

ESTIMATING THE PARAMETERS OF A POSITIVE COLUMN OF THE HALOGEN-CONTAINING GLOW DISCHARGE AT MODERATE PRESSURES

The methodical recommendations for estimating the plasma parameters of an electronegative glow discharge containing halogens at moderate pressures (up to 40 Torr) with the use of simple analytic formulae and without numerical modeling are given. The initial data are easily measurable discharge parameters such as a discharge current, a voltage and a gas mixture pressure and composition as well. It is shown how one can easily consider such important plasma features as non-Maxwellian electron energy distribution function and halogen molecules dissociation by electron impact. As a result, such plasma parameters as the absolute degree of electronegativity, the value of border coordinate between ion-ion and electron-ion plasmas, and the forms of transversal profiles of electron and negative ion concentrations can be evaluated. The comparison of the results with the ones given by a global numerical model shows the suitability of said analytic approach to estimating plasma parameters of real discharges.

GAS DISCHARGE, ELECTRONEGATIVE GAS, HALOGEN, POSITIVE COLUMN.

Collision processes between charged particles taking place in the plasma of a positive column (PC) of the direct current (DC) glow of electronegative (EN) discharge at low and moderate pressures (up to 40 Torr) are of undoubted interest. In Ref. [1], simple analytic expressions connecting the elementary collision rates to each other and to energetic and geometric parameters of transversal profiles of charged particles density were presented. This could be very useful for practical calculations of PC plasma parameters since this makes it possible to calculate the values of mentioned plasma parameters fast, easily and without any numerical modeling.

But in fact, the declared in Ref. [1] improvement of the adequacy of theoretical treatment of EN discharge was reduced to introducing ion diffusion into the theory (indeed, it had not been taken into consideration before). The problem of practical estimation of PC plasma parameters was virtually omitted in

Ref. [1]; therefore it remains uncertain how exactly the theory [1] describes the properties of the real discharges rather than the model ones only.

The present paper offers the technique of calculation of the parameters used in the theory. The technique is based on initial, easily measurable discharge characteristics such as a current, a voltage and gas composition. As the result, it allows us to estimate the values of some important PC plasma parameters which cannot be measured directly. Hence, the present work completes the theoretical paper [1], expanding their practical application area.

Moreover, one of the present work goals is to check the accuracy of PC plasma parameters calculations for real discharges by using analytical expressions obtained in Ref. [1].

In the present paper, the same notation symbols are used as are in [1]: ν_i , ν_a are ionization and attachment frequencies; ρ_i is an ion-ion recombination constant;

$$v = v_i / D_{ap}^*; \quad \alpha = v_a / D_{an}^*; \quad \eta = n_{e0} \rho_i / D_{ap}^*;$$

$$D_{ap}^* = \mu_p T_e / (ex_W^2); \quad D_{an}^* = D_{ap}^* / \mu;$$

T_j, μ_j are temperatures and mobilities of the particles of the kind j ; indices e, p, n correspond to electrons, positive ions and negative ions; $\mu = \mu_p / \mu_n$; x_W is an absolute wall coordinate, and for cylindrical geometry (CG) $x_W = R_i$ (R_i is an inner radius of the discharge tube); n_{e0} is electron density in the discharge center; X is a reduced dimensionless transverse coordinate, in particular, for CG $X = r / R_i$; X_0 is the reduced coordinate of the boundary between e - i and i - i plasmas [1]; $\tau_j = T_j / T_e$; $\tau_s = \tau_p + \tau_n$; $n(X) = n_e(X) / n_{e0}$; $N(X) = n_n(X) / n_{e0}$; N_0 is the ratio of negative ion and electron densities in the discharge center (at $X = 0$), i. e. $N(0)$ at $\tau_s \rightarrow 0$; N_{τ_s} is the same quantity as N_0 but it is taken at $\tau_s > 0$.

From the expressions obtained in Ref. [1] which relate to the values of α , η and v , it could be hypothetically possible to calculate the electron temperature T_e or even the E/N (the ratio of the longitudinal electric field to the concentration of neutral particles), how it can be made from Schottky's relation for electropositive plasmas [2]. But the information on T_e is not highly profitable for the further calculations, particularly for the estimations of characteristics of plasma radiation, because the electron energy distribution function (EEDF) in plasma containing molecular gases is non-Maxwellian. Checking calculations of the power of excimer UV radiation for the discharge in the mixture of Xe и Cl₂ made by model [4] (if we let the EEDF be Maxwellian there) gave the UV-power values of about 2 – 3 times lower than the ones obtained experimentally. And since the EEDF is non-Maxwellian, the problem of E/N calculation becomes as complex as a full-scale numerical global discharge model is. It is irrelevant to pose such a problem having a purpose to reach the maximal simplicity of the estimations of transversal density profiles of charged particles.

