A MONTE CARLO SIMULATION OF NEON ISOTOPE SEPARATION IN A DC DISCHARGE THROUGH A NARROW CAPILLARY

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Abstract

A numerical simulation was undertaken on the neon isotope separation in a DC arc discharge through a narrow capillary The mass transport phenomenon of neutral particles as well as ions was treated by the direct simulation Monte Carlo (DSMC) method. The numerical results qualitatively agreed with existing experimental ones, concerning not only the isotope separation phenomena, but also the pressure difference between the region of the anode and that of the cathode.

1. INTRODUCTION

Here, we briefly report a numerical study on isotope separation in a steady state DC arc discharge of neon along a thin closed capillary. It was experimentally found that the cathode region was enriched in the heavier isotopes, while the anode region in the lighter isotopes [1–6]. From the theoretical point of view, Matsumura and Abe pointed out that the friction force caused by the drift motion of ions onto neutral particles should be the most essential [1]. Later, Karchevskii *et al.* took account of the momentum transfer collisions between various particles in the plasma, which created a difference between isotopes in the diffusive friction force between ions and neutrals, which was given the term "ion wind" [4, 7] Karchevskii and Potanin suggested another mechanism [8]. The lower mobility of the heavier ions results in the higher degree of ionization of the heavier isotopic species. Consequently, the heavier isotopes are enriched into the cathode region, which was referred to as "isotopic cataphoresis". It was also found that the pressure in the anode region became always higher than that in the cathode region in a steady state DC arc discharge along a thin capillary [9]. When we discuss the isotope separation in the DC discharge, the theory should account for the pressure difference simultaneously. It is, however, still difficult for us to find the sufficient account in the foregoing theories. Anyway, we should

consider the dynamics of neutral particles as well as ions

In addition, although all the existing theories are based upon the fluid dynamic treatment of the neutral species and ions in the plasma, the Knudsen number of the neutral particles in a DC discharge capillary becomes 0 01 – 0.1. We should treat such rarefied systems by the Boltzmann equations, which can be solved stochastically by the DSMC (Direct Simulation Monte Carlo) method [10] To confirm the experimental results and the theories, we undertake the numerical simulation of the isotope separation in a DC discharge of neon by the DSMC method in the present study.

Here, we cannot apply the conventional PIC (Particle In Cell) DSMC method [11], for two reasons First, in the PIC method, we cannot simulate the motions of neutral particles. Secondly, the discretization of the domain should be the order of the Debye length, which requires too much memory. Moreover, the time step should be less than the order of the inverse of the plasma angular frequency, which requires impracticable long CPU time for neutral particles We can, however, calculate movements of the heavy particles (neutrals and ions) in a fixed average electric field with a realistic memory size and with reasonable CPU time Since the measured isotope enrichment factor was proportional to the length of the positive column, the essential separation is accomplished in the positive column. All we have to simulate is the transport phenomena, especially of neutral particles, in the positive column, not in the various dark spaces. Still, we have to include the effect of electron collisions, since the pressure in the anode region is always higher because of electron wind.

2. NUMERICAL PROCEDURE

As a preliminary numerical simulation, we treated the system with its Knudsen number 0.1 We discretized the domain by the order of the mean free path of heavy particles, 0.236 mm, and by the time step of the 1/5 of their mean free time, 0.1 μs, which corresponded the total discharge pressure 2 Torr. We assumed a uniform electric field in the longitudinal (z) direction E_z, since we considered that the space had no electric charge, namely $n_i = n_e$ anywhere at any time. This is because the discretization length is much larger than the Debye length. The value of E_z was adopted from the experiments [6]. In the radial (r) direction, we assumed an ambipolar electric field, whereas in the very close region to the wall (considered to be the sheath region), we assumed no radial electric field [9]

