PHOTOEMISSIVE CHARGING OF MONODISPERSED AEROSOL PARTICLES UNDER ATMOSPHERIC PRESSURE

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The photoemissive charging of liquid aerosol particles that settle at atmospheric pressure has been studied theoretically and experimentally. Results have been compared, and satisfactory agreement has been achieved.

The determination of the magnitude and sign of the charge of an aerosol particle interacting with ultraviolet radiation is a timely problem because these parameters determine the course of important processes, such as the development of a thunderstorm in the atmosphere and the form of the trajectories of motion of charged particles in electromagnetic fields.

The present studies are devoted to theoretical and experimental consideration of processes of photoemissive charging of monodispersed aerosol particles.

We determine the applicability of a quasistationary approximation for the photoemissive charging of spherical particles that are settling under atmospheric pressure. In [1], it was shown that the charge carriers in the given situation are represented by free electrons and ions.

We estimate the relaxation time for the concentration field of the charge carriers around a particle.

The process of charging the particles in the presence of their own electrostatic field contains two components of charge transfer: diffusion and drift [2-5]; therefore, the maximum value of the relaxation time can be estimated as the sum of the relaxation time of the diffusion process and the drift time.

To determine these times, one should know the characteristic distance covered by the charge carriers. We define the characteristic distance as the thickness of a diffusion boundary layer, which for molecular diffusion near a spherical surface located in the flow is of the form [6]

$$d = Br\,\mathrm{Re}^{-\frac{1}{2}}. \quad (1)$$

Here B = 0.3 is a coefficient, $\mathrm{Re} = 2v_s r/\nu$.

We estimate the relaxation time assuming that the charge is carried by O_2^- ions. The electron mobility is considerably higher; therefore, the electron relaxation time is less. We determine the diffusion relaxation time [7]:

$$\tau_{\mathrm{dif}} = \frac{d^2}{4D_0}, \quad (2)$$

the value of D_0 is adopted from [8]:

$$D_0 = \frac{u_0 kT}{e}, \quad (3)$$

U_0 is defined in [8] as

$$u_0 = 0{,}815\,\frac{el}{m_0 v_0}. \quad (4)$$

We define the second component of the relaxation time, the drift time of the O_2^- ions:

$$\tau_{\mathrm{dr}} = \frac{d}{v_{\mathrm{dr}}}, \quad (5)$$

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where [8]

$$v_{\text{dr}} = u_0 E_{\text{sur}}. \tag{6}$$

We determine the total relaxation time additively:

$$\tau = \tau_{\text{dif}} + \tau_{\text{dr}}. \tag{7}$$

The time during which a particle stays in the flare zone is equal to

$$t_{\text{f}} = \frac{L}{v_{\text{s}}}. \tag{8}$$

Numerical estimates according to Eqs. (2), (5), (7), and (8) show that for the characteristic parameters of the process $\tau \sim 10^{-5}$ sec, $t_{\text{f}} \sim 10^{-2}$ sec.

Therefore, the inequality $t_{\text{f}} \gg \tau$ holds. This allows one to use a quasistationary approximation when solving the problem on photoemissive charging of settling particles. Thus, for cases $Q < 0$ and $Q > 0$, in [1] it has been obtained that

$$I = 4\pi R^2 Eune - 4\pi R^2 Df(\text{Re}, \text{Sc})\frac{dn}{dR}e, \; Q < 0; \tag{9}$$

$$I = -4\pi R^2 Eune - 4\pi R^2 Df(\text{Re}, \text{Sc})\frac{dn}{dR}e, \; Q > 0. \tag{10}$$

Integration of Eqs. (9) and (10) with boundary conditions $R \to \infty$ and $n \to 0$ and with the binding condition of flows at the surface of the particle $I = I_{\text{s}} - 4\pi r^2 ven_{\text{sur}}$ gives an expression, for the photocurrent from the surface of spherical particles depending on the magnitude of their charges, which in the region of the negative and positive charges is of the form

$$I = \frac{QuI_{\text{s}}}{4\pi r^2 v\varepsilon_0\left(1 - \exp - \dfrac{Qu}{4\pi rDf(\text{Re}, \text{Sc})\varepsilon_0}\right) + Qu} \; Q < 0, \tag{11}$$

$$I = \frac{QuI_{\text{s}}}{4\pi r^2 v\varepsilon_0\left(\exp \dfrac{Qu}{4\pi rDf(\text{Re}, \text{Sc})\varepsilon_0} - 1\right) + Qu} \; Q > 0. \tag{12}$$