But if we prescribe the value of E/N as an input parameter, together with the concentrations of neutral components of gas mixture as it will be shown below, the estimation of

plasma parameters of glow EN discharge becomes a relatively simple task wherein no any numerical model is needed.

The value of E/N for discharges in halogen-containing gas mixtures can be taken from the published data [5 – 7] or estimated experimentally, e. g. by the movable electrode method as it was made in Ref. [5 – 7].

If E/N and gas mixture composition are known, it makes sense to use the free software BOLSIG [8]. There both EEDF and D_e, μ_e, v_a, v_i , as well as the other electron collision frequencies, can be calculated by numerical solution of the Boltzmann kinetics equation. It is the simplest way to take the non-Maxwellian EEDF into consideration and hence to improve the adequacy of all following results.

With the known values of μ_e and E/N , it is possible to estimate the n_{e0} value from the measured discharge current, keeping in mind that the transversal electron density profile at strong electronegativity is almost flat in the great part of the discharge cross section [1, 3, 4] (see also Fig. 1, *b* at the end of this paper).

Ionic temperatures, also corrected for ion heating by electric field, can be calculated by using expressions given in Ref. [9]; ion mobilities can be taken from the data of Ref. [10], and ion-ion recombination rates for halogens ρ_i – from Ref. [11 – 13].

Estimating real EN discharge parameters, we should take into account the reduction of halogen molecules concentration (relative to their concentration without the discharge) due to molecule dissociation caused by electron impact taking place even at small discharge currents [4]. Let us describe the iterative procedure of estimating halogen molecules concentration through the example of a discharge in the mixture of Xe и Cl₂.

All the iterations were made with the use of the above-mentioned free software BOLSIG. For the first iteration, we set the initial gas composition as the filling one. After closing run we should take from BOLSIG output data the sum of excitation collision frequencies for $B^1\Pi, B^3\Pi$ and $C^1\Pi$ states of the Cl₂ molecule (in the sum they give the total electron impact dissociation frequency). In BOLSIG, all the collision frequencies are normalized to the total gas mixture concentration of $3.5 \cdot 10^{16} \text{ cm}^{-3}$; so

we should divide the said sum by $3.5 \cdot 10^{16}$ and by the relative fracture of Cl_2 to obtain the rate coefficient for molecule dissociation $\langle \sigma_d v_e \rangle$ in cm^3/s . Then, from the expression [14]

$$N_{\text{Cl}_2} = \frac{A + 8BC - \sqrt{A^2 + 16ABC}}{8C} \quad (1)$$

we can find the concentration of remaining chlorine molecules.

In the expression (1),

$$A = n_{e0} \langle \sigma_d v_e \rangle, \quad B = N_{\text{Cl}_2,0} 293 / T_g,$$

$$C = K_{ar} N_{\text{Xe}0} 293 / T_g,$$

where T_g is gas temperature in K; $N_{\text{Xe}0}$, $N_{\text{Cl}_2,0}$ are concentrations of xenon and molecular chlorine in cm^{-3} at filling (at 293 K);

$$K_{ar} \cong 6,2 \cdot 10^{-29} / T_g^{3/2} \quad (\text{cm}^6/\text{s}) \quad [4]$$

is a volume recombination coefficient for chlorine atoms in the gas mixture with the predominance of inert gas.

As the input value for the first iteration, we used $N_{\text{Cl}_2,0}$, then EEDF and $\langle \sigma_d v_e \rangle$ were found, and finally we took the new value of N_{Cl_2} . But the EEDF depends on the molecular chlorine concentration. So, we should continue the iterative process until we achieve the stable values of N_{Cl_2} , D_e ; μ_e , v_i and v_a as well. For the second iteration, we should set the initial gas composition with N_{Cl_2} obtained at the first one.

Strictly speaking, for each (except the first) iteration we should take into account the appearance of atomic chlorine in the gas mixture. This would require complementing the BOLSIG database where the atomic chlorine cross-sections are missing initially; these sections can be taken from Ref. [14, 15]. But the helpful moment is that the electron collision cross-sections for chlorine, including the ionization cross-section, are close to the ones for heavy inert gases. Checking calculations shows that if chlorine concentration is 5 or more times lower than that of the inert gas, the introduction of atomic chlorine in the model affects the values of α , v and D_{ap}^* very weakly and only leads to their variations of no more than 4 %. In such a case, the presence of atomic chlorine can be neglected.

