Excerpt from the Proceedings of the 2011 COMSOL Conference in Boston

Simulation of an Atmospheric Pressure Direct Current Microplasma Discharge in He/N_2

Lizhu Tong[1]
[1]Kesoku Engineering System Co., Ltd.
1-9-5 Uchikanda, Chiyoda-ku, Tokyo 101-0047, Japan, tong@kesco.co.jp

Abstract: A study of an atmospheric pressure direct current microplasma discharge in He/N_2 is performed using COMSOL Multiphysics. The calculation of heat transfer is fully coupled with the plasma simulation so as to resolve the gas heating in discharges. A simple circuit model is used to decide the discharge voltage so that the current-voltage (*I-V*) characteristics are obtained. The *I-V* characteristics in He agree well with the available experimental data and it is found that with the addition of some amount of N_2 to He, N_2 is highly ionized. N_2 ions play an important role in the discharge kinetics, even if its amount is very small such as 0.002~0.02%.

Keywords: Microplasma, Atmospheric pressure, DC discharge, Gas heating.

1. Introduction

Microplasmas are characterized by their small size (characteristic dimensions, of tens to hundreds of microns) and high gas pressure (100 Torr–1 atm), yielding nonequilibrium plasmas[1]. Recently, atmospheric pressure nonequilibrium plasmas (APNEPs) have been widely studied for environmental and biological applications as well as material processing [2,3]. The atmospheric pressure direct current (dc) micro-discharge, generally across a short gas gap between metal electrodes, is one of the easy methods of generating an APNEP.

For the extensive use of dc microplasmas at atmospheric pressure to meet industrial needs, it is essential to generate plasmas more efficiently and to understand the discharge properties, such as discharge current-voltage (*I-V*) characteristics, discharge gas composition, as well as the effect of gas temperature. The dc microdischarges at atmospheric pressure have been investigated in a He flow [1,4,5]. Wang and co-workers [1] used the fluid approach to simulate a dc atmospheric pressure microdischarge in He and they found that the microdischarge resembled a macroscopic low pressure dc discharge in many respects. They also indicated that gas heating played an important role in atmospheric pressure micro-discharges, in which the peak gas temperature increased from 370 to 650 K. Choi and co-workers [4] used the particle-in-cell Monte Carlo method to obtain the electron and ion kinetics in a dc atmospheric pressure microdischarge in He. A constant gas temperature of 300 K was used in their study. Belostotskiy and co-workers [5] reported their research about the effect of gas heating on the *I-V* characteristics in dc microdischarge in He at different operating pressures (300-800 Torr). Arkhipenko and co-workers [6] studied the effect of cathode temperature on atmospheric pressure dc glow discharges in He and they found that the cathode temperature varied from 350 to 900 K for the uncooled cathodes.

Some investigations of atmospheric pressure discharges in He/N_2 mixtures have also been carried out for analytical spectrometry [3,7,8]. The addition to He of some amount of a doping gas such as N_2 is shown to have a significant influence on the emission intensity of the analytes. Martens and co-workers [3] reported their research on atmospheric pressure glow discharge in He, including 10 ppm of nitrogen, by using fluid and Monte Carlo simulations. They indicated that at atmospheric pressure the positive column region became the dominant and that the He_2^+ ions played an important role. Wang and co-workers [7] performed their measurements by adding a small amount (0.02mol%) of N_2 to the He gas feed. The measured *I-V* characteristics were presented and the maximum gas temperature was found to be about 550 K. Petrov and co-workers [8] studied the capillary surface wave discharges at atmospheric pressure in He/N_2. They indicated that the N_2 molecules were highly ionized through Penning ionization with the metastable He atoms.

However, the role of N_2 in He/N_2 micro-discharges is still not completely clear, *e.g.*, the effect on the *I-V* characteristics. In this work, different small amounts of N_2 are added to the He gas to study the effect of gas composition on dc microdischarge properties. The gas heating is calculated by solving the equation of heat transfer, coupled with the plasma simulation. The effect of cathode temperature on the *I-V* characteristics is also studied.

