} \cdot \vec{n} = -\vec{\Gamma}_{N_2^+} \cdot \vec{n}$
Electric potential (ϕ)	$\phi _{x=0}(t) = V_{\max} \sin(2\pi f t), \quad \phi _{x=L}(t) = 0$
Electron energy ($\mathcal{E}$)	$\vec{\Gamma}_e \cdot \vec{n} = \frac{5}{3} \mathcal{E} \left(\frac{1}{4} n_e \sqrt{\frac{8k_B T_e}{\pi m_e}} \right) - \frac{5}{3} \mathcal{E}_r \sum_i \gamma_i (\vec{\Gamma}_i \cdot \vec{n})$
Heavy-particle temperature (T_h)	$T_h _{x=0} = T_h _{x=L} = 300$

[§] In this table, the subscripts “e”, “ion” and “e” stand for electrons, ions and the electron energy, while the superscript “*” represents the metastable species; $\vec{n}$ is the unit vector pointed to the surface of the electrode; $V_{\max}$ is the amplitude of the applied voltage.

Figure captions

Fig. 1. Schematic diagrams of the planar-type plasma generator (a) and of the calculation domains for the modeling of the discharge region and the plasma jet region (b).

Fig. 2. Profiles of the electron energy (solid line) and the heavy-particle temperature (dashed line) between the electrodes with no consideration of the gas flowing ($Q=0$ slpm) under the discharge current density of $I_d=16$ mA/cm² and the electrode gap spacing of $L=2.4$ mm.

Fig. 3. Distributions of the species number densities between the electrodes under the same operating condition as that in Fig. 2.

Fig. 4. Variations of the electron energy and the heavy-particle temperature at the mid-plane between the electrodes with different gas flow rates ($I_d=16$ mA/cm² and $L=2.4$ mm).

Fig. 5. Experimental and simulated results for the first negative (0, 0) rotational lines of N_2^+ .

Fig. 6. Variations of the species number densities at the mid-plane between the electrodes with different gas flow rates.

Fig. 7. Calculated isotherms for the gas temperature (a), the streamlines of the plasma jet and the profile of the velocity component in the y-direction at 10.0-mm away from the generator exit (b) ($I_d=16$ mA/cm², $L=2.4$ mm and $Q=10$ slpm).

Fig. 8. Profiles of the species number densities along the flow direction in the symmetric plane ($I_d=16$ mA/cm², $L=2.4$ mm and $Q=10$ slpm).

Fig. 9. Variations of the species concentrations with the helium flow rate at $y=2$ mm in the symmetrical plane ($I_d=16$ mA/cm² and $L=2.4$ mm).

Fig. 10. Variations of the calculated electron and heavy-particle temperatures with the helium flow rate, and comparison with the measured gas temperatures of the plasma jet at 2 mm away from the generator outlet in the symmetric plane ($I_d=16$ mA/cm² and $L=2.4$ mm).

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10