THE THEORETICAL MODEL OF CATAPHORESIS IN A PERIODICALLY PULSED d. c. GLOW DISCHARGE

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The simple theoretical model of cataphoresis in a pulse d. c. glow discharge is developed by using the equations for cataphoresis in the stationary glow discharge and the equations for ordinary diffusion in a cylindrical tube with endbulbs. These differential equations are solved numerically by means of the difference schemes for various discharge parameters. The results are presented in the non-dimensional form which can be applied for any discharge conditions.

ТЕОРЕТИЧЕСКАЯ МОДЕЛЬ КАТАФОРЕЗА В ПЕРИОДИЧЕСКОМ ИМПУЛЬСНОМ ТЛЕЮЩЕМ РАЗРЯДЕ

В работе предложена простая модель катафореза в периодическом импульсном тлеющем разряде, использующая уравнения катафореза для стационарного тлеющего разряда и уравнения для обычной диффузии в цилиндрической трубке с дополнительными объёмами при электродах. Приведено численное решение дифференциальных уравнений для разных параметров разряда, используя метод сеток. Результаты представлены в безрозмерной форме, и могут быть использованы для любых параметров разряда.

I. INTRODUCTION

It is well known that when a d. c. glow discharge is produced in the gas mixture, a separation of the gas constituents is observed. Many authors studied experimentally and theoretically this phenomenon (for more references see [1]). Deutsch was the only one [2, 3] who investigated experimentally cataphoretic processes in the pulse and alternate discharges. In our previous work [4] we also observed by the experimental indirect method the cataphoresis in the pulse d.c. glow discharge. So far nobody has described this process theoretically.

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In the present paper the axial separation in a pulse d. c. glow discharge is theoretically explained. The simplified assumptions regarding relaxation processes (connected with ion currents) and radial effects are used in the formulation of the problem. Equations for cataphoresis in the stationary glow discharge and equations for ordinary diffusion in the cylindrical tube with endbulbs are used to obtain the simple model. Equations of cataphoresis in the periodically pulsed d. c. glow discharge are solved by using difference schemes described in this paper. The calculation have been made for various discharge parameters and for various volumes of the endbulbs of discharge tube.

II. FORMULATION OF THE PROBLEM

We assume that in the cylindrical glass tube containing a Penning gas mixture the periodically pulsed d. c. glow discharge is excited. The cataphoresis effect is caused by the drift current of the admixture ions to the cathode. Our pulse discharge current is "ideal", rectangular, of the duration "τ", i.e., we have neglected the relaxation processes of the electric field and admixture ions density. The repetition period of pulses is "t'", i.e., the pulse frequency is $1/t'$. During the first pulse the admixture ions are moving along the tube to the cathode situated at $z = 0$ of the discharge tube. The result of the unidirectional flow of ions is the occurrence of the concentration gradient of admixture ions and atoms. This gradient leads to the backward diffusion of the neutral admixture atoms during the pulse and mainly in the afterglow period. The afterglow period, i.e. the time between two discharge pulses is not sufficient to make the admixture atom concentration uniform along the axis of the discharge tube [5]. The concentration gradient will only become weaker. During the next pulse the undirectional flow of ions again increases the concentration gradient of the admixture atoms. After the next interruption of the discharge the backward ordinary diffusion will appear again. The same processes will be continued in the next discharge pulses and their afterglow periods. In this way, gradually, the gas segregation can be obtained in a periodically pulsed d. c. glow discharge. The above mantioned processes may be described by the following continuity equations [6, 7, 8] for the admixture ion density $n_i (r, z, t)$:

$$\frac{\partial n_i}{\partial t} = D_i \frac{\partial^2 n_i}{\partial z^2} + D_a \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial n_i}{\partial r} \right) \right] + v_i \frac{\partial n_i}{\partial z} + R_i \tag{1}$$

and the neutral atom density $n(r, z, t)$:

$$\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial z^2} + D \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial n}{\partial r} \right) \right] - R_i \tag{2}$$