When studying photoionization processes in dispersed systems, it is important to know the explicit dependence of the particle charge on time. To do this, we represent the current intensity from the surface of a negatively charged particle as the rate of change of its charge $I = -dQ/dt$. With regard to the accepted notation, Eq. (11) assumes the form

$$\frac{dQ}{dt} = -\frac{QuI_{\text{s}}}{4\pi r^2 v\varepsilon_0\left(1 - \exp - \dfrac{Qu}{4\pi rDf(\text{Re}, \text{Sc})\varepsilon_0}\right) + Qu}. \tag{13}$$

The minus sign in Eq. (13) indicates that photoemission results in the loss of the negative charge by the particle.

A numerical estimation of values on the right-hand side of Eq. (13) shows that for the absolute values $Q \geqslant \dfrac{4\pi rDf(\text{Re}, \text{Sc})\varepsilon_0}{u}$ the exponent approaches rapidly zero and can be neglected. Integration of (13) with regard to this fact for the initial condition $t = 0$, $Q = Q_{\text{in}}$ gives the result

$$\frac{4\pi r^2 v\varepsilon_0}{uI_{\text{s}}}\ln\frac{Q_{\text{in}}}{Q} + \frac{Q_{\text{in}} - Q}{I_{\text{s}}} = t. \tag{14}$$

We can conclude from Eq. (14) that for the magnitudes of charges $Q \geqslant \dfrac{4\pi rDf(\text{Re}, \text{Sc})\varepsilon_0}{u}$ the kinetics of the process does not depend on diffusion, i.e., in this case the primary charge transfer from the surface of the particle is determined by the drift of electrons in the electrostatic field.

We consider the region of negative charges, the absolute value of which satisfies the inequality $0 \leqslant Q < \dfrac{4\pi rDf(\text{Re}, \text{Sc})\varepsilon_0}{u}$.

For the given magnitudes of Q, we expand the exponent on the right-hand side of Eq. (13) in a Taylor series with respect to the parameter $\frac{Qu}{4\pi r Df(\mathrm{Re},\mathrm{Sc})\,\varepsilon_0}$, and truncate it after the first three terms of the expansion.

By integrating (13) with initial conditions $t = 0$, $Q = Q_{\mathrm{in}}$, after some transformations we obtain the dependence of the charge on time in the explicit form

$$Q = \frac{8\pi Df(\mathrm{Re},\ \mathrm{Sc})\,\varepsilon_0\,[vr + Df(\mathrm{Re},\ \mathrm{Sc})]}{vu} - \left[\left(\frac{8\pi Df(\mathrm{Re},\ \mathrm{Sc})\,\varepsilon_0\,[vr + Df(\mathrm{Re},\ \mathrm{Sc})]}{vu} - Q_{\mathrm{in}}\right)^2 + \frac{16\pi D^2 f^2(\mathrm{Re},\ \mathrm{Sc})\,\varepsilon_0 I_{\mathrm{s}} t}{vu}\right]^{1/2}. \tag{15}$$

We obtain the rate of variation of the charge on a particle when $Q > 0$ by using (12):

$$\frac{dQ}{dt} = \frac{QuI_{\mathrm{s}}}{4\pi r^2 v\varepsilon_0\left(\exp\frac{Qu}{4\pi r Df(\mathrm{Re},\ \mathrm{Sc})\,\varepsilon_0} - 1\right) + Qu}. \tag{16}$$

We expand the exponent in the denominator of (16) for values of the charge in the range $0 \leqslant Q < \frac{4\pi r Df(\mathrm{Re},\mathrm{Sc})\,\varepsilon_0}{u}$ in a Taylor series, restricting ourselves to the first three terms of the expansion.

