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INVESTIGATIONS OF THE DECAYING PLASMA OF ELECTRONEGATIVE GASES

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The influence of different processes on the decay of a low-pressure plasma containing negative ions is discussed. Experimental results and calculations modeling the decay of a multicomponent plasma are given. The problems of creation of ion-ion sources are considered.

Two viewpoints on the elementary processes occurring in the decaying plasma of electronegative gases exist at present. In the opinion of Smith et al. [1], when the gas pressure is low the decay is caused by the diffusion processes of a multicomponent plasma. The representatives of the other viewpoint [2] assume that the principal reaction is the attachment of electrons to molecularly excited molecules (of hydrogen). The supporters of both the first approach and the second one are close in stating the experimental fact: the current of negative electrons to the walls of a discharge tube grows in afterglow; both convincingly confirm the mechanism of this phenomenon by considerations and simple calculations. However, the question as to the nature of the phenomenon seems quite fundamental since such investigations will sooner or later be embodied in plasma devices, namely, in the sources of negative ions. The operating efficiency of the latter will depend on the correspondence of our knowledge to actual plasmachemical processes. Since both viewpoints appear quite convincing, one must conduct the experiment so as to unambiguously reveal the principal mechanism of plasma decay.

In this work, we have presented results of an investigation of a decaying low-pressure oxygen plasma [3]. The discharge was initiated in a cylindrical tube of diameter 3.4 cm and length 40 cm; the pulse-repetition rate was 1.5 kHz and the pulse period to pulse duration ratio (off-duty factor) was 10.

The plasma was diagnosed using a molybdenum cylindrical probe of length 0.35 cm and radius 0.005 cm. The measurements were carried out with an electronic device which made it possible to directly obtain the dependence $I(t)$. To do this, a point with a certain potential $eU >> kT$ was fixed on the volt-ampere characteristic and the behavior of the probe current in the phase of afterglow was tracked. Figure 1 gives $I(t)$ measured for a probe potential of 6.3 V; the oxygen pressure varied from 0.07 to 0.3 torr, while the discharge current was 80 mA. It is seen from the figure that the electron current arrives at the probe for a substantial period of time after the discharge pulse. As the gas pressure increases the electrons leave the discharge volume more slowly.

To describe a decaying plasma use is usually made of the balance equations, which in the case of a three-component plasma have the form

$$\partial n_e / \partial t + \mathrm{div}\, \Gamma_e = (v_i - v_a)\, n_e + v_d n_n - \beta_{ei} n_e n_p \,, \quad (1)$$

$$\partial n_p / \partial t + \mathrm{div}\, \Gamma_p = v_i n_e - \beta_{ei} n_e n_p - \beta_{ii} n_n n_p \,, \quad (2)$$

$$\partial n_n / \partial t + \mathrm{div}\, \Gamma_n = v_a n_e - v_d n_n - \beta_{ei} n_e n_p - \beta_{ei} n_e n_p \,, \quad (3)$$

where

$$\Gamma_j = - D_j \nabla n_j + (-1)^j\, b_j\, E n_j \,; \quad j = \mathrm{e, p, n} \,. \quad (4)$$

In what follows, we assume that the condition of quasineutrality of the plasma $n_p = n_e + n_n$ is fulfilled and ionization is absent from the afterglow. As is seen from system (1)–(3), the electron density decreases as a result of:

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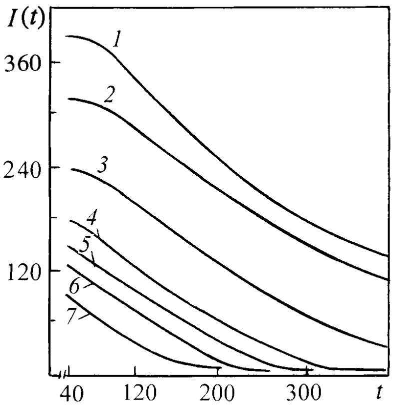