where the first terms on the right-hand side represent the ordinary diffusion of the ions and neutrals along the axis of a tube, the second terms represent the radial diffusion of ions or neutrals towards the wall. The third term in Eq. (1) represents a forced diffusion due to a uniform axial electric field and the last terms in equations (1) and (2) the rate of homogeneous ionization which is a source for ions (Eq. 1) and a sink for neutrals (Eq. 2). The quantities D_i, D and D_a are ionic, neutral and ambipolar diffusion coefficients, respectively and v_i is the drift velocity of ions. Eqs. (1) and (2) describe cataphoresis during the pulse discharge, but in the afterglow period the term $v_i\,\partial n_i/\partial z$ which represents the forced diffusion is equal to zero, i.e., in the afterglow periods these equations are identical with ordinary diffusion equations.

To solve Eqs. (1), (2) we shall proceed in the following way. In the case when the total effect of the radial terms can be neglected, we obtain by multiplying Eqs. (1), (2) by r, by integrating from the tube axis at $r=0$ to the wall at $r=R$ and adding these equations

$$\frac{\partial(\bar{n}_i+\bar{n})}{\partial t}=D_i\frac{\partial^2 \bar{n}_i}{\partial z^2}+D\frac{\partial^2 \bar{n}}{\partial z^2}+v_i\frac{\partial \bar{n}_i}{\partial z}.$$
(3)

The bars in Eq. (3) indicate radially averaged quantities. If we suppose that the level of ionization of the admixture atoms $\Theta=n_i/n$ is low (i.e. $\Theta\ll1$) and independent of z, then by neglecting the first term on the right side of Eq. (3), Eq. (3) can be written in the following nondimensional form:

$$\frac{\partial N}{\partial \tau}=\frac{\partial^2 N}{\partial \eta^2}+\alpha\frac{\partial N}{\partial \eta},$$
(4)

where $N(\eta,\tau)=n(z,t)/n_0$, n_0 is the initial admixture atom concentration uniformly dispersed in the discharge tube, $\tau=t\,D/L^2$ is the normalized time, $\eta=z/L$ is the normalized coordinate, L is the distance between the cathode and the anode in the discharge tube, $\alpha=\Theta v_i L/D$.

Equation (4) must be solved under the boundary conditions which represent also the influence of the endbulbs of the volumes V_1 and V_2 at the cathode and the anode, respectively. Normalized boundary conditions are [6, 7]:

$$\delta\frac{\partial N}{\partial \tau}=\frac{\partial N}{\partial \eta}+\alpha N \quad \text{at} \quad \eta=0,$$
(5)

$$-\varepsilon\frac{\partial N}{\partial \tau}=\frac{\partial N}{\partial \eta}+\alpha N \quad \text{at} \quad \eta=1,$$
(6)

where $\delta=V_1/\pi R^2 L$, $\varepsilon=V_2/\pi R^2 L$. The initial distribution is $N(\eta,0)=1$, the coefficient $\alpha\neq0$ during the pulse discharge, i.e., for the time $\tau\in\langle k\tau_p, k\tau_p+\tau_d\rangle$,

where τ_d is the normalized pulse duration, τ_p id the normalized pulse period, $k=1,2,...,n$ and $\alpha=0$ in the afterglow periods, i.e., for the time $\tau\in\langle k\tau_p+\tau_d,(k+1)\tau_p\rangle$.