After the run of the second iteration with

new gas composition, one should again calculate $\langle \sigma_d v_e \rangle$ and then N_{Cl_2} using formula (1). For the third iteration, the value of N_{Cl_2} obtained at the second one should be taken as the initial and so on.

As it was mentioned in Ref. [4], the described iterative process converges very quickly. The third iteration leads to changes in N_{Cl_2} , D_e , μ_e , v_i and v_a values no more than 6 % compared with the results of the second iteration, and the fourth (in comparison with the third) does no more than the fractions of one percent. So, the values of D_e , μ_e , v_i and v_a , obtained even at the second iteration could potentially be used for estimating D_{ap}^* , α , v , η , τ_p and τ_n (see Table 1).

Since BOLSIG outputs the separate values of D_e and of μ_e , the value of T_e , which is necessary to calculate τ_p and τ_n , should be obtained from Einstein's relation as eD_e / μ_e .

Then, we need to find the criterion χ_0 [1]:

$$\chi_0 = \sqrt{\frac{v + \alpha}{N_0 \tau_S}} X_0, \quad (2)$$

which value determines the kind of ionic diffusion.

For this purpose, we firstly need to find D_{ap}^* . To do this, the equation

$$v - \frac{\alpha}{3\mu} \cdot \frac{1 + X_0 + X_0^2}{X_0^2} \approx \frac{2}{X_0(1 - X_0)} \quad (3)$$

should be solved numerically where the value

$$X_0 \approx \left[\frac{48v_a}{\mu\rho_i n_{e0}} \left(\frac{D_{ap}^* \tau_S}{v_i + \mu v_a} \right)^2 \right]^{1/6}$$

is to be substituted.

Here it is pertinent to note that the expression (3) is valid for cylindrical geometry, and in the present paper only this case is analyzed. The relevant expressions for the plane geometry were presented in Ref. [1].

Furthermore, the found D_{ap}^* value has to be substituted into

$$\chi_0 = \frac{1}{\sqrt{N_0}} \left(\frac{48v_a}{\mu\rho_i n_{e0}} \sqrt{\frac{v_i + \mu v_a}{D_{ap}^* \tau_S}} \right)^{1/6},$$

where

$$N_0 = \sqrt{\frac{v + \alpha}{(\mu + 1)\eta}} = \sqrt{\frac{v_i / \mu + v_a}{(\mu + 1)\rho_i n_{e0}}}. \quad (4)$$

Table 1

The comparison between numerical results of calculations obtained in previous and present works

Parameter	Parameter value			
	Model data [4]	Present work		
		$k = 2$	$k = 3$	$k = 4$
D_{ap}^*, s^{-1}	$1.14 \cdot 10^5$	$1.24 \cdot 10^5$	$1.33 \cdot 10^5$	$1.31 \cdot 10^5$
ν	30.3	32.1	30.2	30.4
α	5.83	6.29	5.51	5.58
η	$1.10 \cdot 10^{-3}$	$1.02 \cdot 10^{-3}$	$9.51 \cdot 10^{-4}$	$9.66 \cdot 10^{-4}$
N_{τ_0}	113.3	129.0	127.0	126.0
X_0	0.92	0.91	0.91	0.90
β	0.018	0.018	0.017	0.017

The notes: (a) Parameter k means the number of iterations made for the refinement of N_{Cl_2} value. (b) β is used to estimate the electron profile in the discharge core [1].

If the calculated value $\chi_0 \leq 1.9$, we have the case of strong ionic diffusion (SD), otherwise it is the case of moderate diffusion (MD). In the last case, one needs to find the value of D_{ap}^* once more through solving numerically for D_{ap}^* the equation

$$\frac{\alpha}{\mu} (1 + X_0 + X_0^2) \approx \approx 3 \frac{\nu + \alpha}{\mu + 1} \cdot \left(\frac{N_0 \tau_s}{\nu + \alpha} + X_0^2 - \frac{3}{\sqrt{2}} X_0 \sqrt{\frac{N_0 \tau_s}{\nu + \alpha}} \right),$$

where

$$X_0 \approx \sqrt[3]{1 - 6 \frac{1 + \tau_p}{\nu - \alpha / \mu}},$$

then to calculate χ_0 again (according to Eq. (2)).