We considered that the discharge space was closed, and surrounded by the cylindrical glass capillary for the side wall, and by the virtual electrodes for both ends of the capillary, and we neglected the existence of various dark spaces, since it was experimentally found that the separation was accomplished in the positive column We assumed that the simulated space should correspond to the positive column. We set 5 cells in the radial direction and 200 cells in the longitudinal direction to simulate a capillary of 2 36 mm in its inner diameter and 4 72 cm in its length

As an initial condition, 30000 sample molecules were set in the capillary, 10 % of which were taken to be ^{22}Ne, and the rest of them ^{20}Ne, by which we approximately simulated the natural isotopic abundance of neon. Each sample molecule was marked with its charge state We neglected multiply charged ions since the electron temperature was experimentally measured to be 2 eV at most The initial ion density was set to be 5×10^{13} cm^{-3}, which corresponded to the ionization degree 0 5 % and the discharge current about 5 A in our experiments [6] The electron temperature T_e at this time was also estimated from the experiments as $1\ 8 \times 10^4$ K, which was supposed to be constant throughout the inside of the capillary at any

time. The initial positions of ions were set to be uniform in the longitudinal direction, and to be of $J_0(2.405r/R)$ distribution in the radial direction. Their velocities were calculated according to the Boltzmann distribution with the temperature T_i, to which the drift velocity, a function of the reduced electric field E_z/n (n is the number density of the heavy particles), was added. Neutral particles were set uniformly in the capillary, and their velocities were calculated according to the Boltzmann distribution with the temperature T_g. For the initial temperatures of heavy particles, we assumed $T_i = T_g = 1300$ K. We moved these sample molecules 3-dimensionally in the Cartesian (x,y,z) coordinates.

Every reflection on the capillary wall was treated as a diffusive reflection. We assumed the wall temperature to be 300 K. All the ions that came into the wall were reflected as neutral molecules. The reflection on the electrodes was treated similarly. All the ions that arrived at the cathode were neutralized, while all the ions that happened to come to the anode were made to remain ions. If a sample ion came to the cathode at a certain time step, we chose a neutral particle, by using a random number at the closest cell to the anode and of the same radial cell, made it ionized. Thus, the total number of the sample ions was kept constant. The temperature of the virtual electrodes was supposed to be the same as that of the wall.

We neglected the collisions of charged particles with other charged particles owing to the low ionization degree. We treated the collisions of a sample neutral molecule with other sample molecules by the maximum collision frequency method [12]. Since the system was axially symmetric, all the collisions were simulated 2-dimensionally with the rotational transformation from (r, θ, z) to $(r, 0, z)$ [13].

In a general scheme to simulate a weakly ionized rarefied gas up to the present time, collisions of electrons are calculated with background neutral molecules (the null collision method [14]). In the present simulation, on the contrary, we made electrons in the background, according to the Boltzmann distribution of the electron temperature with the drift velocity. Here, we needed the electron density in the cell. The electron density was assumed to be equal to the ion density because of rough discretization. Since the ionization degree was rather small, the calculated ion density contained unrealistic distortion. We made a general smoothing operation on the ion density in the z-direction at each calculational time step, and we redistributed the density according to the J_0 distribution in the r-direction.

For the electron impact ionization, we should not apply the same procedure as elastic collisions, since it was already proved that almost all the ions were ionized by metastable-metastable collisions, or by cumulative ionization in the positive column [15]. As a phenomenological approximation, whenever an ion recombined, we made a neutral molecule ionized in the cell whose z-coordinate was equal to that of the recombined ion, whereas its radial coordinate was chosen by a random number according to the calculated electron distribution multiplied by neutral density, which corresponds to the local balance of electron impact ionization and recombination at the wall due to the ambipolar diffusion. We can keep the initial average electron density over the capillary in this scheme.

In respect of various atomic parameters required in the present simulation, the elastic cross section of neutral-neutral collisions was calculated from self diffusion coefficient [16]. The neutral-ion charge transfer collisions was taken from [17]. To estimate the elastic ion-neutral scattering cross sections, we referred Ref. [18] including the polarization force. We took the cross sections of electron-neutral elastic scattering from Hayashi [19]. We refer C. S. Chang *et al.* for the electron and the ion drift velocities [20].