2. Numerical Model

The model study for atmospheric pressure discharges is not similar to low-pressure discharges. It is known that the high pressure gives rise to more collisions and also to other types of collisions (such as three-body collisions, etc) [3]. In He/N_2 mixtures, a strong interaction between He and N_2 species takes place. The high rate coefficients of Penning ionization and of charge transfer reactions are expected to result in a decrease of He metastable atom density and in an increase of N_2 ion density.

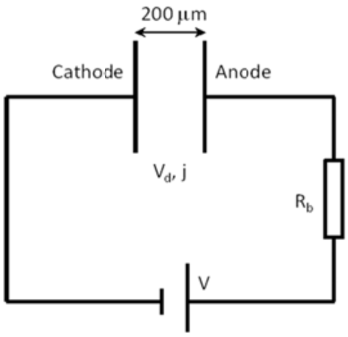

Figure 1. Schematic of a microplasma model.

The schematic of a parallel-plate micro-plasma model is shown in Fig. 1. In this work, x = 0 at cathode and x = 200 μm at anode. A dc power supplied voltage V and a ballast resistor R_b are used to generate a microdischarge. V_d is the discharge voltage and j is the discharge current density. V_d is solved by

$$V = V_d + jAR_b, \tag{1}$$

where A is the area of each electrode. In this work, A is set 0.006 cm^2, which is consistent with that used in the earlier experiment [7]. The obtained V_d is inputted into the plasma simulation as a boundary potential condition. j is obtained from the solution of the governing equations included transport of charged and neutral species, and Poisson's equation for the electric field, which is performed in the Plasma Module in COMSOL Multiphysics. The species taken into account in the model include the ions He^+, He_2^+, N_2^+, the metastable He atoms: $He(2^1S)$, $He(2^3S)$, He^*, as well as electrons. Since only a small amount of N_2 is used, the excited N_2 molecules, the dissociated N atoms, and the ions N^+, N_4^+, N_3^+, are not considered in this work [2]. The reactions among species included in the model are given in Table 1, in which the cross sections σ and reaction rate coefficients k for electron impact collisions are taken from ref. [3,9,10]. The reaction rate coefficients k among heavy species (ions and neutral species) are taken from ref. [2,3,8].

Table 1: The reactions included in the model.

No.	Reaction	σ / k ($cm^3\ s^{-1}$/ $cm^6\ s^{-1}$)
1	$e^- + He \rightarrow e^- + He(2^1S)$	$f(\varepsilon)$
2	$e^- + He \rightarrow e^- + He(2^3S)$	$f(\varepsilon)$
3	$e^- + He \rightarrow 2e^- + He^+$	$f(\varepsilon)$
4	$e^- + He(2^1S) \rightarrow 2e^- + He^+$	$f(\varepsilon)$
5	$e^- + He(2^3S) \rightarrow 2e^- + He^+$	$f(\varepsilon)$
6	$2e^- + He^+ \rightarrow e^- + He$	$3\times10^{-20}\,(T_g/T_e)^4$
7	$2e^- + He^+ \rightarrow e^- + He^*$	$3\times10^{-20}\,(T_g/T_e)^4$
8	$e^- + He_2^+ \rightarrow He + He^*$	$8.9\times10^{-9}(T_g/T_e)^{1.5}$
9	$e^- + N_2 \rightarrow 2e^- + N_2^+$	$f(\varepsilon)$
10	$e^- + N_2^+ \rightarrow N_2$	$4.8\times10^{-7}(T_g/T_e)^{0.5}$
11	$He^+ + 2He \rightarrow He_2^+ + He$	6.5×10^{-32}
12	$He(2^1S) + He \rightarrow 2He + h\nu$	6.0×10^{-15}
13	$He(2^3S) + 2He \rightarrow He_2 + He$	2.5×10^{-34}
14	$He(2^3S) + He(2^3S) \rightarrow He^+ + He + e^-$	2.9×10^{-9}
15	$He^+ + N_2 \rightarrow N_2^+ + He$	1.3×10^{-9}
16	$He_2^+ + N_2 \rightarrow N_2^+ + 2He$	1.1×10^{-9}
17	$He^+ + N_2 + He \rightarrow N_2^+ + 2He$	2.2×10^{-29}
18	$He_2^+ + N_2 + He \rightarrow N_2^+ + 3He$	1.6×10^{-29}
19	$He(2^3S) + N_2 \rightarrow N_2^+ + He + e^-$	6.3×10^{-11}
20	$He(2^1S) + N_2 \rightarrow N_2^+ + He + e^-$	1.5×10^{-10}
21	$He(2^3S) + N_2 + He \rightarrow N_2^+ + 2He + e^-$	2.9×10^{-30}