If χ_0 is near 1.9 in both cases, it is of no importance what kind of ionic diffusion is accepted. As it was demonstrated in Ref. [1], using formulae either for SD or for MD yielded similar results in comparison both with each other and with the “numerical experiment” (about that see Refs. [1, 16]).

After determining the kind of ionic diffusion and obtaining the values of ν , α , η , τ_p , τ_n , μ , it is possible (quite easily and without any numerical modeling) to estimate, using expressions deduced in Ref. [1], the following plasma parameters:

(a) The degree of electronegativity N_{τ_0} . For SD,

$$N_{\tau_0} = \frac{1}{4} \left(\frac{48\alpha}{\mu\eta} \cdot \frac{\nu + \alpha}{\tau_s} \right)^{1/3}$$

or (if $\alpha \geq 1$ and $\tau > 0.05$)

$$N_{\tau_0} \approx \sqrt{\frac{3\alpha}{\mu\eta}}.$$

For MD,

$$N_{\tau_0} \approx \sqrt{\frac{\nu + \alpha}{(\mu + 1)\eta}} \cdot \left[1 - \frac{1}{I_0(\sqrt{2}\chi_0)} \right],$$

where I_0 is the modified Bessel function of zero order.

(b) The border coordinate between i - i and e - i plasmas X_0 . For SD,

$$X_0 \approx \left[\frac{48\alpha}{\mu\eta} \left(\frac{\tau_s}{\nu + \alpha} \right)^2 \right]^{1/6};$$

for MD,

$$X_0 \approx \sqrt[3]{1 - 6 \frac{1 + \tau_p}{\nu - \alpha / \mu}}.$$

(c) Transversal profiles of negative ion concentration.

For SD,

$$N(\chi) \approx N_0 \frac{\chi_0^2}{4} \left[1 - \left(\frac{\chi}{\chi_0} \right)^2 \right],$$

for MD,

$$N(\chi) \approx N_0 \left[1 - \frac{I_0(\sqrt{2}\chi)}{I_0(\sqrt{2}\chi_0)} \right]. \quad (5)$$

Here $\chi = \sqrt{\frac{\nu + \alpha}{N_0 \tau_s}} X$.

(d) Transversal profiles of electron concentration. If $X \leq X_0$, then

$$n(X) \approx 1 - \beta X^2, \quad (6)$$

where

$$\beta = \frac{1}{4N_0} \frac{\mu\nu - \alpha}{\mu + 1},$$

and if $X_0 < X \leq 1$,

$$n(X) \approx (1 - X) / (1 - X_0). \quad (7)$$

Using the above expressions, we should keep in mind that all of them are deduced in Ref. [1] under the assumption that such parameters as ν , α , η , τ_p , τ_n are constant along the radial coordinate. This limits the area of practical applications of obtained results to discharges of small current densities (not more than 4 – 8 mA/cm²) where gas temperature is near the room one and E/N is roughly constant over the plasma cross-section. For example, the low-current stage of EN glow discharge in the mixture of inert gases and halogens [17 – 19]

satisfies the said conditions, and its study is of big practical interest because these discharges provide quite high efficiency of excimer UV radiation output (up to dozens of percent).

As an example, let us consider the estimation of PC parameters of a real discharge in the mixture of Xe and Cl₂ in a cylindrical tube of an inner radius of 0.6 cm. The values of E/N and discharge current were 105 Td (1 Townsend = 10⁻²¹ V·m²) and 2 mA respectively, $T_g \approx 350$ K, and $n_{e0} = 2.3 \cdot 10^9$ cm⁻³. These characteristics just correspond to the low-current discharge stage. Here $\rho_i = 5.5 \cdot 10^{-8}$ cm³/s and $\mu \approx 0.77$. By the program BOLSIG, it was found that $v_i^{(2)} \approx 3.98 \cdot 10^6$ s⁻¹, $v_a^{(2)} \approx 7.8 \cdot 10^5$ s⁻¹, and $N_{Cl_2}^{(2)} = 3.07 \cdot 10^{15}$ cm⁻³. As an illustration: $v_i^{(3)} \approx 4.02 \cdot 10^6$ s⁻¹, $v_a^{(3)} \approx 7.4 \cdot 10^5$ s⁻¹, $N_{Cl_2}^{(3)} = 2.98 \cdot 10^{15}$ cm⁻³. The upper index in parentheses shows the iteration number of above-mentioned iterative process of N_{Cl_2} refinement. Additionally (with the use of BOLSIG), it was obtained that $T_e \approx 6.05$ eV, $\tau_p \approx 0.014$ and $\tau_n \approx 0.018$.