The time required for the isotopic equilibration in the capillary was estimated to be about 0 003 sec, i.e 30,000 time steps for the present system. After that, we summed up the particle density and the velocity to evaluate the mass transport phenomena in a steady state, and we calculated up to 700,000 time steps. One run took about 15 days by a work station HP9000/735 Consequently, we can summarize the procedure as in Fig. 1.

3. RESULTS AND DISCUSSION

As a result, Fig 2 shows the isotopic distribution in the capillary in a steady state. The heavier isotope was found to be enriched into the region of the cathode, while the lighter isotope into the region of the anode. In addition to the qualitative agreement, if we extrapolate the experimental results [6], we

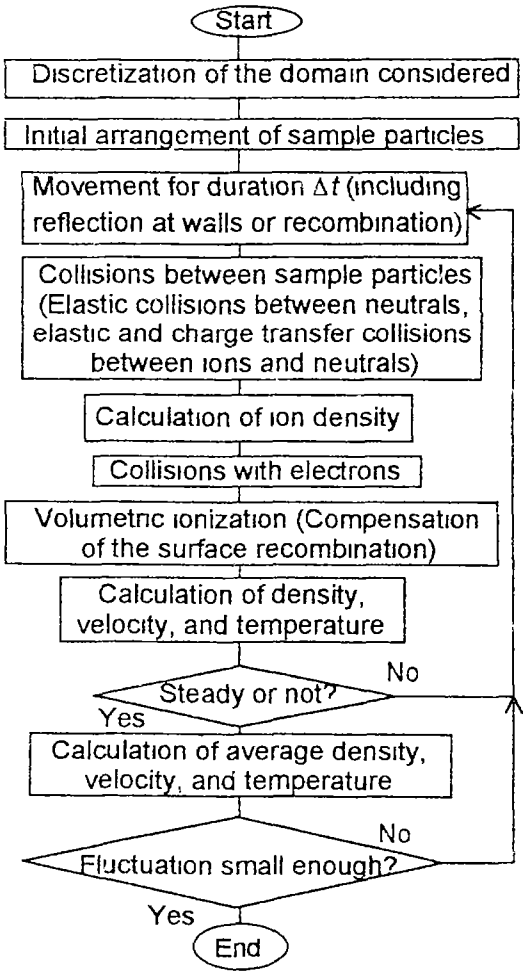

Fig 1. Outline of the present simulation.

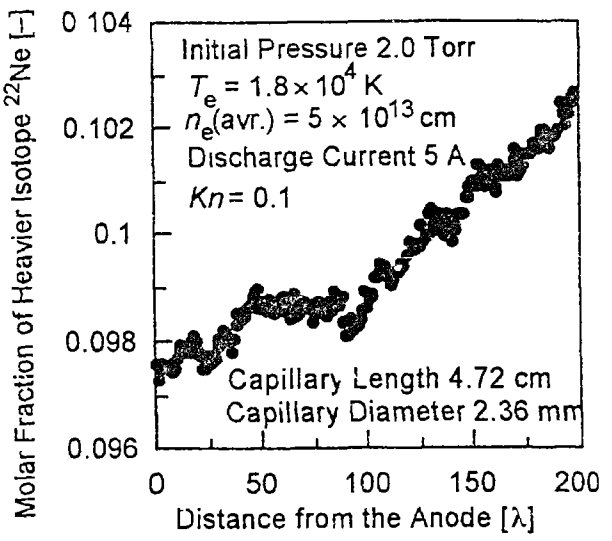

Fig. 2. Calculated molar fraction of the heavier isotope ^{22}Ne in the axial direction. λ is the average mean free path of neutral particle, 0.236 mm.

