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DIAGNOSTICS OF HIGH PRESSURE MICROWAVE DISCHARGE PLASMA BY LANGMUIR PROBE

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ABSTRACT

Probe characteristics measured in high pressure microwave excited helium and argon plasmas are characterized using simple algebraic functions. A microwave power from 100-400 W is used to generate helium and argon plasmas. Evolutions of electron concentration and temperature with increasing microwave power have been studied. It is found that electron concentration increases linearly with increasing microwave power both for 400 and 500 torr. The rate of production of ions in helium discharge is higher than that of argon because of higher direct electron impact ionization rate coefficient of helium. Electron temperature decreases with increasing microwave power because electron concentration increases with increasing power and consequently electron temperature decreases due to the screening of microwaves inside the plasma column.

INTRODUCTION

High pressure discharges are drawing much attraction for the reactive plasma processing(1-4) due to the production of a large amount of reactive species like ions, excited atoms and free radicals. The convenient features of the microwave excited high pressure non-thermal plasmas include, lower gas flow rate, efficient power coupling from the generator to plasmas and devoid of electrode contamination. High pressure discharges have been applied to UV light sources, gas discharge lasers and ozone production for water and air purification.

Plasma parameters are pre-requisite criteria for plasma source design and discharge performance optimization. Langmuir probe is a widely used diagnostic tool for the determination of local plasma properties. So, it is important to select a proper theoretical probe model, which allows reliable characterization of the measured probe characteristics under certain experimental conditions.

Several theoretical continuum probe models(5-15) have been proposed to analyze the probe characteristics measured in the pressure range where conditions of $\lambda_e << \lambda_D << r_p$ are fulfilled, where λ_e, λ_D and r_p are the mean free path of electrons, Debye length and probe radius, respectively. Most of the theories mentioned above do not fulfill our experimental conditions except that of Su *et al.*(10), Cohen(11) and Talukder *et al.*.(15) Su *et al.* considered such cases where the electron concentration n_e is sufficiently high and where both ions and electrons make numerous collisions with the neutral particles before being collected by the probe. Under such conditions, they found that the sheath thickness became comparable to r_p for highly negative probe potentials and then calculated the probe characteristics for different values of Debye ratio D_λ, where $D_\lambda = r_p/\lambda_D$. Cohen[11] used the same theoretical probe model as proposed by Su *et al.*[10] assuming that λ_e and the ion mean free path λ_i are shorter than the sheath thickness. Cohen found that the probe characteristics never saturate even at large probe voltage V_p although they tend more nearly to saturate as D_λ becomes larger due to the influence of the probe potential which extends far beyond the space charge sheath. When the probe collects current, the probe potential decays only with D_λ^{-1} at a long distance from the probe. This indicates that the Debye shielding in the space charge sheath is incomplete. The expression of the results, however, is not convenient for practical probe data analysises. Talukder *et al.*[15] derived algebraic equations using Cohen's results for the determination of plasma parameters directly for any D_λ from 50 to 1600 and for any electron to ion temperature T_i ratio $\tau = T_i/T_e$ from 0 to 1.

THEORY

A. Electron current : The motion of the charged particles can be described by the collision dominated processes of diffusion and drift. In high density plasmas, ions and electrons make many collisions with the neutral particles before being collected by the probe, taking into account only elastic collisions, allowing one to use simplified continuity equations without source terms. But in reality many different collisional processes can take place within the ion sheath, which cause the generation of ions. For example, collisions between two metastable helium atoms result in two-step processes in the production of an ion and a fast electron. Conditions for which large current due to secondary electrons flows through the sheath, the direct electron impact ionization of neutrals, stepwise ionization of metastable species or even three-body recombination of ions and electrons can be considered. But the contribution of these effects to the total ion flux at moderate negative potential of the probe is small as compared to the ion flux extracted from the bulk plasma.

The electron random current I_{er} can be written[10] as

$$I_{er} = 4\pi r_p e n_o D_e , \tag{1}$$

with

$$D_e = \frac{1}{3}\lambda_e v_e, \quad \lambda_e = \frac{1}{n_o \sigma_{mt}}, \quad v_e = \sqrt{\frac{8\kappa T_e}{\pi m_e}}, \tag{2}$$

where v_e, m_e and σ_{mt} are the thermal velocity, mass and total collision cross section for momentum transfer of electrons, respectively. The random electron flux per unit probe surface area, Γ_{er} can be written as

$$\Gamma_{er} = \frac{I_{er}}{4\pi r_p^2 e} = \frac{n_o v_e}{4}\left(\frac{4}{3}\frac{\lambda_e}{r_p}\right). \tag{3}$$

It should be noted that Γ_{er} is reduced by a factor of $4\lambda_e/3r_p$ as compared to the case where collisions are absent in the sheath region.