The heat transport of the gas is solved by

$$\rho c_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + Q, \tag{2}$$

where ρ is the gas density, T is the gas temperature, c_p is the specific heat, k is the thermal conductivity of the gas, and Q is the heat source term summing all the reactions shown in Table 1.

3. Results

3.1 Current-voltage (*I-V*) characteristics

Figure 2 shows the simulated the *I-V* characteristics. The boundary condition for electric field calculation is specified as $V_c = 0$ on the cathode and $V_a = V_d$ on the anode. The temperature for both electrodes is assumed to be 350 K. The secondary electron emission coefficient is decided by comparing the available experimental data [7]. It is found that at $\gamma = 0.11$ for ions He^+ and He_2^+, the calculated *I-V* characteristics for pure He microdischarges are in good agreement with the experimental data. The same γ value is used for ions N_2^+ in He/N_2 microdischarges. The secondary electron temperature are assumed to be 5 eV [1]. The calculations are performed for pure He, He/ 0.002%N_2, He/0.02%N_2 microdischarges. As seen in Fig. 2, even if a very small amount (0.002% mol) N_2 is added, the *I-V* characteristics are still affected. In the case of 0.02%mol N_2 added in He, the *I-V* characteristics greatly decrease.

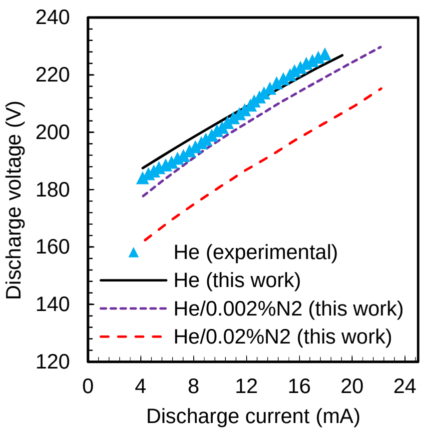

Figure 2. Current-voltage (*I-V*) characteristics for He, He/0.002%N_2, and He/0.02%N_2 microdischarges.

3.2 Discharge structure

The plasma properties for He/0.02%N_2 micro-discharges at $V = 420$ V are given in Figs. 3 and 4. As can be observed from Fig. 3, the electrical potential increases very quickly from 0 V at the cathode to about 210 V at a distance of 40 μm from the cathode. This small region has been called as the cathode dark space (CDS) region [3]. Figure 3 also indicates a large positive column fills most of the discharge region. The electron temperature profile shows a maximum of 20 eV in the cathode sheath and decreases rapidly to much smaller values (about 1~2 eV). Figure 4 shows the density profiles of electrons and ions He^+, He_2^+, and N_2^+. Although a small amount (0.02%mol) of N_2 is added to He, the generated N_2^+ ions are distributed over a large