The subsequent integration of (16) with the use of the initial conditions for $t = 0$, $Q = Q_{\mathrm{in}}$ gives the dependence of the charge on time

$$Q = \left[\left(Q_{\mathrm{in}} + \frac{8\pi Df(\mathrm{Re},\ \mathrm{Sc})\,\varepsilon_0\,[rv + Df(\mathrm{Re},\ \mathrm{Sc})]}{vu}\right)^2 + \frac{16\pi D^2 f^2(\mathrm{Re},\ \mathrm{Sc})\,I_{\mathrm{s}}\varepsilon_0 t}{vu}\right]^{1/2} - \frac{8\pi Df(\mathrm{Re},\ \mathrm{Sc})\,\varepsilon_0\,[rv + Df(\mathrm{Re},\ \mathrm{Sc})]}{vu}. \tag{17}$$

For charges $Q \geqslant \frac{4\pi r Df(\mathrm{Re},\mathrm{Sc})\,\varepsilon_0}{u}$, it is advisable to solve Eq. (16) numerically. For sufficiently large negative Q on a droplet, when the photoemissive current becomes saturated, the change in the absolute value of the droplet charge after the droplet passes the flare zone is of the form

$$\Delta Q = I_{\mathrm{s}} t_{\mathrm{f}}. \tag{18}$$

Substituting here $I_{\mathrm{s}} = e\gamma\Phi$ in explicit form, we obtain the value of the quantum efficiency

$$\gamma = \frac{\Delta Q}{e\Phi t_{\mathrm{f}}}. \tag{19}$$

To check the above discussion of the course of the photoemissive process and equations, quantitatively describing the phenomenon under consideration, an experimental investigation of interaction of ultraviolet radiation with an aerosol particle has been conducted.

The investigation was conducted on the experimental set-up whose diagram is represented in Fig. 1. The set-up contains a vessel 1, serving as an electrostatic screen. The generator of monodispersed droplets 2 is assembled in the upper part of the vessel and is fed from the source of alternating voltage 2a. The initial charge on the droplets is varied by a constant voltage source 3, the magnitude of which is controlled by an indicator 3a. In the lower part of the vessel there is a collector that represents a "Faraday cylinder" 4. The current formed by the droplets is recorded by a current meter 5 and a recorder 5a.

A plate 6 covered by a two-layer vaseline—oil film serves to measure the droplet sizes.

The droplets from the generator of monodispersed droplets are irradiated by a source of ultraviolet radiation 8, fed from the supply unit 8a, through a window with a light filter 7, which can be shut off by the shutter 7a. Parameters of the flow of ultraviolet radiation are measured by the actinometer 9 located on the vessel wall opposite to the window and recorded by the device 9a. The train of droplets, formed by the generator of monodispersed droplets, passes through the vessel and settles out on the measuring electrode.

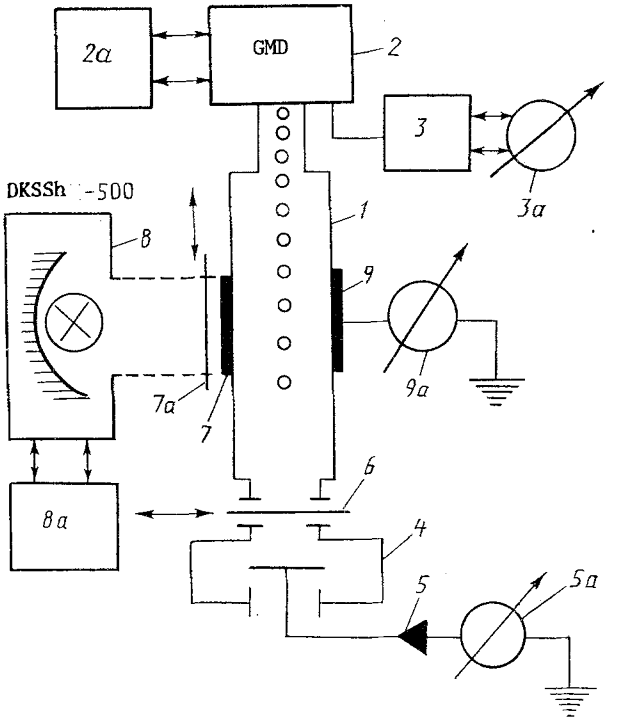

Fig. 1

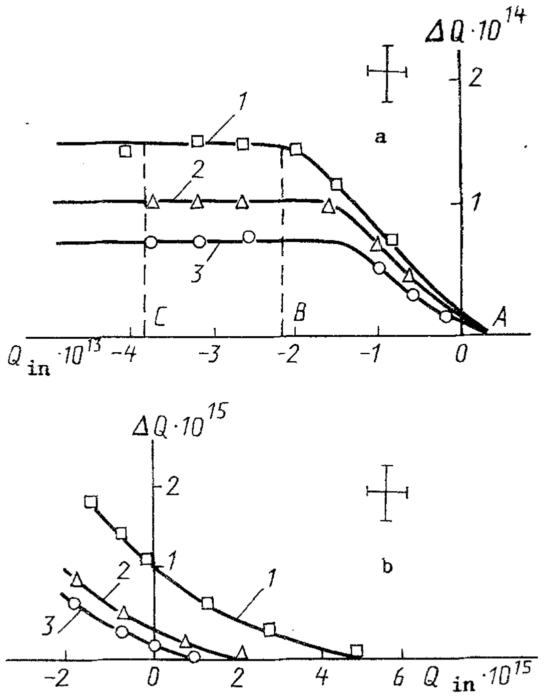

Fig. 2

Fig. 1. Diagram of the experimental set up.