From Eq. (3), I_{er} is rewritten as

$$I_{er} = eA\Gamma_{er} = \frac{4}{3}\pi r_p e n_o \lambda_e v_e, \tag{4}$$

where $A = 4\pi r_p^2$ is the surface area of a spherical probe. The value of electron current I_e for an arbitrary probe voltage is expressed[15] presumably by introducing a function $I_{en}(\varphi_p, D_\lambda)$ as

$$I_e = I_{er} I_{en}(\varphi_p, D_\lambda), \tag{5}$$

where φ_p is the normalized probe potential and $I_{en}(\varphi_p, D_\lambda)$ is empirically presumed as

$$I_{en}(\varphi_p, D_\lambda) = \frac{A_1 \exp\left\{\dfrac{\varphi_p - A_2(D_\lambda)}{A_3(D_\lambda)}\right\}}{1 + \exp\left\{\dfrac{\varphi_p - A_2(D_\lambda)}{A_3(D_\lambda)}\right\}}, \tag{6}$$

and A_1, A_2 and A_3 are approximated by the rational functions of D_λ, respectively as

$$A_1(D_\lambda) = \frac{2.551 + 4.165\times10^{-2} D_\lambda}{1 + 2.011\times10^{-2} D_\lambda}, \tag{7a}$$

$$A_2(D_\lambda) = \frac{0.835 + 1.29\times10^{-2} D_\lambda}{1 + 6.36\times10^{-2} D_\lambda}, \tag{7b}$$

and

$$A_3(D_\lambda) = \frac{2.104 + 1.107\times10^{-2} D_\lambda}{1 + 3.59\times10^{-2} D_\lambda}. \tag{7c}$$

Introducing Eqs. (7) along with Eq. (6), one can easily fit $I_{en}(\varphi_p, D_\lambda)$ to any D_λ from 50 to 1600.

B. Secondary electron emission current : Generally, the probe current I_p consists of I_e, the current I_i of ions and the current I_{em} caused by the secondary electrons emitted from the probe surface. In high pressure high density plasmas, a large amount of secondary electrons[16,17] can be emitted from the probe due to the potential emission (Auger neutralization of ions or Auger de-excitation of metastable atoms). In such a case, I_p is modified to

$$I_p = I_e - I_i - I_{em} \,. \tag{8}$$

With decreasing pressure, the contribution of I_{em} to I_p decreases because of increasing wall losses of metastable atoms.

Under the influence of metastable atom fluxes to the probe the secondary electron emission current I_{em} deduced by Kagan *et al.*[18] given by

$$I_{em} = eA\gamma_m \Gamma_m = \frac{en_{mo}A\gamma_m}{4}\sqrt{\frac{kT_m}{2\pi m_M}}, \tag{9}$$

where n_{m0}, T_m and m_M are the density, temperature and mass of metastable atoms, respectively; Γ_m is the metastable flux to the probe and γ_m is the secondary electron emission coefficient. Eq. (9) is applicable[4,15] only for the ion sheath thicknesses smaller than the mean free path of metastable particles and to the gas pressure of about 12 torr. So that at high pressure, a continuum probe model for a thin and a thick sheath should be considered.

1. THIN SHEATH APPROXIMATION

The ion sheath is very thin at lower probe potential and close to the probe surface and consequently the sheath edge can be considered as the probe surface. Hence, the metastable concentration at the probe surface is zero. The secondary electron emission current[4] due to metastable atoms flux to the probe under the conditions considered can be written as

$$I_{em} = -e\gamma_m\, An_{mo}\sqrt{D_m n_o C_m}\,, \tag{10}$$

where D_m and C_m are the metastable diffusion coefficient and cumulative ionization rate coefficient, respectively. The thin sheath approximation can be used for slightly negative or positive probe potentials.

