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EFFECT OF PENNING IONIZATION ON THE BALANCE OF CHARGED PARTICLES IN PLASMA OF A STATIONARY REFLEX DISCHARGE

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A model of stationary reflex discharge, which is based on the volume-averaged (global) one, has been considered in the case of weakly ionized non-isothermal plasma. The temperature and the density of plasma electrons are calculated. The process of cathode sputtering and the formation of sputtered ions by means of the electron impact ionization, Penning ionization, and sputtered ion charge exchange at gas ions in plasma are analyzed. The dependence of the ionization degree of sputtered atoms on plasma parameters is determined. The influence of the Penning ionization on the balance of charged particles in plasma is considered.

Key words: Penning ionization, plasma, ionization degree, charge exchange, reflex discharge.

1. Introduction

The reflex discharge, which is also known as the Penning discharge, was proposed for the first time by F.M. Penning [1] in order to create a vacuum manometer. Nowadays, the major domain of reflex discharge applications includes the vacuum equipment, physics of atomic and electron collisions, physics of charged particle beams, plasma physics, applied plasma technologies, and so on. However, despite the long-term and numerous researches, there is no satisfactorily complete physical model that would describe the reflex discharge. This circumstance is partially connected with the fact that the reflex discharge has a number of modes depending on the working gas pressure and the magnetic field [2], with the parameters of the discharge and the formed plasma being substantially different in different modes. The reflex discharge at pressures higher than $p = 1.33 \times 10^{-2}$ Pa and low plasma ionization degrees is the most often used for the creation of plasma sources on its basis. In this case, an optimum relation between the plasma parameters and the energy contribution is often required.

In work [3], a model of stationary reflex discharge on the basis of the volume-averaged (global) model [4, 5] was proposed and analyzed. A comparison of

experimental results with calculated data [3] showed that the latter differ from the former to within a coefficient of about 2. Such an agreement between the results is satisfactory for rather a simple model of reflex discharge. However, in work [3], the ionization of the sputtered atoms of the cathode material owing to the Penning ionization, i.e. the ionization of a neutral atom at the collision with an atom in the metastable state, was not taken into consideration. At the same time, this phenomenon can considerably affect the ionization degree of sputtered atoms in weakly ionized plasma [6]. The course of the Penning ionization process is rather efficient, because the excitation energy of gas atoms in the metastable state exceeds the ionization potential of metal atoms.

In this work, the model proposed in work [3] will be developed further. Here, we consider the Penning ionization of sputtered cathode atoms and its influence on the balance of charged particles in plasma.

2. Volume-Averaged (Global) Reflex Discharge Model

While estimating the parameters of low-pressure plasma discharges, when the particle losses are mainly occurs owing to the diffusion, the volume-averaged (global) model is widely applied (see, e.g., works [4, 5, 7–12]). The overwhelming majority of those works are devoted to the consideration of both stationary and pulsed discharges without a magnetic

field, except for work [7], where the particle losses owing to the diffusion in a magnetic field were neglected altogether. In the general case, the reflex discharge is a discharge in a magnetic field. Therefore, in work [3], with regard for this and some other features of the discharge, a discharge model on the basis of volume-averaged (global) model was proposed.

In this work, the weakly ionized non-isothermal plasma of a stationary reflex discharge is considered. According to work [3], the total adsorbed power in the discharge equals

$$P_{\text{abs}} = N_e v_B \times (A_{\text{eff}} E_{\text{iz}} + A_R h_R E_e + A_L h_L E_i), \quad (1)$$

