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Numerical Study on the Gas Temperature of Microwave Discharge Rare Gas Plasmas as a Rarified Gas Dynamic System

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We report numerical simulation of gas temperature of microwave discharge plasma of rare gases by weighted-DSMC method. Our numerical simulation showed that neutral particles are heated by collisions with ions. It is found that the average temperature becomes higher for heavier rare gas discharge plasmas, which agrees with our previous experiments. It is also found that ions are accelerated by ambipolar electric field in the discharge tube. The axial dependence of the gas temperature becomes more remarkable for the heavier rare gas plasmas. The temperature gradient results in neutral depletion at the central region of the cylindrical plasma, which is also more remarkable for the heavier rare gas plasmas. It further causes the discrepancy of radial ion distribution from the Bessel distribution.

Keywords: DSMC Analysis, discharge plasmas, rare gas plasmas, neutral particles, gas temperature, rarefied flow, non-equilibrium, neutral depletion

1. Introduction

Rare gas-diluted oxygen plasmas are widely applied to various engineering processes, such as semiconductor engineering, microelectronics fabrication, innovative material preparation or surface modification. Particularly, the dissociation degree or the quantum states of the dissociated oxygen atoms are often examined in electronic engineering, since Kr-O_2 mixed plasma is frequently applied to oxidation process in microelectronics to improve electronic characteristics of prepared materials. However, there is another important parameter, gas temperature. It is one of the crucial parameters in these processes since it may affect the quality of the processed materials drastically [1]. It may play an important role to control the prepared materials by modifying plasma parameters. Up to the present time, however, the gas temperature of low-pressure discharge oxygen plasmas has not been sufficiently examined, particularly, its variation with respect to the admixture ratio of rare gas molecules.

We have experimentally studied rotational temperature of OH radicals in oxygen plasmas by optical emission spectroscopy (OES) in order to examine approximate value of its gas temperature [2]. In our experimental conditions, the discharge pressure is relatively high (~ several Torr), and as a result, the collisional equilibration is established between neutral molecules including dissociated radical fragments, although the state of non-equilibrium still exists between ions and neutral particles due to acceleration by ambipolar electric field. Consequently, we consider that the rotational temperature of OH radicals is an approximate value to the gas temperature in the rare gas-diluted oxygen plasma. We also examined the effects of rare gas admixture on its gas temperature. As a general conclusion, we found that the gas temperature decreased with He, Ne and Ar admixture, whilst it increased with Kr and Xe when the volumetric admixture ratio exceeded 80 %. As a qualitative discussion, we considered that this is mainly due to the difference in their thermal conductivity, or in the dissociation degree of oxygen molecule, etc. Now it is necessary for us to discuss the effects of rare gas admixture into oxygen discharge on its gas temperature from the quantitative viewpoints. In this report, as a first step to the foregoing subject, we conduct a numerical analysis of neutral particles as well as ions in these rare gas discharge plasmas without oxygen. In our previous experiments, we set the discharge pressure at about a few Torr with its tube radius 26 mm, and consequently, the Knudsen number becomes about 0.1, where the plasma should be considered to be a rarefied fluid. Therefore, we should solve the Boltzmann equation to understand the fluid-dynamical phenomena of these rarefied neutral particles in the plasma. We chose the DSMC simulation method, where atomic and molecular collision processes are taken into account more easily than conventional fluid dynamic simulation. In the present simulation, to understand the non-equilibrium state of neutral particles, experimentally observed values

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of electron kinetics (electron temperature and density) are treated as input parameters to analyze the neutrals. We also give the observed rotational temperature as an initial condition of the neural and ion temperatures to analyze the radial temperature distribution of neutrals. These values are summarized in Table 1 [2].

2. Numerical Procedures

First, we consider the Knudsen number of the neutral particles in the discharge tube of inner diameter 26 mm. For example, when we treat the krypton microwave discharge plasma, the discharge pressure is about 1 Torr and the corresponding mean free path of the krypton molecule becomes about 0.285 mm for room temperature, which indicates the Knudsen number $Kn = 1.1 \times 10^{-2}$. When the plasma is generated by microwave discharge, the gas temperature is considered to be much higher than the room temperature, particularly for the heavy rare gas molecules in the central region of the discharge tube. Consequently, the mean free path becomes longer and the Knudsen number becomes larger. And hence, we chose the DSMC method to simulate the transport phenomena of the neutral particles in the discharge tube as a stochastic method to solve the Boltzmann equation to describe rarefied gas dynamics.