2. THICK SHEATH APPROXIMATION

If the ion sheath region is free from electrons and there are no destruction or production of metastable atoms due to the stepwise ionization or electron impact, a model for thick ion sheath approximation can be used. The metastable induced secondary electron emission current[4] under the conditions considered can be written as

$$I_{em} = -e\gamma_m A D_m \frac{n_{mo}}{\delta_{sh}}, \tag{11}$$

where

$$\delta_{sh} = \frac{3}{2}\sqrt[3]{\frac{1}{3}\lambda_e\lambda_D^2\varphi_p^2}, \tag{12}$$

is the ion sheath thickness obtained[19] by solving one-dimensional Poisson's equation taking into account negligible electron concentration within the sheath and zero electric field at the sheath. Eq.(12) can be used for high electron concentration and consequently the high production rate of metastable excited atoms.

Taking our experimental conditions into account $P = 500$ Torr, $T_m = 1000$K, $D_m = 5.69 \times 10^{-4}$ m^{2}s^{-1}, $C_m = 1.14 \times 10^{-13}$ m^{3}s^{-1}, $n_{mo} = 7.6 \times 10^{21}$ m^{-3}, $\gamma_m = 10^{-1}$ and introducing the above values in Eqs. (4) and (10), it is obtained that $I_{ir} = 9.95$ μA and $I_{em} = 11.2$ mA. So it is reasonable to consider that at high pressures, I_{em} is much higher than I_i. Therefore, I_i can indeed be neglected with respect to I_{em}, hence Eq. (8) can be written as

$$I_p = I_{er}I_{en}(\varphi_p, D_\lambda) - I_{em}. \tag{13}$$

Now one can easily apply Eq. (13) by introducing Eqs. (6) and (10) to analyze the probe characteristics measured in high pressure plasmas with high metastable atoms concentration. Probe data have been analyzed by an iterative procedure employed by Talukder *et al.*[15]

EXPERIMENTAL

The probe characteristics were measured in the high pressure pulsed microwave excited plasmas. Block diagram of the experimental setup and the cross sectional view of the discharge chamber are shown in Figs. 1 and 2, respectively. Description of the experimental setup was published elsewhere.[2] The discharge chamber is made of a copper rectangular waveguide of 54 mm high, 108 mm wide and 130 mm long. Open ends of the rectangular waveguide are closed by pyrex glass plates in order to make the discharge chamber. Two cylindrical tungsten pipes of 4 and 6 mm in inner and outer diameters,

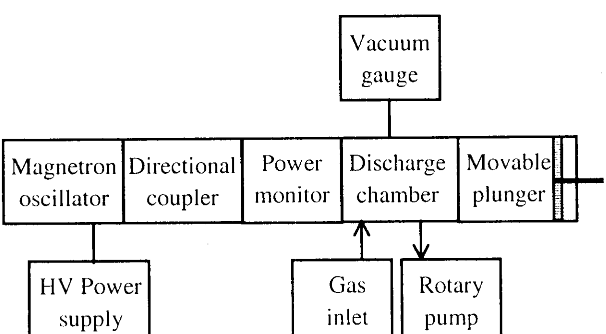

Fig. 1. Block diagram of the experimental setup.

respectively are inserted perpendicularly through the H-plane of the rectangular waveguide, as shown in Fig. 2. The spacing between the stub tips was 10 mm. In order to make a Langmuir probe, a tungsten wire of 0.8 mm in diameter embedded in the insulation with an alumina tube, is inserted through one of the stubs as shown in Fig. 2. The probe tip of 1 mm long is placed at the center of the stub and along its axis. A 2.45 GHz pulsed microwave with a 120 Hz repetition frequency is used. A digital oscilloscope was used to record probe characteristics. Fig. 3 shows the typical probe characteristics measured at 400 and 500 torr in microwave excited argon and helium plasmas at different microwave power.

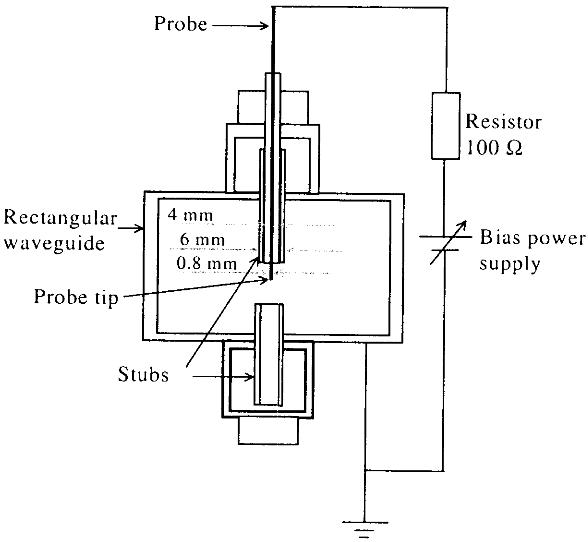

Fig. 2. Cross sectional view of the discharge chamber and probe setup.