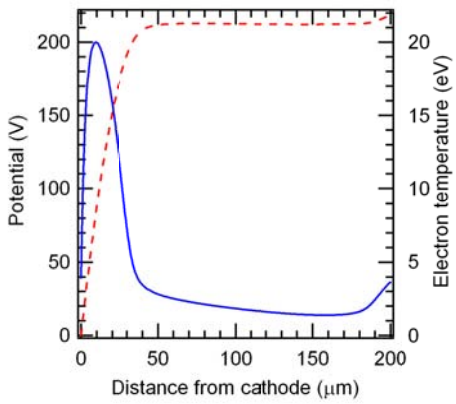

Figure 3. Electrical potential (dashed line) and electron temperature (solid line) in He/0.02%N_2 microdischarges.

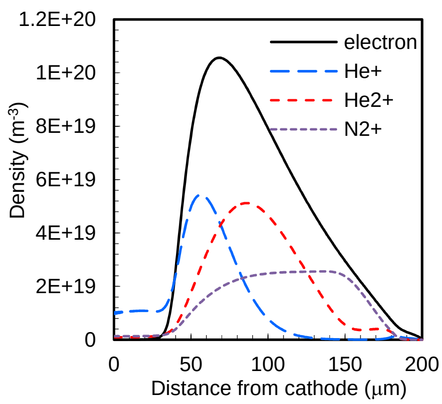

Figure 4. Density profiles for electrons and ions He^+, He_2^+, and N_2^+ in He/0.02%N_2 microdischarges.

discharge region with a maximum of about half of those of He^+ and He_2^+ ions, which could be deduced by the high Penning ionization of No. 19-21 and charge transfer reactions of No. 15-18 shown in Table 1.

3.3 Effect of cathode temperature

The simulated results for the effect of cathode temperature in He/0.02%N_2 micro-discharges are given in Figs. 5-7. The cathode temperature is assumed to be 350, 450, and 550K and the anode temperature is fixed at 350 K. The other calculation conditions are the same as section 3.2. With the increase in cathode temperature from 350 to 550 K, the peak temperatue of gas temperature increase from 580 to 700 K. It is well known that the electrical conductivity σ is strongly dependent on temperature, which can be approximated by

$$\sigma = \frac{1}{\rho_0[1+\alpha(T-T_0)]}, \qquad (3)$$

where ρ_0 is the electrical resistivity at T_0, α is the proportional coefficient, T_0 is the reference temperature, T is the gas temperature. The decrease of σ with the increase in temperature requests a higher discharge voltage to sustain the plasma discharge, as shown in Fig. 6.

Figure 7 shows the *I-V* characteristics in He/0.02%N_2 microdischarges at different cathode temperatures. With the increase in cathode temperature, the *I-V* characteristics rise up.

4. Conclusions

This paper presents the simulation results of atmospheric pressure direct current microplasma discharges in He/N_2. The fully coupled calculation of plasma discharge and heat transfer is realized. A simple circuit model is used to obtain the discharge voltage regarded as the boundary potential condition in the plasma simulation.

Results show that the present work efficiently reveals the plasma properties in He/N_2 microdischarges, especially for the effect of a small amount of N_2 added to He as well as the effect of cathode temperature on the *I-V* characteristics. The present work can provide the useful information of the use of COMSOL

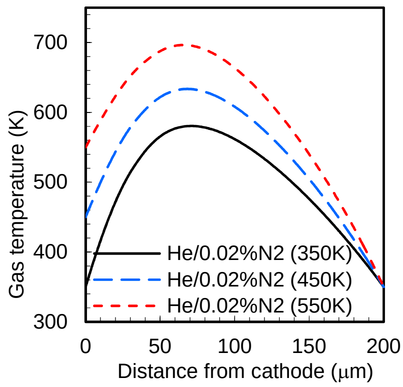

Figure 5. Gas temperature profiles in He/0.02%N_2 microdischarges at different cathode temperatures.