We discretize the domain considered by a mean free path λ of the neutral molecules in the plasma as an initial gas-ion temperature, which is generally by far larger than the Debye length. We treat an axially symmetric system, and we uniformly discretize the system only radially, and not azimuthally. We set a time step Δt as one-fifth time of mean free time. We set the inner diameter of the discharge tube D = 26 mm, which was the same as the discharge tube used in our previous experiments. We treat a sufficiently long discharge tube and assume a periodic boundary condition for the longitudinal direction. Consequently, there is no volumetric charge in each control volume, and the condition of quasi neutrality $n_e = n_i$ holds everywhere in the domain considered. Then, we can eliminate the plasma oscillation, and in consequence, we are allowed to put electrons into the backgrounds of the domain. The time step may also be chosen as a practical value to simulate kinetics of neutral particles [3, 4]. We assume that the electron temperature is constant throughout the discharge area and put the experimentally observed value by double probe. Electron density is calculated to be the same as the ion density owing to the charge neutrality.

Table.1 Input parameters in the present simulation

Discharge species	Electron temperature T_e [K]	Average electron density n_{e0} [cm^{-3}]	Initial gas and ion temperature $T_g = T_i$ [K]
He	4.8×10^4	1.4×10^{12}	812
Ne	4.0×10^4	8.0×10^{11}	812
Ar	2.1×10^4	1.5×10^{12}	1160
Kr	1.8×10^4	1.7×10^{12}	1972
Xe	1.6×10^4	3.5×10^{12}	2088

The main objective of the present study is to understand the kinetics of neutral particles in the rare gas plasmas. Since the radial momentum transfer from the electrons to the neutral particles is considered to be small and rather isotropic, we neglect the collisions of neutral particles with electrons. However, the momentum transfer from the ions are essential since the motion of ions are not isotropic in the radial direction, and what is more important, the transferred momentum is a considerable amount for each collision. Then, we must calculate the ion motion in the discharge tube under the electric field in the cylindrical discharge tube, together with the motions of neutral particles. We treat only a steady state of the transport phenomena, and as a result, we adopt an ambipolar electric field as follows:

$$\phi(r) = \mathrm{Max}\left[\frac{k_B T_e}{e}\ln \mathrm{J}_0\left(\frac{\alpha r}{R}\right), -\frac{6k_B T_e}{e}\right], \qquad (1)$$

where J_0 is the Bessel function of the zero-th order, α the first zero-point of J_0, k_B the Boltzmann constant, e the elementary charge, R the inner semidiameter of the discharge tube [3, 4]. The effect of the magnetic field is sufficiently small and negligible to the movement of heavy particles.

The major problem to treat ions in the weakly ionized plasmas with the same DSMC scheme with neutral particles, lies in the respect that the ionization degree is as low as 10^{-5}, and consequently, the statistic quality of ions become very poor in comparison with that of neutrals. One of the original idea of the present study is to change the weight of ions and neutrals and to set the same number of sample particles in the domain at the initial state. That is, we apply weighted DSMC scheme to treat neutrals and ions simultaneously in the same statistical accuracy [5]. Although this scheme is frequently applied to the simulation of kinetics of gas mixture in a rarefied flow, application to the ionized gases is not often reported. We make an attempt to apply the weighted-DSMC scheme to simulate kinetics of ions and neutrals in weakly ionized gases.

Our previous experiments showed that the electron temperature was a few eV at most for the plasma to be simulated, and in consequence, we can justifiably neglect the existence of multiply charged rare gas ions [2]. We consider only singly charged ions. And the discharge pressure was set at about 1 Torr, we also neglect dimer molecules and ions [6].

As initial conditions, we set 20,000 neutral sample particles uniformly in the discharge tube with their velocity described by the Boltzmann distribution with the gas temperature T_g, while the same number of ions are spatially distributed in proportional to $\mathrm{J}_0(\alpha r/R)$ with the same velocity distribution as of the neutrals. We approximated the initial gas and ion temperature by the rotational temperature of OH radicals in our previous OES measurement (see Table 1).

According to the general DSMC scheme, we move every sample particle by the common duration Δt including wall reflection in the Cartesian coordinate (x, y, z). After the movement, we consider mutual collisions between the sample particles. We assume diffusive reflection for all the reflections at the tube wall with a constant temperature 400 K for any rare gas plasma discharges. Ions impinging the wall are assumed to be lost from the system, but we preserve the number of sample ions with generation of a new ion in the system. For this process, we choose a sample cell in proportion to the probability of ionization. We choose the cell according to the electron density multiplied by neutral density to simulate the direct electron impact ionization due to the low ionization degree by a random number. And we set another new ion in the chosen cell. This time, the velocity of the new ion is stochastically determined according to the Boltzmann distribution of the ion temperature in the chosen cell at the simulated moment.