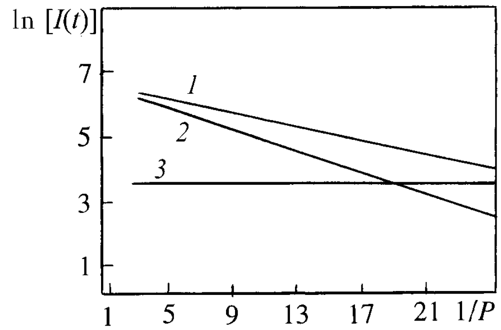


Fig. 1. Current of negative particles at different pressures of the neutral component: 1) 0.3, 2) 0.2, 3) 0.15, 4) 0.1, 5) 0.09, 6) 0.08, and 7) 0.07 torr. $I(t)$, μA; t, μsec.

Fig. 2. Dependence of ln $[I(t)]$ on $1/P$ of the electron current [1) t = 40 and 2) 100 μsec] and of the current of positive ions [3) t = 400 μsec]. ln $[I(t)/I(t_0)]$, rel. units; $1/P$, torr^{-1}.

(a) diffusional flux to the walls of the discharge volume, (b) recombination on positively charged particles, and (c) attachment to the particles of the gas with the formation of negative ions.

Let recombination be the main process of disintegration of electrons and div $\Gamma_e = n_e/\tau_e$; then $\partial n_e/\partial t + n_e/\tau_e = -\beta_{ei} n_e n_p$; after simple transformations we have

$$\frac{\partial n_e}{\partial t} = \left[\sqrt{\beta_{ei}(\alpha+1)}\, n_e + \frac{1}{2\sqrt{\beta_{ei}(\alpha+1)}\,\tau_e}\right]^2 - \left[\frac{1}{2\sqrt{\beta_{ei}(\alpha+1)}\,\tau_e}\right]^2 . \tag{1a}$$

When recombination predominates this means that the last term can be disregarded because of its smallness; consequently, by performing the integration

$$\frac{1}{\sqrt{\beta_{ei}(\alpha_0+1)}\, n_e(t) + \dfrac{1}{2\sqrt{\beta_{ei}(\alpha_0+1)}\,\tau_e}} - \frac{1}{\sqrt{\beta_{ei}(\alpha+1)}\, n_e(0) + \dfrac{1}{2\sqrt{\beta_{ei}(\alpha+1)}\,\tau_e}} = \sqrt{\beta_{ei}(\alpha+1)}\, t ,$$

we obtain the equation which relates $1/n_e$ and t by a linear law. If it is necessary to take into account all the quantities, integration yields

$$\frac{1}{2}\ln\frac{y+b}{y-b} = \sqrt{\beta_{ei}(\alpha+1)}\, t ,$$

where y corresponds to the first term on the right-hand side of (1a), while b corresponds to the second term.

If the main process is disintegration of electrons as a result of their attachment to neutral particles, the corresponding solution of the equation of balance of the electrons on the condition that $\nu_a(t)$ = const [4] has the form $n_e(t) = n_e(0)\exp(-\nu_a t)$ and the exponent increases either in proportion to P for the process of double attachment of the electrons or in proportion to P^2 for the process of triple attachment of the electrons to molecules.

When the principal mechanism of disintegration of the electrons is their diffusion, the characteristic time is in inverse proportion to the gas pressure [4].

Based on this analysis it seems possible to determine the dominant process of decrease in the number of electrons in a decaying plasma from the corresponding dependence on time. Thus, Fig. 2 plots ln $[I(t)]$ as functions of $1/P$. The points corresponding to the curves are measured at different pressures but, being taken at the same instants of time, fall well on a straight line, which corresponds to a decrease in the number of electrons as a result of diffusion.