III. SOLUTION OF THE CATAPHORESIS EQUATIONS BY THE FINITE DIFFERENCE METHOD

The finite difference method is very effective for the solution of Eqs. (4—6). Therefore the approximation of Eq. (4) was made by replacing the time derivatives by forward differences and the space derivatives by central differences. The truncation error of this approximation is $0(\Delta\tau+(\Delta\eta)^2)$. $\Delta\tau$, $\Delta\eta$ are meshwidths of the normalized time and space coordinates. The described approximation leads to the simple explicit difference formula for new density values at the time $\tau+\Delta\tau$ and at $\eta\in(0,1)$. The stability condition of this explicit difference formula is:

$$\Delta\tau\leq\frac{(\Delta\eta)^2}{2}.$$
(7)

The boundary conditions were approximated with the same precision in a similar way as that used by Crank [9]. Let Eq. (4) be valid at the boundary points, then two fictitious points are necessary for the approximation of this equation at the distance $\pm\Delta\eta$ from the electrodes. The values of N at these points are obtained from the difference approximations of Eqs. (5) and (6). The final forms of the difference schemes are:

$$N(0,\tau+\Delta\tau)=N(0,\tau)+\frac{\Delta\tau\left[\dfrac{2}{(\Delta\eta)^2}N(\Delta\eta,\tau)-N(0,\tau)\left(\alpha^2-\dfrac{2\alpha}{\Delta\eta}+\dfrac{2}{(\Delta\eta)^2}\right)\right]}{1-\alpha\delta+\dfrac{2\delta}{\Delta\eta}}$$
(8)

$$N(1,\tau+\Delta\tau)=N(1,\tau)+\frac{\Delta\tau\left[\dfrac{2}{(\Delta\eta)^2}N(1-\Delta\eta,\tau)+N(1,\tau)\left(\alpha^2+\dfrac{2\alpha}{\Delta\eta}+\dfrac{2}{(\Delta\eta)^2}\right)\right]}{1+\alpha\delta+\dfrac{2\varepsilon}{\Delta\eta}}.$$
(9)

From the stability analysis by means of the matrix method [10] it follows that if $2\alpha<1/\Delta\eta$, then expression (7) is also the stability criterion for all values of quantities α, δ, ε used in our calculations. The concrete errors such as truncation errors were controlled by dividing the space and time meshwidths. To obtain errors in our calculation smaller than 2 % it was sufficient to divide the space interval to 50 steps. The time steps were determined by expresion (7).

IV. RESULTS AND DISCUSSION

Using the difference method we have solved first the cataphoresis equations for the stationary glow discharge, because these results could be compared with their analytic solutions [6, 7]. Our results obtained by the numerical way are practically identical with the analytic results.

In the cataphoresis studies it is important to know the admixture atom distribution in the steady state, i.e. after a long time. All the following results of the steady state distributions are referred to the end of the afterglow period. For the interpretation of the time dependences of the admixture atom densities at the electrodes it is useful to use the time constants of the cataphoresis τ_m. In this paper the time constant of the cataphoresis τ_m is the time when the admixture atom concentration attains the value of 0.63 % of the value $(N_c - 1)$; N_c is the admixture atom concentration at the cathode in the steady state.

In the calculations we have used the normalized period of the pulses $\tau_d = K_1\Delta\tau$, the normalized duration of the discharge pulse $\tau_p = K_2\Delta\tau$, where K_1, K_2 are nature numbers. If these numbers are changed in the intervals $K_1 \in \langle 1; 60\rangle$, $K_2 \in \langle 2; 120\rangle$ with the ratio $t/t' = \tau_d/\tau_p = K_1/K_2$ constant, the resulting gradient of the admixture atoms slowly decreases if K_1 and K_2 simultaneously increase. For the limiting values of K_1, K_2 from our used intervals and at the ratio $K_1/K_2 = 1/2$, the calculated steady state concentrations at the electrodes differ by less than 1 %. The extreme values of these constants were not examined. In all the following results $K_2 = 40$.