We neglect the Coulomb collisions since we do not treat kinetics of electrons. The values of scattering angle of ions are basically small, and the ion-ion collision frequency is much smaller than the ion-neutral one. We treat the mutual collisions of sample particles by the maximum collision number method [8]. Since we treat an axially symmetric system, we conduct the judgment of the collisions by the Riechelman-Nanbu method [9]. That is, the collisions are treated 2-dimensionally after the rotational transformation of all the sample particles from (r, θ, z) to (r, 0, z) plane. After the judgment of the collision and the calculation of the scattering angle, we again return the position of every sample particle to the original position (r, θ, z) together with the scattering angle, then we continue the DSMC movements and collisions.

Concerning the collision cross sections between neutral particles, we apply diffusion cross sections Q_n, which were experimentally determined from the measurement of self-diffusion coefficients [10]. Since we treat molecular motions only at the thermal velocity, we suppose that the diffusion cross sections are constant with respect to the relative velocities of collision particles. We treat the molecules as rigid spheres, and assume the collisions are spatially isotropic in the center of gravity system.

We must consider two independent processes for ion-neutral collisions, i.e., charge transfer collision and pure elastic collision. We refer the former cross section Q^* to Duman et al. [11], whereas the latter Q_i to Karvchevskii and Potanin [12]. Here, we must remember that the weight of sample particles is different for the ions and the neutrals. That is, the velocity of the ions must be updates whenever they make collisions, whilst that of neutral particles is altered by the probability determined by the weight ratio of sample particles. With these procedures, we can keep various conservation laws of the statistical ensemble of sample particles. To summarize, Fig. 1 shows flowchart of the DSMC simulation of neutral particles and ions adopted in the present study.

3. Results and discussion

Based upon the numerical procedure described in the previous section, we carried out numerical simulation of kinetics of heavy particles in rare gas discharge plasmas under the initial conditions specified in Table 1, and we obtained radial component of velocity v_r, temperature T and density N of neutral particles (a) and ions (b) in Figs 2 - 4, respectively.

Figure 2 shows that the radial velocity component v_r of neutrals is almost zero over the discharge area, whereas that of ions becomes larger as they come close to the tube wall for any kinds of rare gas discharge plasmas. This is because the ambipolar electric field E_r increases with the radial coordinate r and consequently the ions are strongly accelerated. When we compare the ion velocity of different kinds of rare gas, we find that lighter rare gas ions are more accelerated. As a most straightforward reason, when the potential drop is the same for different ions, the energy conservation leads to the fact that the velocity obtained is proportional to $m^{-1/2}$, where m is the mass of the ion. Second, as a minor effect, the experimentally observed electron temperature was higher for the lighter rare gas

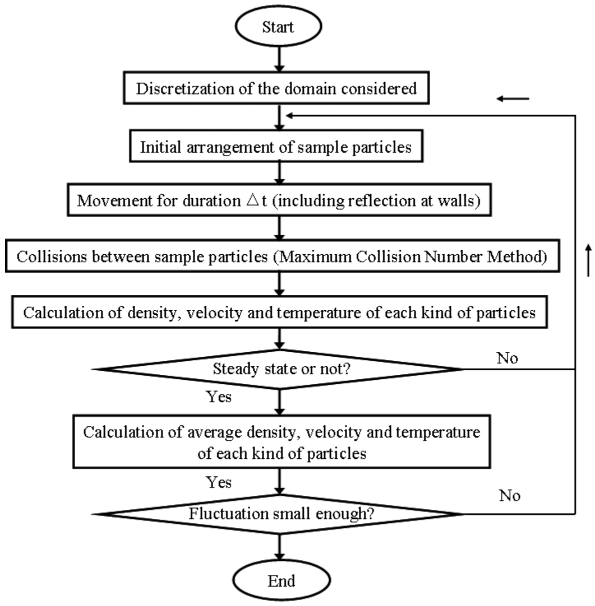

Fig. 1. Schematic outline of the present simulation. First, the domain considered is discretized by the mean free path presumed. Next, sample particles are set according to the initial conditions, followed by a movement in a given electric field, and then the occurrence of collisions is judged by the maximum collision number method for each sample particle. After establishment of steady state, density, velocity and temperatures are calculated by the accumulation of kinetics of sample particles till sufficient statistics are accomplished.