Fig. 2. Experimental dependence of the photoemissive charge ΔQ on the initial charge Q_{in}: a) in the region of negative charges; b) in the region of positive charges for the droplets of aqueous solutions of dyes; 1) rhodamine 6G; 2) chrysoidine; 3) erythrosin (+ represents the maximum confidence intervals).

The current formed by the droplets that hit the measuring electrode was recorded with the help of the VK-2-16 electrometer.

The generator of monodispersed droplets generates N droplets per second, i.e., the current registered by the electrometer and produced by the droplet train $i = QN$, from which it follows that the value of the droplet charge is equal to $Q = i/N$.

The initial charge of a droplet of required size was created by means of a charging unit. While passing through the flare zone created by ultraviolet radiation, the train of droplets was changing the value of its initial charge. The change in the droplet charge due to photoemission can be found as $\Delta Q = Q_{in} - Q_{fin}$, where Q_{in} and Q_{fin} are the initial and the final charges of droplets, measured by the electrometer, respectively.

During the experiments a number of aqueous solutions of dyes were used.

The experimental results obtained are represented in Fig. 2 as dependencies of the emissive charge on the initial droplet charge $\Delta Q = f(Q_{in})$.

As is seen, the form of the curve is similar to the classic form of a volt-ampere characteristic obtained by using the method of the counter potential [8] when investigating a photoeffect.

The point 0 corresponds to the origin for a specified value of the initial droplet charge. The length OA describes the effect of the initial droplet charge $Q_{in} > 0$ on photoemission. It indicates that there is an electron flow returning to the surface, which increases with an increase in the initial droplet charge. The region of the curve OB characterizes the region of negative values of a specified droplet potential $Q_{in} < 0$.

As is seen from Fig. 2, when the negative droplet potential increases, the number of electrons leaving its surface increases. The region BC represents the saturation state of photoemission. This region is used to determine experimentally the quantum output with the help of Eq. (19).

By knowing how long the particle stays within the flare zone (8), the change in its charge during this time, which is determined experimentally, and the value of its initial

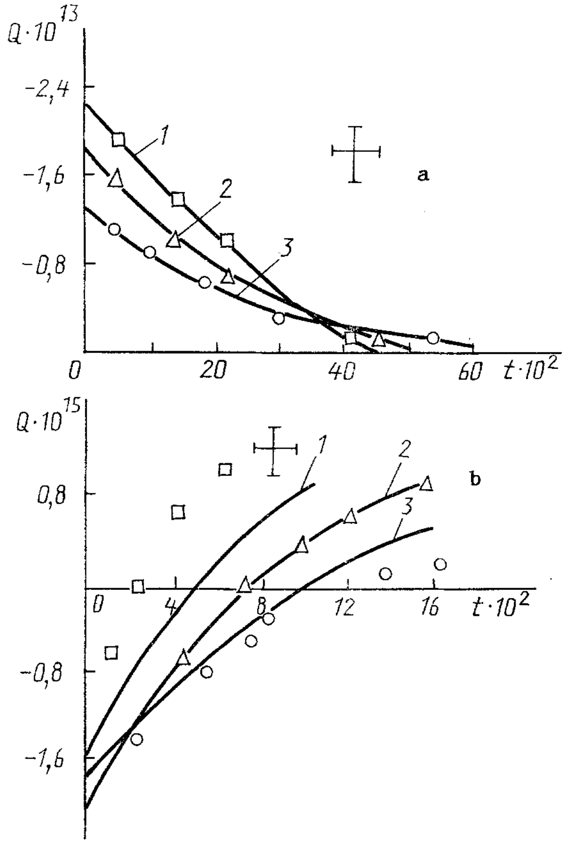

Fig. 3. Theoretical and experimental dependencies of the charge Q on time t. Notation is similar to that of Fig. 1.

charge Q_{in}, one can construct the dependence of the particle charge on time.