In Fig. 1 there is shown the steady state distribution of the admixture atom density along the discharge tube between the cathode and the anode. It is seen that the gas separation is directly proportional to the pulse duration. The resulting profiles are exponential with the parameter $\alpha_{eff} = \alpha t/t'$:

$$N(\eta, \infty) = N_c \exp\{-\alpha_{eff}\eta\},$$ (10)

where N_c is the concentration of the admixture atoms at the cathode. Then the value of N_c can be determined from a mass balance equation:

$$\delta N_c + N_c \int_0^1 \exp\{-\alpha_{eff}\eta\}\, d\eta + \varepsilon N_c \exp\{-\alpha_{eff}\} = 1 + \delta + \varepsilon.$$ (11)

The solution of Eq. (11) gives:

$$N_c = \frac{1 + \delta + \varepsilon}{\delta + \varepsilon \exp\{-\alpha_{eff}\} + \dfrac{1 - \exp\{-\alpha_{eff}\}}{\alpha_{eff}}}.$$ (12)
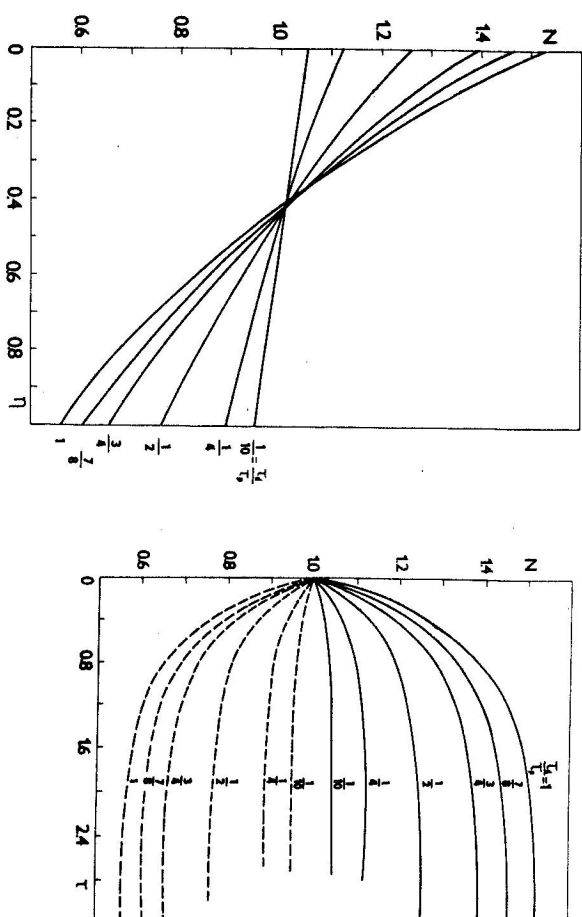

Fig. 1. Steady state distribution of the admixture atom density along the discharge tube for various pulse durations ($\alpha = 1$, $\delta = \varepsilon = 1$).

Fig. 2. Density of the admixture atoms at the cathode (full lines) and at the anode (dashed lines) as a function of time for various pulse durations ($\alpha = 1$, $\delta = \varepsilon = 1$).

The values of N_c calculated from Eq. (12) for various α_{eff} are in good agreement with the values N_c obtained by the numerical method, small differences are in the third significant digit.

It is interesting to note that the resulting composition of the mixture is the same as the initial one, at the distance $\eta = 0.42 - 0.49$ from the cathode for various pulse durations (Fig. 1). This result may be obtained also by using Eq. (10). The coordinate η_1 of the point where the composition is unchanged satisfies the following equation:

$$N_c \exp\{-\alpha_{eff}\eta_1\} = 1.$$ (13)

This equation gives:

$$\eta_1 = \frac{\ln N_c}{\alpha_{eff}}.$$ (14)

The values η_1 were calculated from Eq. (14) for N_c obtained from Eq. (12). η_1 decreases if α_{eff} increases ($\alpha = 1$ const.) from $\eta_1 = 0.49$ ($\alpha_{eff} = 1/10$) to $\eta_1 = 0.42$ ($\alpha_{eff} = 1$).