where N_e is the electron density in plasma [m^{-3}]; $N_e = N_i$; $v_B = \sqrt{T_e/M_i}$ is the Bohm velocity [m/s]; T_e is the temperature of electrons [eV] ($1 \text{ eV} = q \times 1 \text{ eV} = 1.6 \times 10^{-19} \text{ J}$, where q is the elementary charge [C]), M_i is the ionic mass [kg]; E_{iz} the average energy required for the formation of an ion-electron pair or the ionization cost; E_e the average kinetic energy taken away by an electron (provided the Maxwellian distribution, its value amounts to $2T_e$); $E_i = (T_e/2) + qV_s$ is the average kinetic energy taken away by ions, which is equal to the sum of the ion energy before entering the layer and the energy acquired in the layer; V_s is the cathode potential drop, which is approximately equal to the discharge voltage V_{dc} ; and A_{eff} is the effective area of charged particle losses [m^2]. The latter equals $A_{\text{eff}} = A_R h_R + A_L h_L$, where A_L and A_R are, respectively, the area of the base and the lateral surface of the cylinder [m^2]; and h_L and h_R are the axial and radial, respectively, ratios between the plasma density at the plasma boundary and the plasma density at the center.

According to works [13, 14], in the case of weakly ionized non-isothermal plasma, the quantities h_L and h_R equal

$$h_L = 0.86 \left(3 + \frac{L}{2\lambda_i} \right)^{-1/2}, \quad (2)$$

$$h_R = h_{R_0} (1 + G + G^2)^{-1/2}, \quad (3)$$

where $h_{R_0} = 0.8(4 + R/\lambda_i)^{-1/2}$ is the value of h_R in the absence of a magnetic field; $\lambda_i = 1/N_0\sigma_{\text{mi}}$ is the mean free path of an ion in the gas [m]; N_0 is the density of neutral particles [m^{-3}]; and σ_{mi} the transport cross-section of ion scattering by a neutral

particle [m^2]. At the collision of ions with the own gas neutrals, $\sigma_{\text{mi}} = 2\sigma_{\text{cx}}$, where σ_{cx} is the cross-section of resonant charge exchange [m^2]. The parameter G equals [14]

$$G = \frac{0.64}{\sqrt{4 + \frac{R}{\lambda}}} \frac{R\lambda_e}{r_e r_i},$$

where $\lambda_e = 1/N_0\sigma_{\text{me}}$ is the mean free path of an electron in the gas [m]; σ_{me} the transport cross-section of electron scattering by a neutral particle [m^2]; $r_e = v_{T_e}/\omega_{\text{ce}}$; $r_i = v_s/\omega_{\text{ci}}$; ω_{ce} and ω_{ci} are the electron and ion, respectively, cyclotron frequencies; v_{T_e} is the thermal velocity of an electron [m/s]; and v_s the ion sound velocity [m/s].

From Eq. (1), we can obtain a relation between the density and the adsorbed power in the discharge:

$$N_e = \frac{P_{\text{abs}}}{v_B (A_{\text{eff}} E_{\text{iz}} + A_R h_R E_e + A_L h_L E_i)}. \quad (4)$$

The balance equations for the charged particles in plasma, provided that the process of particle losses owing to the diffusion dominates over the recombination, can be written down in the form

$$N_e v_B A_{\text{eff}} = K_{\text{iz}} N_0 N_e V, \quad (5)$$

where V is the plasma volume [m^3], and K_{iz} the rate constant for the neutral particle ionization [m^3/s]. Equation (5) can be rewritten in the form

$$\frac{v_B}{K_{\text{iz}}} = \frac{N_0 V}{A_{\text{eff}}}. \quad (6)$$

By solving it, the electron temperature T_e as a function of the neutral particle density can be determined.

The interaction of plasma formed in the reflex discharge with the cathode surface results in the destruction of a cathode material owing to the corpuscular sputtering of this material into plasma and its subsequent ionization [15]. The balance equation between the neutral atoms that arrive into the reflex discharge plasma owing to the cathode sputtering by gas ions and the ions of cathode material looks like

$$Y_g N_g^+ A_L h_L v_B + Y_m N_m^+ A_L h_L v_B^m = \frac{N_m}{\tau_i} V + \frac{N_m}{\tau_D} V, \quad (7)$$

where Y_m and Y_g are the sputtering coefficients of the cathode material by metal and gas ions, respectively; N_m^+ the density of metal ions [m^{-3}]; N_g^+ the