Figure 3 gives the dependences $I(t)$ and ln $[I(t)]$; the gas pressure is 0.07 torr. The current of negative particles decreases monotonically, reaching a steady level at $t_0 > 190$ μsec. This means that in afterglow the electron density does not increase. When $t > t_0$ the probe volt-ampere characteristic becomes symmetric, i.e., by this time the ion-ion plasma is formed [6]. The time of its decay is determined by the codiffusion of the positive and negative ions:

$$\tau_{\text{n.p}} = \frac{R^2}{2.4^2 D_{\text{n.p}}}, \quad D_{\text{n.p}} = \frac{D_{\text{p}} b_{\text{n}} + D_{\text{n}} b_{\text{p}}}{b_{\text{n}} + b_{\text{p}}}. \tag{5}$$

The process of decay to t_0 is of greater interest. To describe the systems with a large number of elements one introduces the relaxation times of the system. Its evolution usually occurs in order of smallness of the characteristic time. As is seen from Fig. 3, such as approach is unsuitable for our problem. From a time of 40 μsec the electron component decays relatively slowly; then from approximately 130 μsec the electrons leave the discharge volume rather rapidly and after t_0 only the ion-ion plasma is left in the tube. A theoretical description of the processes occurring in the decaying plasma of electronegative gases can be found in [7]. Rozhanskii et al. assume the ionization-recombination processes to be absent and set zero boundary conditions for three sorts of charged particles. Since the radial distributions of all the components are in proportion to the Bessel function of zero order, for the electron density and the concentration of negative ions we obtain the nonlinear system

$$\frac{\partial n_{\text{e}}}{\partial t} = -(2.4/R)^2 n_{\text{e}} D_{\text{e}} \frac{2 b_{\text{p}} n_{\text{p}}}{b_{\text{e}} n_{\text{e}} + b_{\text{n}} n_{\text{n}} + b_{\text{p}} n_{\text{p}}}, \quad \frac{\partial n_{\text{n}}}{\partial t} = \frac{n_{\text{n}} D_{\text{n}}}{n_{\text{e}} D_{\text{e}}} \frac{\partial n_{\text{e}}}{\partial t}. \tag{6}$$

The condition of absence of the current to the tube walls $\Gamma_{\text{e}} + \Gamma_{\text{n}} = \Gamma_{\text{p}}$ determines the radial field

$$E = \frac{D_{\text{p}} \nabla n_{\text{p}} - D_{\text{e}} n_{\text{e}} - D_{\text{n}} n_{\text{n}}}{b_{\text{p}} n_{\text{p}} + b_{\text{e}} n_{\text{e}} + b_{\text{n}} n_{\text{n}}}. \tag{7}$$

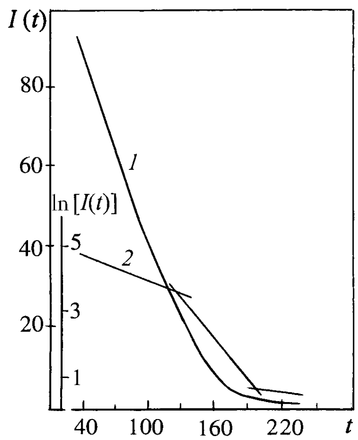
In the opinion of Rozhanskii et al. [7], as long as $n_{\text{e}} >> n_{\text{p}} b_{\text{p}}/b_{\text{e}}$ the decay is determined by the codiffusion of the electrons and ions which arrive at the wall; the flux Γ_{n} is small and the field (7) is close to a Boltzmann field for both the electrons and the negative ions. The plasma is depleted of electrons at this stage. At the end of it, when $n_{\text{e}} \approx n_{\text{p}} b_{\text{p}}/b_{\text{e}}$, the field (7) ceases to confine electrons and the rate of decrease of n_{e} becomes higher. The rate of decrease is determined by unipolar diffusion whose time constant is equal to

$$\tau = (R/2.4)^2 (1 + b_{\text{n}}/b_{\text{p}}) \frac{1}{2D_{\text{e}}}.$$

The flux Γ_{n} increases sharply; further decay is determined by ion-ion diffusion. The transition at $t_0 = 3.72$ msec observed in [1] should also be compared to the transition of the electron-ion diffusion to ion-ion diffusion. This instant and further decay correspond to a symmetric volt-ampere characteristic on the tube axis, while the region of the weak field, occurring at the electron stage of decay at the center of the tube and extending with time, corresponds to the region of the ion-ion plasma.