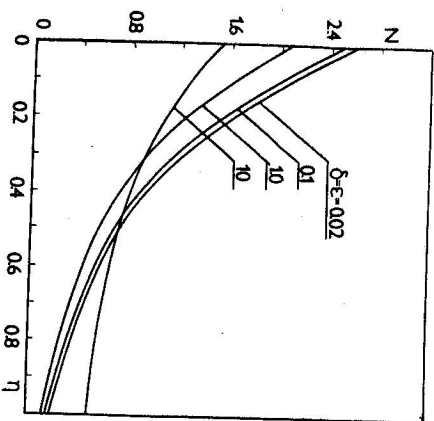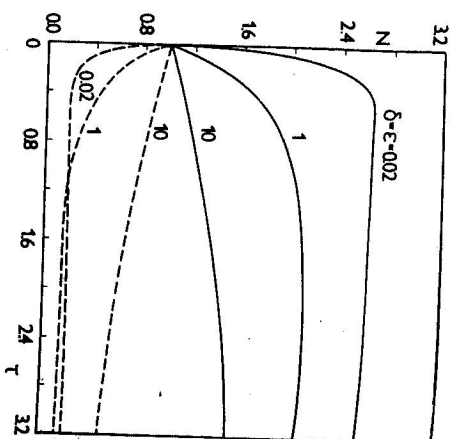Therefore, from the results obtained by the numerical integration of the cataphoresis equations in the pulse d.c. glow discharge, we can conclude that the resulting profiles of the admixture atoms are the same as the profiles in the stationary d.c. glow discharge with the parameter $a_{eff} = a\tau_d/\tau_p = at/t'$.

The admixture atom concentrations at the cathode and the anode as functions of nondimensional time are shown in Fig. 2 for various pulse durations. It can be seen that after a certain time (nearly the same for various pulse durations) the gas separation attains the steady state distribution. Then the time constants of the cataphoresis in the pulse d.c. glow discharge τ_m are determined by the dimensions of the discharge tube and are practically independent of the pulse duration.

The ratios δ and ϵ can be changed in many ways. We refer our results to the changes of δ and ϵ by changing V_1 and V_2, respectively if the volume of the discharge tube equal to $\pi R^2 L$ is constant, or to the changing of the diameter of the discharge tube R if V_1, V_2 and L are constant. The case when δ and ϵ are changed by elongating the discharge tube length L is not taken into account, because in this case also the normalized variables τ, η, a are changed. In Figs. 3 and 4 there are shown the steady state distribution and the time dependences, respectively, of the admixture atom densities at the cathode and at the anode for various volumes of the endbulbs. In the case of $\tau_d/\tau_p = t/t' = 7/8$ the effect of the endbulbs is similar to that in the stationary glow discharge. The steady state kconcentration is decreasing at the cathode and increasing at the anode, respectively, if the volume of the
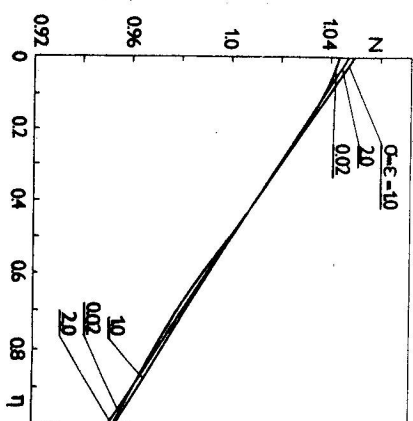
Fig. 3. Steady state distribution of the admixture atom density along the discharge tube for various volumes of the endbulbs ($a = 3$, $\tau_d/\tau_p = 7/8$).

Fig. 4. Density of the admixture atoms at the cathode (full lines) and at the anode (dashed lines) as a function of time for various volumes of the endbulbs ($a = 3$, $\tau_d/\tau_p = 7/8$).
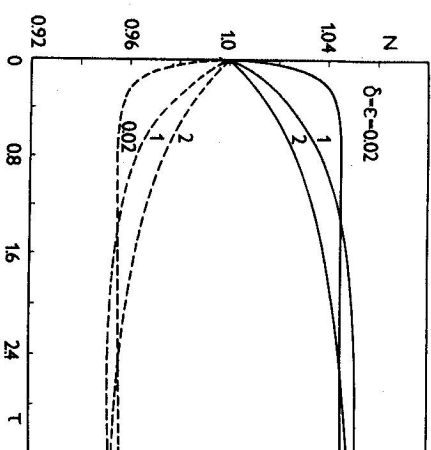
Fig. 5. Density of the admixture atoms at the cathode (full lines) and at the anode (dashed lines) as a function of time for various volumes of the endbulbs ($a = 1$, $\tau_d/\tau_p = 1/10$).