density of gas ions [m⁻³]; v_B^m the Bohm velocity of metal ions [m/s]; N_m the density of neutral atoms of a cathode material [m⁻³]; $\tau_D = \Lambda/D_m$ is the characteristic diffusion time of a sputtered particle [s]; Λ is the characteristic diffusion length [m]; D_m the diffusion coefficient of a sputtered atom in the gas [m²/s]; and τ_i the characteristic time of the sputtered atom ionization [s]. Making allowance for the processes of electron impact ionization, charge exchange at gas ions, and Penning ionization, the characteristic time of the sputtered atom ionization equals [15]

$$\tau_i = \frac{1}{K_{iz}N_e + K_{CT}N_g^+ + K_{PI}N_g^m}, \quad (8)$$

where K_{CT} is the rate constant of gas ion charge exchange at a neutral metal atom [m³/s]; K_{PI} the rate constant for the Penning ionization of metal atoms by metastable gas atoms [m³/s]; and N_g^m the density of metastable gas atoms [m⁻³]. In view of expression (8), formula (7) looks like

$$N_m = \frac{A_L h_L [Y_g N_g^+ v_B + N_m^+ v_B^m (Y_m - 1)] \tau_D}{V}. \quad (9)$$

Since the sputtering is one of the major processes that result in the transport of a cathode material into plasma, the sputtering coefficient Y has to be determined. This coefficient depends on the charge, mass, and energy of a bombarding ion, the incidence angle, as well as on the target material and the temperature [15]. The process has an energy threshold of sputtering. In gas discharges, the ion obtains the energy for sputtering in the layer with cathode potential drop V_s (the cathode layer) [5, 16], which is approximately equal to qV_s . In the first approximation, we assume that $V_s \approx V_{dc}$. From the stationary condition for the given independent discharge [3], it follows that $V_s \approx V_{dc} = W_{iz}/q\gamma$, where γ is the secondary emission coefficient, and W_{iz} the average energy spent for the formation of an ion-electron pair by a primary electron emitted from the cathode surface. Despite that the physical senses of the quantities W_{iz} and E_{iz} are identical, their values are not identical in the general case, which is associated with different approaches to the determination of those quantities. This fact makes it possible to obtain rather a rough estimate for the V_s -magnitude. On the other hand, V_s can be estimated by equating its magnitude to the discharge ignition potential V_b . The corresponding value of the

sputtering coefficient will be some overestimated in this case, because $V_b > V_{dc}$. According to work [2], the condition of self-sustained discharge ignition in a non-uniform electric field can be written in the following form:

$$\ln \left(1 + \frac{1}{\gamma} \right) = \int_{r_1}^R \alpha(r) dr, \quad (10)$$

where α is the first Townsend coefficient (the number of ionization events per unit path length; R the anode radius; and r_1 a radius, at which an electron has a sufficient energy for the ionization. In the case of discharge in a magnetic field, the Townsend coefficient can be estimated from the equation [17]

$$\alpha = A_0 P \sqrt{1 + \omega_{ce}^2 \tau_e^2} \times \exp \left(-\frac{B_0 P}{E} \sqrt{1 + \omega_{ce}^2 \tau_e^2} \right), \quad (11)$$

where P is the gas pressure [Pa]; the constants A_0 [m⁻¹Pa⁻¹] and B_0 [V/m/Pa] are determined by the type of a gas [5]; τ_e the time of electron collisions [s]; and E the electric field strength [V/m]. In the case of gas discharge without a magnetic field ($B = 0$), the first Townsend coefficient equals $\alpha = A_0 P \exp(-B_0 P/E)$ [5, 16]. By solving Eq. (10) with regard for Eq. (11), it is possible to determine the discharge ignition potential V_b .