Rozhanskii et al. [7] note that the situation becomes significantly different if T_{e} in the plasma is even slightly higher that T_{n}, which can occur in volume recombination or destruction of metastable particles. Then, according to the solution of (6)–(7), at the electron stage of decay Γ_{n} turns out to be directed from the wall, i.e., as long as the temperature and concentration of the electrons are significant, the radial field rakes up the negative ions to the center of the tube and the region of the electron-ion plasma adjoins the wall.

This publication supplements the probe investigations presented in [5], since the cycle of measurements was performed under the same discharge conditions. The aim of the work is to determine plasma parameters and to study their evolution with time. According to the measurements, in the active phase of the discharge $T_{\text{e}} = 1.96$ eV and $\alpha = 1$; within 40 μsec after the discharge pulse $T_{\text{e}} = 0.03$ eV and $\alpha = 3$. The radial profile of the electrons in afterglow coincides with the Bessel function and $n_{\text{p}}(r)$ and $n_{\text{n}}(r)$ are close to it [5].



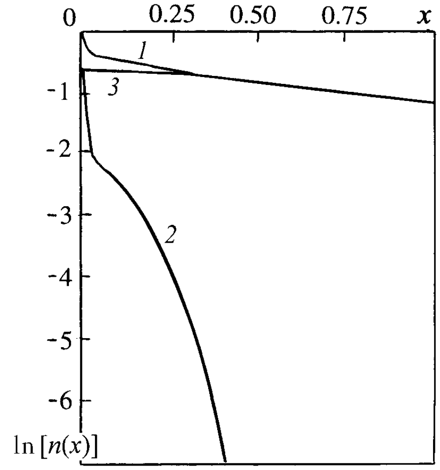


Fig. 3. Current of negative particles on the tube axis at P = 0.07 torr (1); ln [$I(t)$] (2). $I(t)$, μA; ln [$I(t)$], rel. units; t, μsec.

Fig. 4. Numerical calculations of the evolution of densities (relative units): 1) ln $[n_p(x)/n_p(0)]$; 2) ln $[n_e(x)/n_p(0)]$; 3) ln $[n_n(x)/n_p(0)]$.

To elucidate the influence of plasmachemical processes we performed model calculations. Since the discharge tube is of cylindrical symmetry, the radial profiles are close to Bessel ones throughout the decay, and there is no longitudinal field in afterglow, we reduce the computations to a one-dimensional problem. The expression for the diffusion coefficient of the corresponding particles is borrowed from [4]:

$$D_{a.p} = D_p \frac{(1+\gamma+2\alpha\gamma)\left(1+\alpha\dfrac{b_n}{b_e}\right)}{(1+\alpha\gamma)\left(1+\alpha\dfrac{b_n}{b_e}+(1+\alpha)\dfrac{b_p}{b_e}\right)}, \tag{8a}$$

$$D_{a.n} = D_p \frac{(1+\gamma+2\alpha\gamma)\left(\dfrac{b_n}{\gamma b_e}\right)}{1+\alpha\dfrac{b_n}{b_e}+(1+\alpha)\dfrac{b_p}{b_e}}, \tag{8b}$$

$$D_{a.e} = D_p \frac{1+\gamma+2\alpha\gamma}{1+\alpha\dfrac{b_n}{b_e}+(1+\alpha)\dfrac{b_p}{b_e}}. \tag{8c}$$