RESULTS AND DISCUSSION

Fig. 4(a) and 4(b) show the development of electron concentration in helium and argon plasmas for 400 and 500 torr, respectively. In both cases, electron concentration increases linearly with increasing microwave power. This indicates that the absorption of microwave power increases with increasing pressure. By comparing Figs. 4(a) and 4(b), it is seen that electron concentration is higher in helium than that of argon plasmas. This phenomenon can be explained in the following way. The temperature of gas discharge plasma is substantially lower than the ionization potential E_i, because strong ionization takes place when the energy of electrons is less than the ionization potential by a factor in the range of 5 to 10. Atoms are ionized by the high energy electrons in the tail of the Maxwellian energy distribution function. The production rate of ion is estimated[20] by

$$P_i = n_g n_e k_i , \tag{14}$$

where n_g is the neutral particle concentration and the rate coefficients of ionization can be estimated by

$$k_i = 4\pi\left(\frac{m_e}{2\pi\kappa T_e}\right)^{3/2}\int_{E_i}^{\infty} v^3\sigma(v)\exp(-m_e v^2 / 2kT_e)dv, \tag{15}$$

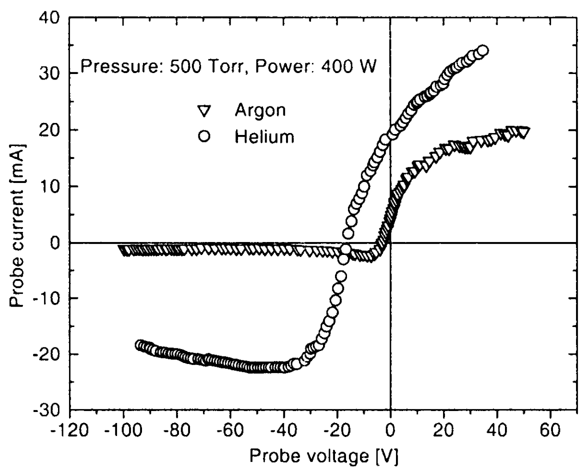

Fig. 3. Current-voltage probe characteristics measured at 500 torr with a 400 W microwave power.

assuming the Maxwellian velocity distribution function of electrons. Taking into account only the direct electron impact ionization process and our typical experimental conditions P = 500 Torr, neutral particle temperature T_g = 1000 K, $n_g = 1.64 \times 10^{25}$ m^{-3}, for (i) argon plasma T_e = 0.95 eV, $n_e = 7.17 \times 10^{18}$ m^{-3} and E_i = 15.759 eV, introducing these values in Eqs. (15) and (14), we obtain $k_i = 2.45 \times 10^{-24}$ m^3s^{-1} and $P_i = 8.49 \times 10^{19}$ $m^{-3}s^{-1}$, and for (ii) helium plasma T_e = 2.95 eV, $n_e = 1.09 \times 10^{20}$ m^{-3} and E_i = 24.59 eV, we obtain $k_i = 1.07 \times 10^{-19}$ m^3s^{-1} and $P_i = 5.65 \times 10^{24}$ $m^{-3}s^{-1}$. From the above estimations conclusion can be drawn that the production rate of ion by direct electron impact ionization is much higher in helium than those of argon plasmas.

Fig. 5(a) and 5(b) show the development of electron temperature for helium and argon plasma at 400 and 500 torr, respectively. Electron temperature both in helium and argon plasma decreases slowly with increasing microwave power. Because with increasing power electron concentration increases, consequently penetration of microwaves within the plasma column decreases, and as a result electron temperature decreases. The difference in electron temperature between 400 and 500 torr, both in helium and argon plasmas, is due to the increase of collision frequency between the electron and neutral particle with increasing pressure and consequently electron temperature decreases. Electron temperature in helium is higher than that of argon plasmas because the higher electron temperature is mainly responsible for the energy gap between the ground state and the excited or ionized state[21] of atoms. In order to provide validity of the probe theory used for the experimental probe data analyses, let us take into account the plasma parameters obtained by the present theory: $T_e = 2.95\ eV$, $n_e = 1.09 \times 10^{20}\ m^{-3}$ for helium, we obtain. $\lambda_D = 1.22 \times 10^{-6}\ m$. On the other hand the radius of the probe is $r_p = 4.9 \times 10^{-4}\ m$, that is $\lambda_D << r_p$, which justify the validity of selecting this probe theory used for analyzing the probe characteristics measured in high pressure plasmas.