can find rather quantitative agreement of the simulated result with the experiment. It was also found that the enrichment coefficient was improved in the low pressure region

Secondly, the distributions of the number density of each neutral molecule are shown in Fig. 3. They became higher in the anode region than in the cathode region When we neglected the electron collisional processes or the radial electric field, such a

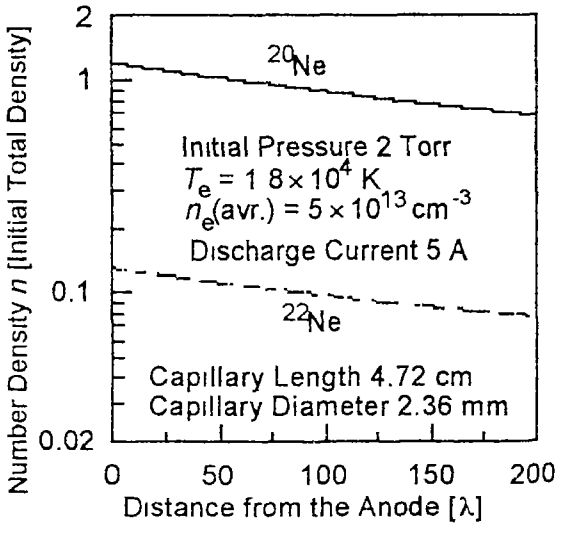

Fig. 3. Calculated number density of each isotope of neon in the axial direction. λ is the average mean free path of neutral particle, 0.236 mm.

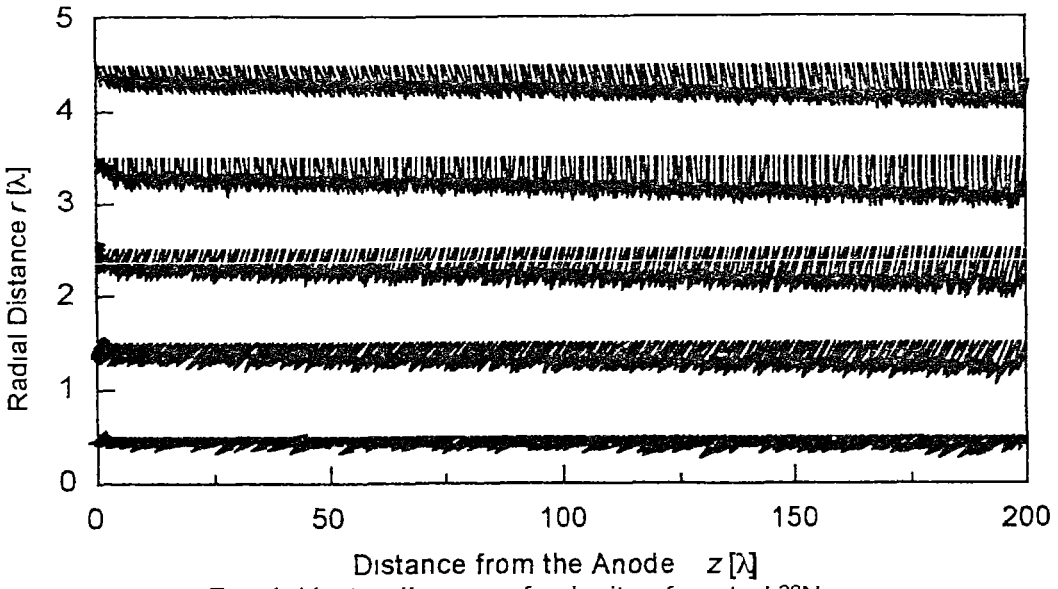

Fig. 4. Vector diagram of velocity of neutral ^{20}Ne.

result was never obtained. It means that the behavior of electrons, including the ambipolar electric field, plays an essential role in a positive column.