Fig. 6. Steady state distribution of the admixture atom density along the discharge tube for various volumes of the endbulbs ($a = 1$, $\tau_d/\tau_p = 1/10$).
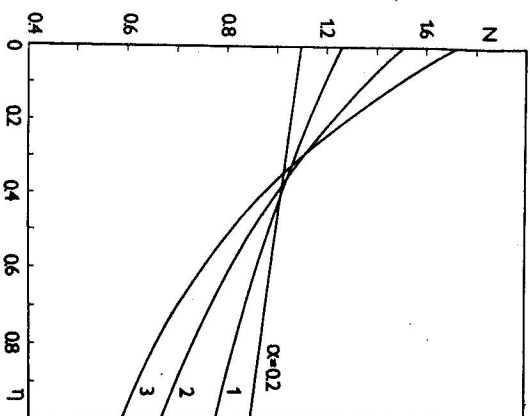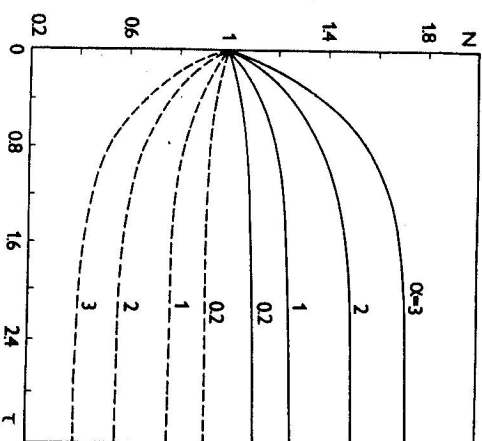

Fig. 7. Steady state distribution of the admixture atom density along the discharge tube for various values of the parameter a ($\delta = \epsilon = 1$, $\tau_d/\tau_p = 1/2$).

Fig. 8. Density of the admixture atoms at the cathode (full lines) and at the anode (dashed lines) as a function of time for various values of the paremeter a ($\delta = \epsilon = 1$, $\tau_d/\tau_p = 1/2$).

endbulbs increases. The time constant of cataphoresis is directly proportional to the quantities $\delta = \varepsilon$. For the short pulse duration $\tau_a/\tau_p = 1/10$, τ_m is also directly proportional to $\delta = \varepsilon$ (Fig. 5), but other effects at the electrodes are presented (Fig. 6) in the steady state profiles of the admixture atom density. For $\delta = \varepsilon > 1$ the steady state profiles are similar to those of the long pulse duration but for smaller values of $\delta = \varepsilon$, for example $\delta = \varepsilon = 0.02$, the derivative of the admixture atom density profiles at the electrodes becomes low. It seems that the ordinary diffusion has in such cases a strong influence on this profile.

The effect of the parameter α is the same as in the stationary glow discharge. With increasing α the final gas separation of the admixture atom density gradient increases (Fig. 7). The dependences of the admixture atom density at the cathode and at the anode on time for various α are shown in Fig. 8. The time constant of the cataphoresis in the pulse d. c. glow discharge decreases if the parameter α increases.

V. CONCLUSION

By using explicit difference schemes we have been able to solve the equations for cataphoresis in periodically pulsed d. c. glow discharge. The calculations have confirmed the assumption that also in a pulse d. c. glow discharge excited in the gas mixture there exists a gas segregation of gaseous constituents and the created concentration gradient depends on the pulse duration. This phenomenon is important in any equipment which uses a pulse d. c. glow discharge excited in the gas mixtures, e. g., gas lasers, expecially in afterglow measurements.

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