The calculation of χ_0 value indicated the presence of moderate diffusion in the considered case rather than the strong one. The resulting profiles $N(X)$ and $n(X)$ are shown in Fig. 1.

In the Table, the numerical results of described calculations are presented. Parameter k

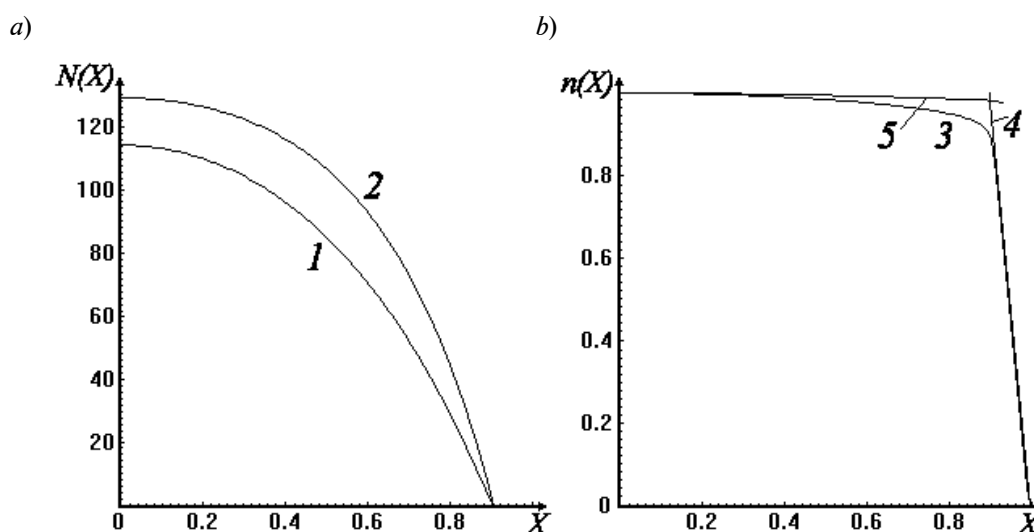


Fig. 1. Radial concentration profiles of negative ions (a) and electrons (b) in the plasma of an EN discharge. The calculations were carried out using the “global” numerical model [4] (curves 1 and 3), and formulae (4), (5) (curve 2), (7) (curve 4), (6) (curve 5).

The initial data for the calculations are given in the text

means the number of iterations made for refining N_{Cl_2} value which is affected by electron impact dissociation; β is used for the estimations of electron profile $n(X)$ at $X \leq X_0$ (see formula (6)), i. e. in the discharge core [1].

Most plasma parameters given in the table cannot be measured directly, so it is not possible to verify our calculations by any experimental data. Therefore, in the table and in Fig. 1 the results obtained from the complex numerical “global” model [4] for the same considered discharge are additionally presented for comparison. The model [4] can quantitatively predict not only discharge plasma character-

istics but also the experimentally measurable concentration profiles of excited atoms and molecules, and the characteristics of UV excimer radiation from the discharge as well. We can see from the table that such a comparison shows quite satisfactory agreement of the results given both by the “global” model and by simple analytical formulae.

As a conclusion, we can say that the presented analytical formulae can provide acceptable accuracy of estimations of plasma parameters of real EN discharges under conditions discussed above.

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Головицкий А.П. ОЦЕНКА ПАРАМЕТРОВ ПОЛОЖИТЕЛЬНОГО СТОЛБА ТЛЕЮЩЕГО РАЗРЯДА СРЕДНЕГО ДАВЛЕНИЯ, СОДЕРЖАЩЕГО ГАЛОГЕНЫ.

Даны методические рекомендации по оценке параметров плазмы положительного столба электроотрицательного тлеющего разряда низкого и среднего давления, содержащего галогены, при помощи несложных аналитических формул, исходя из внешних, легко измеряемых и контролируемых параметров разряда, и без привлечения сложного численного моделирования. Путем сравнения результатов аналитических расчетов с результатами глобальной численной модели названного разряда продемонстрирована пригодность использования аналитических выражений для оценок параметров плазмы реальных разрядов.

ГАЗОВЫЙ РАЗРЯД, ЭЛЕКТРООТРИЦАТЕЛЬНЫЙ ГАЗ, ГАЛОГЕН, ПОЛОЖИТЕЛЬНЫЙ СТОЛБ.

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