Thirdly, it should be noted that in the present simulation, the drift velocity of ions v_{id} should be also calculated like the swarm method [21]. It leads to $v_{id} \approx 1.3 \times 10^2$ m/s at $E_z/p \approx 2$ V/(cm•Torr), for ^{20}Ne, which results in its mobility $\mu_i \approx 3.6 \times 10^3$ cm^2/(V•s), and is considered to agree with its reported value very well [22].

In addition to the movement of ions, we can simulate the dynamics of neutral particles, which are generally neglected in the electron or ion swarm calculation. Figure 4 shows the vector diagram of velocity of neutral light isotope ^{20}Ne. The present analysis revealed the existence of the return flux of neutral particles to the anode near axis region, whereas the ion wind was dominant at the near wall region, and as a whole, the neutral neon flows to the anode to compensate the ion flux. It was also confirmed that the average ion temperature over the cross sectional area of the capillary was found to be higher than that of neutrals, which is shown in Fig. 5 These results qualitatively agree with the experiments of an argon plasma for an Ar ion laser by Ovsepyan [23]

4. CONCLUDING REMARKS

Mass transport phenomenon in the positive column was numerically simulated by the direct simulation Monte Carlo (DSMC) method based on atomic collisional processes. In consequence, we can conclude that the present simulation satisfactorily explains the experimental results, concerning not only isotope separation phenomena but also the pressure difference between the cathode area and the anode area. The isotope separation phenomena result from the atomic collisional processes in the DC positive column Of course, there remains much uncertainty in the present treatment of collisional cross sections. As a preliminary simulation, however, the present results show a possible application of the DSMC method for the transport phenomena of neutral particles, which exist as background of a positive column. This kind of simulation is considered to be useful to analyze the transport phenomena, including neutral particles, of DC discharge through a narrow path

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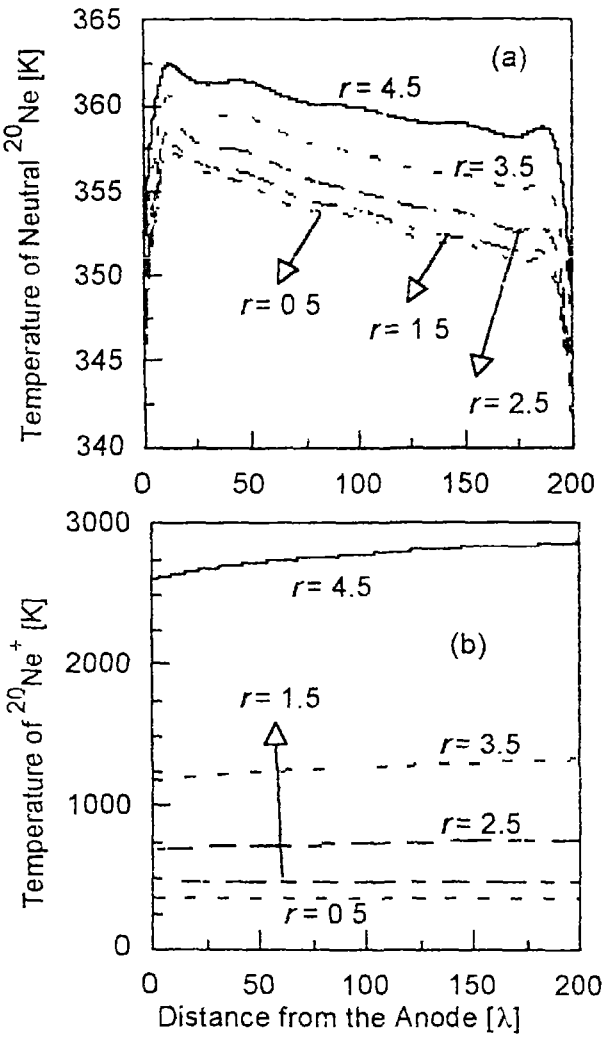

Fig. 5 Temperature distribution of 20Ne. (a) gas temperature, (b) ion temperature

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