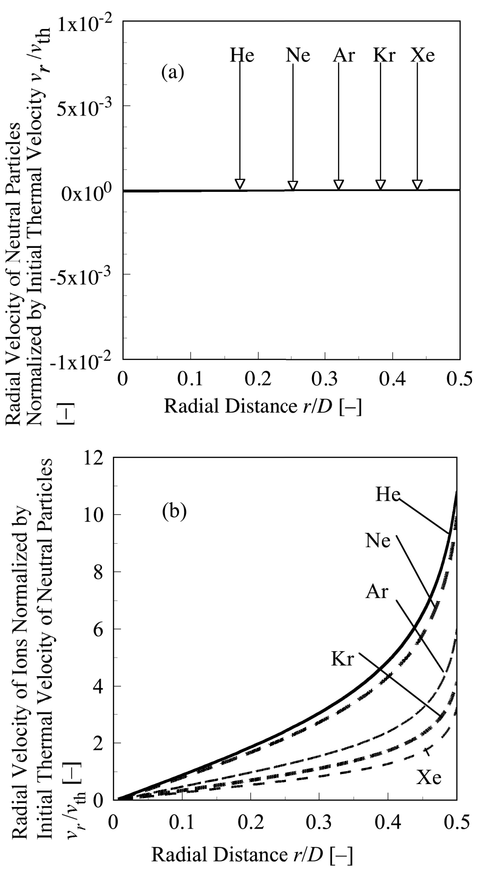

Fig. 2. Radial velocity normalized by initial thermal velocity of neutral particles. (a) neutral particles (almost zero for any discharge species), (b) ions.

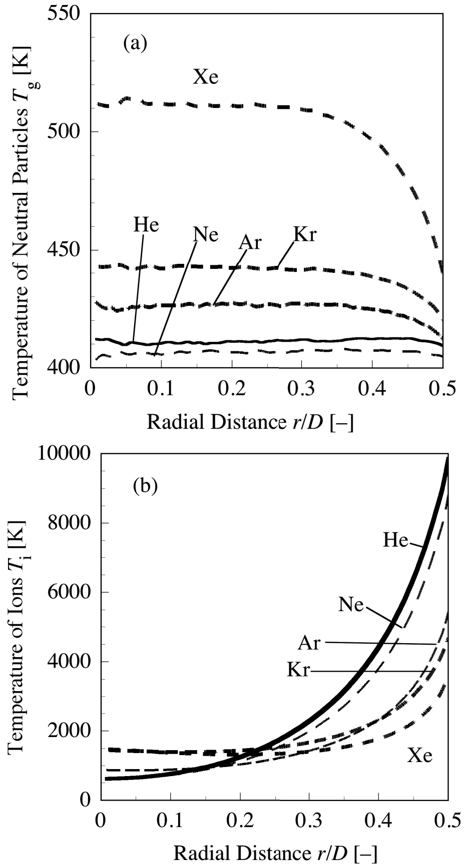

Fig. 3. Temperatures calculated. (a) neutral particles, (b) ions.

discharges with a constant microwave power. Then, the ambipolar electric field becomes stronger for the lighter rare gas discharges, and consequently, the lighter ions move under the stronger electric field.

Figure 3 shows that the ion temperature increases as ions come close to the tube wall, and particularly, that of helium ions becomes as high as 1 eV in the vicinity of the wall. This is because the ambipolar electric field becomes stronger as the radial distance from the axis increases, and, the following collisions with neutral particles randomize the direction of motion, which results in the increase in ion temperature. Meanwhile, in the central region of the discharge tube, the order of the ion temperature becomes inverted from the boundary region with respect to the molecular mass. This is because the temperature of neutral particles there becomes higher for the heavier rare gas molecules, except for helium and neon, as discussed in the next paragraph. For the lighter rare gas ions, the neutral particles as collision partner in the central region has lower temperature and the ions become cooled down more rapidly.

On the other hand, we found that the neutral temperature becomes lower as it comes to the tube wall. Before these simulations, we numerically confirmed that the temperature of neutral particles becomes equilibrated with the wall temperature unless we set ions in the domain considered. Therefore, it is concluded that the neutral particles become heated by collisions with the ions accelerated by the ambipolar electric field. The neutral particles are cooled down by the collisions with the wall. The ion density is also lower there than in the central region, and consequently, the neutral particles are cooled down at the vicinity of the wall.