Let us write the balance equation for metal ions in the following form:

$$N_m^+ v_B^m A_{eff} = K_{iz} N_m N_e V + K_{CT} N_m N_g^+ V + K_{PI} N_m N_g^m V. \quad (12)$$

From Eq. (12), the density of metal ions is determined as follows:

$$N_m^+ = \frac{N_m V (K_{iz} N_e + K_{CT} N_g^+ + K_{PI} N_g^m)}{v_B^m A_{eff}}. \quad (13)$$

In the general form, the balance equation for metastable atoms in the reflex discharge looks like

$$\sum_i R_{Gener,i} = \sum_i R_{Loss,i} + \frac{N_g^m}{\tau_D^m}, \quad (14)$$

where $R_{Gener,i}$ and $R_{Loss,i}$ are the rates of metastable gas atom creation and loss, respectively [m⁻³/s]; $\tau_D^m = \Lambda/D_g^m$ the characteristic diffusion time for a

metastable particle [s]; and D_g^m the diffusion coefficient of metastable atoms in the gas [m²/s]. The reaction rates are written down as the product of the reagent densities $N_{r,i}$ and the reaction rate constant K : $R = K \times \prod_i N_{r,i}$.

The rate constants, cross-sections, and various process parameters used in the calculations were taken from works [18–26]. They are quoted in Table 1.

3. Results of Calculation and Discussion

The plasma parameters of a stationary reflex discharge were calculated for the conditions of the experimental installation described in work [27]. The geometrical dimensions of the installation were as follows: the anode radius $R = 0.08$ m; the distance between the cathodes $L = 0.336$ m; the volume $V = 6.756 \times 10^{-3}$ m³; the areas of the cylinder base and lateral surface $A_L = 0.04$ m² and $A_R = 0.169$ m², respectively. The cathodes were made of copper (Cu). The discharge was ignited at a low pressure of 0.01–12 Pa in the argon (Ar) atmosphere.

By solving Eq. (6) and making allowance for the initial conditions, the dependences of the electron temperature in plasma on the pressure were obtained for the cases of plasma in a magnetic field and without a magnetic field. They are depicted in Fig. 1. As one can see, the temperature of electrons decreases with the increase of the gas pressure and the magnetic field strength. The increase of the initial gas pressure or, equivalently, the increase of the density of neutral gas atoms gives rise to an increase of the probability for electrons to collide with gas atoms and, accordingly, of their energy loss in elastic and inelastic processes. On the other hand, an increase of the density of neutral atoms results in an increase of the plasma density, which can be seen from its dependences on the pressure exhibited in Fig. 2. The role of the magnetic field influence on the plasma density and temperature has two aspects. First, the magnetic field gives rise to an increase of the electron path length in the gas and, accordingly, to an increase of the probability for the electron to collide with gas atoms. Second, the field results brings about a reduction of particle losses from plasma associated with the diffusion. As a result, if compared with the discharge plasma without a magnetic field, the electron temperature decreases a little (see Fig. 1), and the density increases (see Figs. 2 and 3). The plasma