Unlike Rozhanskii et al. [7], we will not assume the plasma to be isothermal but will take the initial conditions n_p = 1, α = 1, and γ = 60, which correlate with the experimental data of [5], for the numerical solution of (1)–(3). Next we must determine $T_e(t)$, which is found from spectroscopic measurements. To do this we obtained the spectrogram of the discharge in its active phase; then we tuned the monochromator to the line of atomic oxygen (732.5 nm). The photons were recorded using a photomultiplier; the signal from it arrived at a multichannel accumulator [8] locked in step with the discharge. In this experiment, the accumulation step was 5 μsec and the number of cells was 64. In such a manner we carried out multiple scanning from the active phase to 310 μsec in afterglow. The time dependence of the number of photons has the form

t, μsec	0	5	10	15	20	25	30	35	40
N	28367	5723	1525	755	609	541	571	513	515

The scanning at $t > 40$ μsec revealed the fluctuation of N about the average value, which is related to the dark current of the photomultiplier. If we set $N = N_0 \exp(-t/\tau)$ at the initial stage of decay, then, by taking the logarithm of the tabulated data, we find $\tau = 3.4$ μsec. Let us assume that the luminosity of the line is in proportion to the electron temperature; then the latter decreases to 0.03 eV in 15–20 μsec. This enables us to introduce a complex temperature dependence for the diffusion (8a)–(8c). The calculation results are given in Fig. 4. The quantity $x = 2.4^2 D_p t/R^2$ is plotted on the x axis, while the logarithm of the concentration of the corresponding component is plotted on the y axis. As is seen from the figure, the decay of the plasma with negative ions should be subdivided into four stages.

At the first stage, we have a sharp decrease in T_e and in the density of the electrons and the positive ions for a constant value of the concentration of the negative ions. At this stage, just as in the active phase of the discharge, Γ_n is directed to the axis of the discharge tube. The modeling results are in good agreement with the theoretical description [7] and experimental data [1, 5]. These are primarily the shapes of the radial profiles of particles in the active phase of the discharge and early afterglow, which are leveled out on the tube axis and drop to the periphery more abruptly than the Bessel function, which corresponds to the blocking of the negative ions by the electron field.

The second stage is characterized by the joint drift of the positive ions and the electrons whose temperature is the same, in practice. Already at the 40th second of the decay the values of the densities were $n_p = 7.96 \cdot 10^9$, $n_n = 6.1 \cdot 10^9$, and $n_e = 1.86 \cdot 10^9$ cm^{-3}. This period is characterized by a substantially smaller slope of the logarithm of the concentration of the positive ions and the electrons than the previous portion, but here the condition $n_e \gg n_p b_p / b_e$ is additionally fulfilled. At the boundary of the first and second stages, we have the reversal of the flux of negative ions; now it is directed toward the wall and increases with α.

At the third stage, $n_e \ll n_p b_p / b_e$ and, as has been mentioned above, the electrons leave the plasma volume in the regime of unipolar diffusion [7]. Because of the decrease in the number of electrons the radial field decreases and the flux of negative ions sharply increases; correspondingly $\Gamma_n \to \Gamma_p$. We can easily find the boundaries of this stage, having plotted ln $[I(t)]$ by the experimental curve (see Fig. 3) or having drawn asymptotic straight lines on the corresponding plot of the model calculations (see Fig. 4).

The fourth stage is characterized by the insignificant influence of the electrons (which appear as a result of the reaction of detachment). This period of afterglow is an ion-ion electronless plasma. Correspondingly the time of its decay is determined from (5).

Comparing the modeling and the experimental results, we note that Fig. 4 shows ln $[n_j(t)]$ while Fig. 3 shows the logarithm of the sum of the electronic current and the ionic current; therefore, one can obtain only indirect information on the behavior of the latter at the first stage. Indeed, mass spectrometric measurements are required to determine the density of the negative ions in a nonequilibrium electron-ion plasma [1, 4]. Modeling makes it possible to find the dependence $n_n(t)$ and to compare experimental data on it in the discharge and afterglow. Thus, according to [5], $n_p = 1.51 \cdot 10^{10}$ and $n_e = 8.2 \cdot 10^9$ cm^{-3} in the active phase and $n_p = n_n = 4.485 \cdot 10^9$ cm^{-3} at the stage of the ion-ion plasma. The calculations show that the change in the density of the negative ions amounts to 35%, i.e., in the active phase, $n_n = 6.9 \cdot 10^9$ cm^{-3}. Thus, the model of formation of the probe layer in electronegative gases [5] turned out to be more accurate than the author himself assumed, while the numerical calculations constructed on the diffusion model reflect the process of decay most completely for these discharge conditions. The modeling revealed the presence of four stages and made it possible to determine the time of departure of the electrons from the discharge volume; this time was $x = 0.4$ (210 μsec), which is also in good agreement with experimental results.