When we compare the neutral temperature between the different kinds of rare gas plasmas, it is generally found that the heavier the rare gas molecules are, the higher the gas temperatures are, except helium and neon plasmas. Generally, this is considered to be an appropriate result for the neutral gas simulation, since the thermal conductivity of rare gas becomes smaller for the heavier molecules. We also found the radial temperature gradient for the neutral particles in the cylindrical discharge tube, which becomes more remarkable for the heavier rare gas plasmas and not so remarkable for neon and helium plasmas. It should be also noted that the present numerical result agrees with our previous experiments very well, including the order of neon and helium gas temperatures, for the same microwave power load and the same discharge pressure [2]. Although the electron temperature of helium plasma experimentally observed is a little higher than that of neon plasma, the difference is only 20 % and the effect is considered to be a minor one. The major reason for the gas temperature inversion for the helium and the neon plasmas is attributed to the difference in the ion density, i.e., the ion density of helium plasma is about twice higher than that of the neon plasma. Consequently, the collision frequency of neutral particles with ions becomes higher, and neutral helium molecules are much more heated than neutral neon molecules. Nevertheless, for any kinds of rare gas plasmas, the difference in the temperature between ions and neutral particles are remarkable, and we should consider that the ions and neutrals are not equilibrated with each other.

The radial distributions of neutral particle number densities are found to keep the pressure constant over the whole discharge region, which is shown in Fig. 4. That is, the neutral particles in the central region generally become depleted than in the near-wall region. It is also consistently found that the neutral density gradient becomes more steep for the heavier rare gas plasmas due to the temperature gradient, mainly caused by the difference in their thermal conductivity. And this causes ion depletion in the central discharge area as shown in Fig. 4(b), particularly for the heavier rare gas plasmas, whose number density of neutral particles becomes remarkably lower there. That is, the neutral depletion in the radially central area results in the lowering of ionization frequency, and finally, the ion density becomes lower there.

4. Conclusion

In the present study, we carried out numerical analysis of transport phenomena of neutral particles and ions of various kinds of rarefied rare gas plasmas in a cylindrical discharge tube by DSMC simulation. We conducted the weighted DSMC method in the simulation, where the sample neutral particles have much higher particle weight than the sample ions to keep statistical correctness of sample ions. Our numerical simulation showed that the neutral particles are heated by collisions with ions that are accelerated by the ambipolar electric field. The increase in temperature of neutral particles is remarkable in the radially central region of the discharge plasma. In the vicinity of the discharge tube wall, the neutral particles showed lower temperature due to the collisions with the tube wall. On the other hand, the radial temperature distribution of ions showed that they become heated as they come close to the wall due to the acceleration by the ambipolar electric field and the following collisions with neutral particles. For any kinds of rare gas plasmas, the ions showed much higher temperature than the neutral particles, which indicates that the system is quite far from the thermodynamic equilibrium. Our numerical simulation also showed that the gas temperature becomes higher for the heavier rare gas molecule plasmas except for helium plasma, which explains our previous experimental results very well including the helium gas temperature exception. We also found remarkable radial distribution of number densities. Neutral particles become depleted in the central region due to the high temperature, which results in the discrepancy of ion density distribution from the Bessel function J_0.

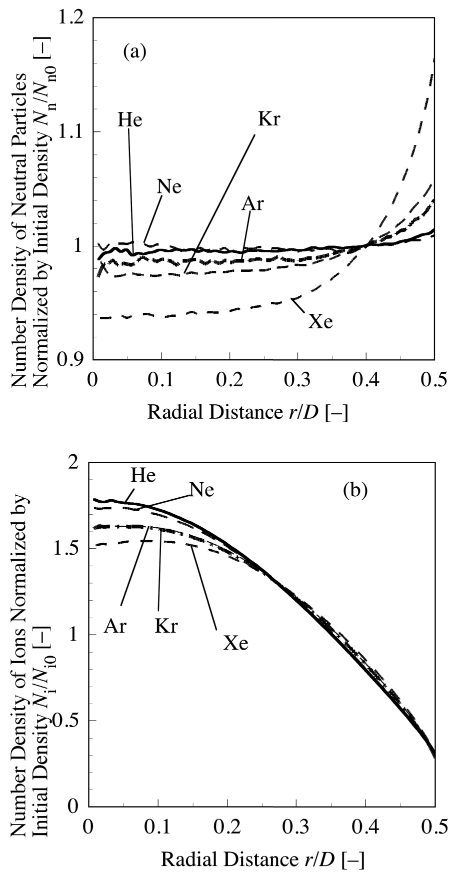

Fig. 4. Number densities calculated. (a) neutral particles, (b) ions.

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