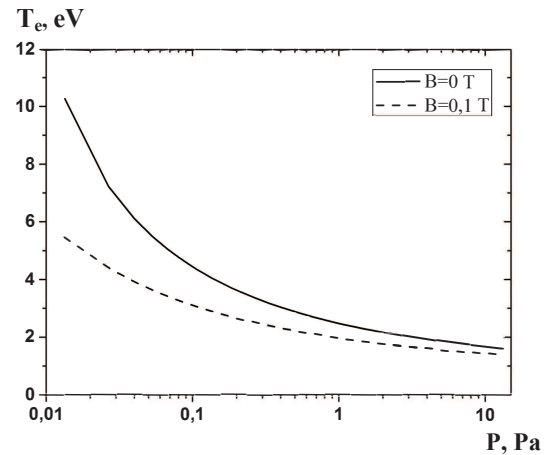


Fig. 1. Dependences of the electron temperature on the pressure

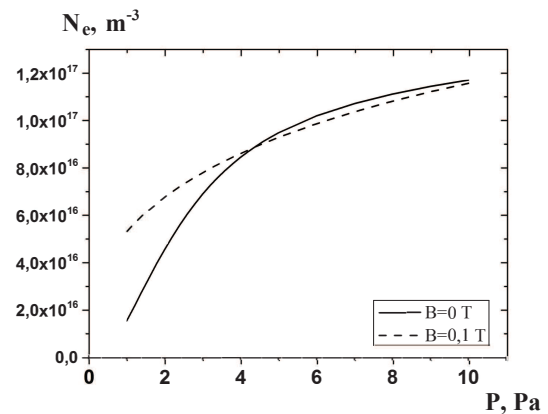


Fig. 2. Dependences of the electron density in plasma on the pressure at $P_{\text{abs}} = 100$ W

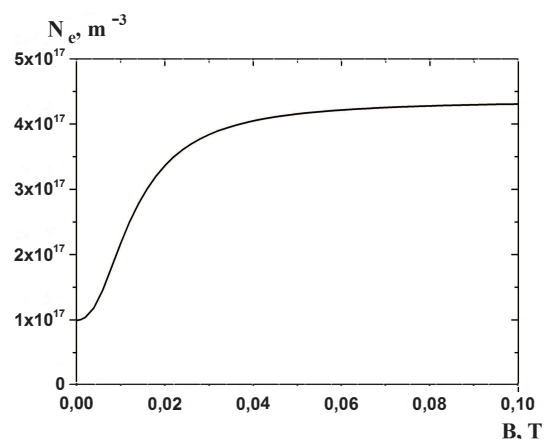


Fig. 3. Dependence of the electron density in plasma on the magnetic induction at $P_{\text{abs}} = 100$ W and $P = 2$ Pa

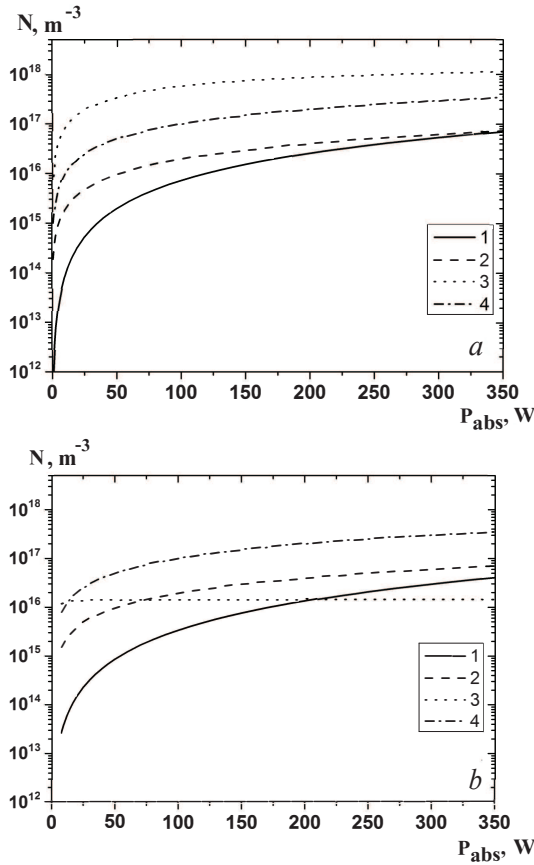


Fig. 4. Dependences of the particle densities in plasma on the adsorbed power in the discharge at $P = 2$ Pa and $B = 0.05$ T; metastable Ar atoms were calculated by formula (15) (panel a) and (16) (panel b); Cu ions (1); neutral Cu atoms (2); metastable Ar atoms (3); electrons (4)

density grows substantially as the magnetic field increases to 0.06 T (see Fig. 3) and weakly changes at higher fields.