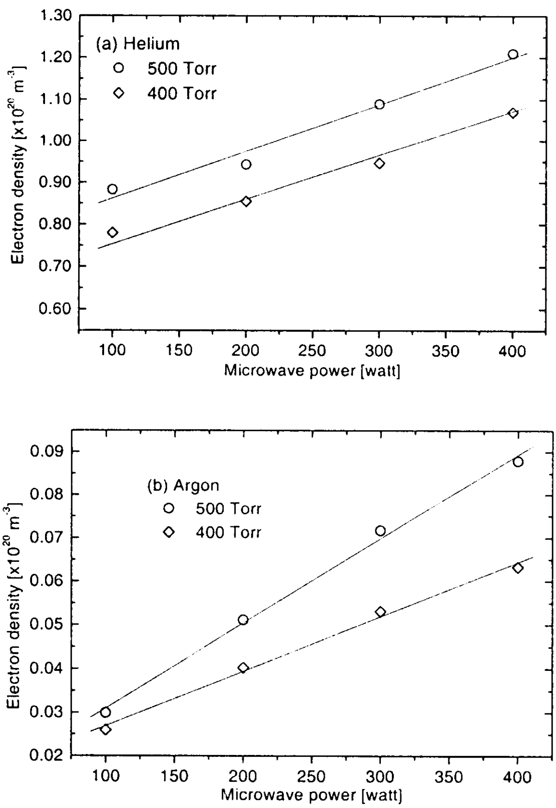

Fig. 4. Evolution of electron concentration with microwave power for (a) helium and (b) argon plasmas.

CONCLUSION

Microwave excited high pressure helium and argon plasmas are produced inside a rectangular waveguide with a microwave power from 100 - 400 w. Electrostatic probes are used for the diagnostics of helium and argon plasmas. By analyzing probe data, it is found that electron concentration increases linearly with increasing microwave power both for 400 and 500 torr. The ion production rate coefficient of helium discharge plasma is higher than that of argon discharge plasma because of higher direct electron impact ionization

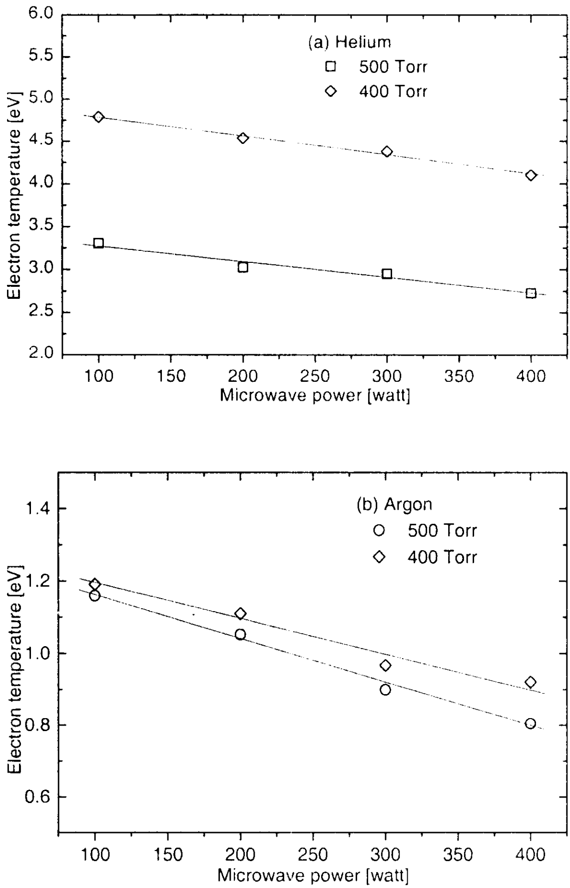

Fig. 5. Dependencey of electron temperature with microwave power for (a) helium and (b) argon plasmas.

rate coefficient of helium. With increasing microwave power electron temperature decreases because electron concentration increases with increasing power and consequently electron temperature decreases due to the screening of microwaves inside the plasma column.

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