A computation of the fluxes of charged particles to the tube walls enables us to obtain the final supplemented picture of decay of the three-component plasma. In Fig. 5, curve 1 describes the behavior of Γ_p, curve 2 describes the behavior of Γ_e, and curve 3 describes the behavior of Γ_n. At the initial stage of decay, there is virtually no current of negative ions to the tube wall, which corresponds to the blocking of the negative ions by the electron field (7). As T_e decreases and α increases, the flux Γ_n increases. It attains its maximum by the end of the third stage when $\Gamma_n \approx \Gamma_p$ by virtue of the quasineutrality condition. Thereafter $\Gamma_n(t)$ exactly follows the behavior of $\Gamma_p(t)$. The results of the numerical experiments presented in Fig. 5 are in agreement with the behavior of the experimental curves of $\Gamma_j(t)$ given in Fig. 7 in [1]; the only difference is in the time scale. Indeed, in [6], the approximation formula to find the time of departure of the electrons from the plasma volume is proposed:

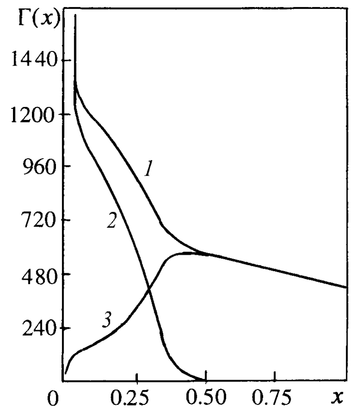


Fig. 5. Numerical calculation of the evolution of fluxes (rel. units): 1) $\Gamma_p(x)$; 2) $\Gamma_e(x)$; 3) $\Gamma_n(x)$.

$$t_0 = \tau_p \ln (1 + 1/\alpha_0)\,, \tag{9}$$

this time is determined by three parameters: the pressure of the neutral component, the initial degree of electronegativity α_0, and the radius of the discharge tube. Thus, for the gas-discharge conditions investigated in [1], the corresponding quantities are P = 0.05 torr, α_0 = 0.1, and R = 5.5 cm. With account for (9) and (8a) we obtain t_0 = 3.77 msec, which is in agreement with the results presented in [1]. It is precisely the fact that a sharp increase in the negative-ion flux is observed in afterglow that has led Katsch et al. [2] to the thought of a substantial increase in n_n in afterglow; Katsch et al. relate it to the dominant role of attachment. Thus, Katsch et al. [2] successfully explain both the disappearance of the electrons and the increase in T_n. But in this work we in no way took into account the reactions of detachment and attachment; however, the time of departure of the electrons (formation of an ion-ion plasma) is in good agreement not only with the results of [3, 6] but with the results of [1] as well.

Owing to the data of [1, 5, 9], we can propose a method of formation of the degree of electronegativity of the discharge. Thus, Thompson [9] finds the value of α = 20 (on the tube axis) in the steady discharge. In [1], α_0 constitutes tenth fractions, while in [5] α_0 = 1. It is easy to see the dependence of the value of α_0 on the off-duty factor of discharge pulses: it is equal to zero in [9], is of about ten in [5], and constitutes several hundreds in [1]. Such a behavior of α_0 is explained by the fact that in the steady discharge the negative ions are blocked in the plasma volume where their accumulation occurs; α is determined by the balance of attachment and detachment. Furthermore, in the pulse-periodic discharge of low pressure, the density is determined by the drift velocity, geometry of the discharge, and duration of the afterglow phase. On the other hand, the time of departure of the electrons depends on the quantity α_0 in accordance with (9). Thus, by decreasing the off-duty factor of the pulse discharge we decrease the value of α_0 and simultaneously make the departure of the electrons from the plasma volume more rapid. To elucidate the dependence of t_0 on the off-duty factor of the discharge we performed measurements in the afterglow of air at P = 0.1 torr and a discharge current of 32 mA; the pulse-repetition frequency was 1.4 kHz. We obtained the following dependence:

Duration of the active phase, μm	100	200	300
Time of departure of the electrons t_0, μsec	450	400	370
Longitudinal field in the active phase, V/cm	9.2	6.4	1.4

As the duration of the discharge current increases, t_0 decreases, which can naturally be related to the accumulation of the negative ions in the active phase (9). The decrease in the longitudinal field in the discharge is also related to the increase in the concentration of the charged particles. If we assume that the disappearance of the electrons is a consequence of activation of the attachment processes, the high value of the longitudinal field must stimulate these processes. Indeed, the principal reaction of formation of negative ions under low-pressure conditions is dissociative attachment. For oxygen the cross-section maximum is observed at 6 eV; the cross section of the reaction becomes larger and shifts toward lower energies only in the case of a significant increase in the gas temperature [4] where the energy of vibrationally excited molecules is involved in the reaction. But, first, the energy contribution decreases. Second, the experimental value of the ion temperature is 0.03 eV [10]; consequently, the neutral gas must have the same temperature. Third, when the attachment predominates, the decay is independent of α_0. Thus, the vibrational mechanism of formation of negative ions seems improbable and the decrease in the field is caused by the accumulation of the charged particles in the active phase. In the case of a hydrogen plasma the picture of decay can be more complex since both the particle mass and the energy of affinity to its molecule are substantially lower (3.8 eV). However, in the case of low pressures [2] the diffusion of the electrons to the walls with their subsequent recombination seems more probable. However, the criterion of truth is an experiment, and it is a great pity that Katsch et al. [2] did not prove the truth of their assumptions by conducting this experiment convincingly.

Having drawn such unexpected conclusions, one can give recommendations on the creation of ion-ion sources. First, one should use a pulse-periodic discharge in which departure of the electrons from the plasma volume occurs. Then the problem of separation of electrons and negative ions is solved, which makes it possible to save on devices that carry out this selection. Second, by varying the gas pressure, the current, and the off-duty factor and also the dimensions of the plasma volume, one can achieve the required value of n_n. On the other hand, in selecting negative ions α_0 decreases and in accordance with (9) the time of departure of the electrons t_0 increases since the system is nonlinear, which should also be taken into account in developing corresponding devices.

Thus, in the work the results of experimental measurements have been discussed and model calculations of the evolution of the densities of charged particles and their fluxes have been performed. The diffusional nature of the observed phenomenon has been substantiated. The mechanism of forming the degree of electronegativity α_0 has been proposed, and practical recommendations on the development of sources of negative ions have been given.

NOTATION

b_e, b_n, and b_p, mobility coefficients of the electrons and of the negative and positive ions; D_e, D_n, and D_p, diffusion coefficients of the electrons and of the negative and positive ions; E, radial field; e, electron charge; $I(t)$, saturation current as a function of time; k, Boltzmann constant; n_e, n_p, and n_n, density of the electrons and of the positive and negative ions; P, gas pressure; R, tube radius; r, running radius; U, potential of the probe; T, particle temperature; t, time; t_0, time of departure of the electrons from the plasma volume; $x = t/\tau_p$; $\alpha = n_n/n_e$; β_{ei}, coefficient of electron-ion recombination; Γ_e, Γ_p, and Γ_n, fluxes of the electrons and of the positive and negative ions; $\gamma = T_e/T_p$; ν_i, ν_a, and ν_d, frequencies of ionization, attachment, and detachment; τ_p, characteristic time of diffusion of the positive ions. Subscripts: 0, initial value; n, negative; p, positive; e, electronic; i, ionization; a, attachment; d, detachment; a.n, ambipolar diffusion of the negative particles; a.p, the same, of the positive particles, a.e, the same, of the electrons.

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