Now, let us consider the formation of ions of sputtered atoms owing to the electron impact ionization, Penning ionization, and charge exchange at gas ions in plasma. For the calculation of the sputtering coefficient and the diffusion time of a sputtered neutral atom, we used a model proposed in work [15] The calculated value of the diffusion time of a sputtered particle τ_D amounted to 1×10^{-4} s (at $P = 2$ Pa) and the ionization time was $\tau_i = 0.07$ s. One can see that $\tau_D < \tau$, which testifies that only a small fraction of sputtered atoms was ionized, and the others turned out on the surface of a discharge chamber owing to the diffusion.

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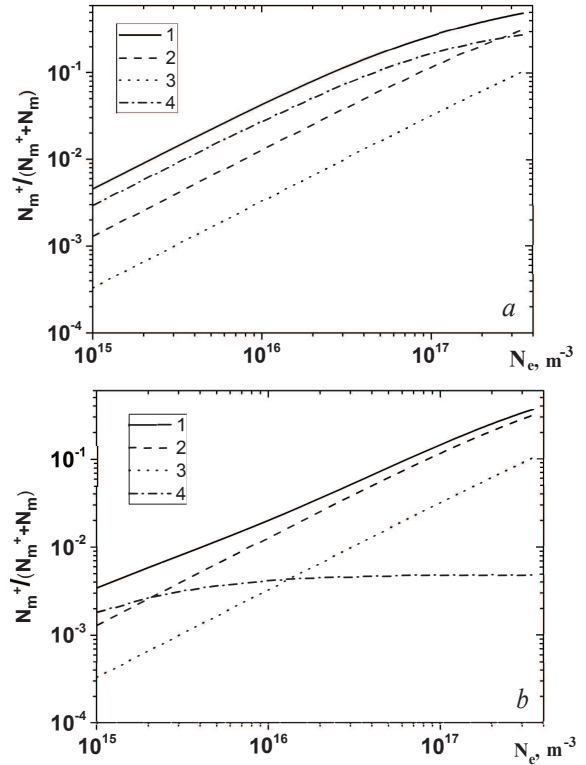


Fig. 5. Dependence of the ionization degree of sputtered copper atoms on the electron density in plasma; metastable Ar atoms were calculated by formula (15) (panel a) and (16) (panel b); the total contribution of processes (1), electron impact ionization (2), charge exchange at gas ions (3), Penning ionization (4)

Taking the above-mentioned processes into account, Eq. (14) for the balance of metastable atoms in the reflex discharge reads

$$N_g N_e K_{12} = N_g^m N_e K_{13} + N_g^m N_e K_{14} + N_m N_g^m K_{21} + \frac{N_g^m}{\tau_D^m}, \quad (15)$$

This equation can be rewritten in a more detailed form:

$$N_g N_e K_{12} + N_g^+ N_e K_{15} = N_g^m N_e K_{13} + N_g^m N_e K_{14} + N_g^m N_e K_{16} + N_g^m N_g^m K_{17} + N_g^m N_g K_{18} + N_m N_g^m K_{21} + \frac{N_g^m}{\tau_D^m}, \quad (16)$$

where the quantities from K_{12} to K_{18} and K_{21} are the rate constants (the subscript designates the number of the process in Table 1). By solving Eqs. (15)

and (16), as well as Eqs. (4), (9), and (13), the dependences of the densities for plasma electrons, metastable Ar atoms, neutral Cu atoms, and Cu ions were obtained, which are shown in Fig. 4. One can see that the choice and the account for elementary processes substantially affects the density of metastable argon atoms and, accordingly, the contribution of the Penning ionization to the ionization processes of sputtered atoms (see Fig. 5).

In one case (see Fig. 4, *a* and formula (15)) where the number of processes that were taken into account was minimum, the calculated density of metastable argon atoms exceeded that of electrons. A similar scenario is also typical, e.g., of the pulsed magnetron discharge at an insignificant degree of plasma ionization. The corresponding plasma parameters were calculated in work [28]. In this case (see Fig. 5), the main process giving rise to the ionization of sputtered atoms is the Penning ionization at the electron density $N_e \ll 2 \times 10^{17} \text{ m}^{-3}$. This conclusion agrees with the results of work [6]. It should be emphasized that the charge exchange of a gas ion at the metal atom was not taken into account in works [6, 28].