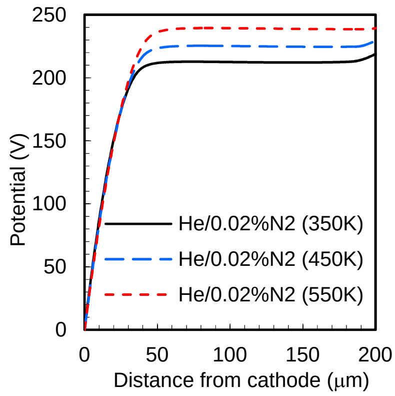

Figure 6. Electrical potential in He/0.02%N_2 micro-discharges at different cathode temperatures.

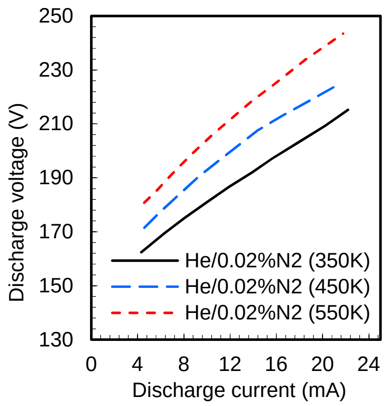

Figure 7. Current-voltage (*I-V*) characteristics in He/0.02%N_2 microdischarges at different cathode temperatures.

Multiphysics in finding the design parameters of atmospheric pressure plasma sources for surface modification.

5. References

1. Q. Wang, D.J. Economou, V.M. Donnelly, "Simulation of a direct current microplasma discharge in helium at atmospheric pressure", *J. Appl. Phys.* , **100**, 023301 (2006)

2. F. Tochikubo, N. Shirai, S. Uchida, "Simulation of atmospheric pressure direct current glow discharge along a miniature helium flow in nitrogen", *Appl. Phys. Exp.*, **4**, 056001 (2011)

3. T. Martens, D. Mihailova, J. van Dijk, A. Bogaerts, "Theoretical characterization of an atmospheric pressure glow discharge used for analytical spectrometry", *Anal. Chem.*, **81**, 9096-9108 (2009)

4. J. Choi, F. Iza, J.K. Lee, C.M. Ryo, "Electron and ion kinetics in a dc microplasma at atmospheric pressure", *IEEE Trans. Plasma Sci.*, **35**, 1274-1278 (2007)

5. S.G. Belostotskiy, V.M. Donnely, D.J. Economou, "Influence of gas heating on high pressure dc microdischarge *I-V* characteristics", *Plasma Sources Sci. Technol.*, **17**, 045018 (2008)

6. V.I. Arkhipenko, A.A. Kirillov, Ya A. Safronau, L.V. Simonchik, S.M. Zgirouski, "Influence of cathode temperature on the parameters of an atmospheric pressure dc glow discharge", *Plasma Sources Sci. Technol.*, **17**, 045017 (2008)

7. Q. Wang, I. koleva, V.M. Donnelly, D.J. Economou, "Spatially resolved diagnostics of an atmospheric pressure direct current helium microplasma", *J. Phys. D: Appl. Phys.*, **38**, 1690-1697 (2005)

8. G.M. Petrov, J.P. Matte, I. Pérès, J. Margot, T. Sadi, J. Hubert, K.C. Tran, L.L. Alves, J. Loureiro, C.M. Ferreira, V. Guerra, G. Gousset, "Numerical modeling of a He-N_2 capillary surface wave discharge at atmospheric pressure", *Plasma Chem. Plasma Proc.*, **20**, 183-207 (2000)

9. A.V. Phelps, http://jila.colorado.edu/~avp/collision_data/electronneutral/eletrans.txt

10. T. Kao, R.K. Janev, "Parametric representtation of electron impact excitation and ionization cross-sections for helium atoms", *Atomic and Plasma-Material Interaction Data for Fusion*, Vol. 3, International Atomic Energy Agency, Vienna (1992), pp. 33-29