In the other case (see Fig. 4, *b* and formula (16)), the addition of processes to Eq. (15), when not only collisions with electrons are taken into consideration, but also collisions of metastable atoms with one another and with atoms in the ground state, gives rise to a weak density change of metastable argon atoms. For example (see Fig. 4), $N_{\text{Ar}}^m = 1.19 \times 10^{16} \text{ m}^{-3}$ at $P_{\text{abs}} = 8 \text{ W}$ and $1.44 \times 10^{16} \text{ m}^{-3}$ at $P_{\text{abs}} = 350 \text{ W}$. Therefore, the contribution of the Penning ionization to the ionization of sputtered atoms becomes substantial at $N_e \ll 2 \times 10^{15} \text{ m}^{-3}$ (see Fig. 5, *b*). The ionization degree of sputtered atoms is a little lower than in the first case (see Fig. 5).

One can see that the application of different sets of processes at calculations brings about a considerable discrepancy between the results obtained. It is natural that, for the description of processes with the participation of metastable atoms to be more complete, it is necessary that every process giving rise to the population of metastable levels in the atom should be made allowance for. In essence, we need in a complete enough model of impact plasma ionization, e.g., like that used in work [29], which involves collisions with electrons and atoms. In this case, the number of processes that should be taken into consideration will be rather large, whereas a complete

set of the rate constants for those processes could be absent. Therefore, a finite number of elementary processes, which is acceptable for the approximation of actual experimental conditions, is made allowance for at calculations. The analyzed spatially averaged (global) model is simple enough, and the results of calculations in its framework have sooner an evaluative character. At the same time, the results of calculations carried out in this and other works (see, e.g., work [6]) demonstrate that the role of the Penning ionization in the ionization of sputtered atoms in a weakly ionized plasma can be substantial. Therefore, the account for a small number of the processes, which are the most probable, in the model can be quite justified. Anyway, the choice of the processes for their consideration at calculations should be based on the available data for the probability of processes running under those or other initial conditions, and on the corresponding comparison between final calculation results and experimental data.

4. Conclusions

A model of stationary reflex discharge, which is based on the spatially averaged (global) model has been developed for the case of weakly ionized non-isothermal plasma. In the framework of the examined model for experimental conditions, the dependences of the electron density and temperature in plasma on the adsorbed power in the reflex discharge, magnetic field strength, and initial gas pressure are calculated. The results of calculations have shown that, under the given conditions and at an adsorbed power of 350 W, the temperature of electrons is about 1–10 eV, and the plasma density reaches a value of $3.5 \times 10^{17} \text{ m}^{-3}$. The processes of cathode sputtering and formation of the ions of sputtered atoms owing to the electron impact ionization, Penning ionization, and charge exchange at gas ions in plasma are considered. It is found that the role of the Penning ionization and, accordingly, its contribution to the ionization degree of sputtered atoms can be considerable, but the calculation results rather strongly depend on the choice of elementary processes with the participation of metastable atoms.

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ВПЛИВ ПЕННІНГОВСЬКОЇ ІОНІЗАЦІЇ
НА БАЛАНС ЗАРЯДЖЕНИХ ЧАСТИНОК У ПЛАЗМІ
СТАЦІОНАРНОГО ВІДБИВНОГО РОЗРЯДУ

Резюме

Розглянуто модель стаціонарного відбивного розряду на основі просторово-усередненої (global) моделі у випадку слабоіонізованої неізотермічної плазми. Проведено розрахунок температури та густини електронів плазми. Розглянуто процеси розпилювання катода та утворення іонів розпорощених атомів за рахунок іонізації електронним ударом, пеннінговської іонізації та перезарядження на іонах газу у плазмі. Визначено ступінь іонізації розпорощених атомів від параметрів плазми. Розглянуто вплив пеннінговської іонізації на баланс заряджених частинок у плазмі.