In Fig. 3, experimental dependencies of the particle charge on time in the region of negative and positive charges are given for droplets of solutions of different dyes r = 140 μm, which settled out through the zone of ultraviolet radiation with characteristic wavelength $\lambda = 240$ nm.

A comparison of experimental data with calculated data obtained according to Eqs. (14), (15), and (17) demonstrates that they agree well.

NOTATION

d, thickness of a diffusion boundary layer, m; r, particle radius, m; R, radius of a concentric sphere circumscribed around the particle, m; ℓ, mean free path of the oxygen ion, m; L, length of the flare zone, m; λ, wavelength, m; v_s, particle settling rate, m/sec; v_{dr}, drift rate of the oxygen ions, m/sec; v, one fourth of the average absolute rate of the oxygen ions, m/sec; v_o, average quadratic rate of the oxygen ions, m/sec; f(Re, Sc), wind factor; Re, Reynolds factor; Sc, Schmidt criterion; ν, kinematic air viscosity, m^2/sec; t, time, sec; t_f, flare time, sec; τ_{dif}, diffusion relaxation time, sec; τ_{dr}, drift time of the oxygen ions, sec; τ, total relaxation time, sec; m_o, mass of the oxygen ion, kg; u_o, mobility of the oxygen ions, $m^2/(V \times sec)$; u, electron mobility, $m^2/(V \times sec)$; n, volumetric electron concentration, m^{-3}; n_{sur}, concentration of the oxygen ions near the particle surface, m^{-3}; D_o, diffusion coefficient of oxygen ions, m^2/sec; D, diffusion coefficient of electrons, m_2/sec; Q, particle charge, C; Q_{in}, initial charge of particle, C; Q_{fin}, final charge of particle, C; e, electron charge; ΔQ, change in the particle charge, C; ε_0, electric constant, F/m; E, electric intensity of the particle field, V/m; E_{sur}, intensity of electrostatic field near the particle surface, V/m; I, photocurrent from the particle surface, A; I_s, saturation photocurrent, A; i, current created by a train of drops, A; Φ, quantum flow toward the particle surface, $m^{-2} \cdot sec^{-1}$; γ, quantum efficiency; N, operating frequency of the droplet generator, sec^{-1}; k, Boltzmann constant, J/K; T, ambient absolute temperature, K.

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FREEZING OF GRANULES IN AN APPARATUS FOR CRYOGRANULATION OF LIQUID MATERIALS

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The numerical solution of the problem of cooling and freezing a spherical drop under conditions of convective heat transfer is studied.

Cryogranulation of liquid materials is the main element of cryodispersion technology for obtaining organic and inorganic powdered materials. The quality of the final product is determined, as a rule, by the degree of monodispersity of the obtained granules and the conditions under which the granules are frozen.

In cryodispersion technology, different types of drop generators [1, 2] as well as systems for maintaining and monitoring the pressure and temperature of the dispersed liquid are employed to obtain a flow of monodispersed drops.

Freezing of the flow of drops is one of the most important stages of cryodispersion technology. In the process of solidification and cooling of the drops of liquid, a crystalline structure is formed, and this ultimately determines the quality of the product. In the process the sizes of the crystals formed are determined by the rate of freezing. In addition, the rate of freezing also affects other specific properties of the granulated material (the possibility of segregation of the components of the solution and others).

Under the conditions of low-temperature treatment the monodispersity can be destroyed owing to merging of drops in flight as well as at the moment that the cooling agent arrives at the surface. To prevent this undesirable phenomenon it is proposed that the freezing of the drops in nitrogen vapor be organized so that there would be enough time for the particles to freeze before the cooling agent reaches the surface.

A diagram of the apparatus for cryogranulation of objects with freezing in a convective flow is shown in Fig. 1. The apparatus operates as follows. The working solution is injected under a fixed pressure from the preparation system 1 into a drop generator 2 and is forced through a draw plate. Oscillations of the piezoelectric element are superposed on the jet emerging from the drop generator, and in a certain range of frequencies the jet is broken up into monodispersed drops. The drops are then subjected to low-temperature treatment in the freezing unit 3. The drops are cooled with nitrogen vapors, whose temperature is close to the saturation temperature at atmospheric pressure. The frozen drops enter the cooled assembly 5. Low temperature is maintained in the freezing unit by feeding liquid nitrogen from a Dewar bottle 6 into a nitrogen jacket 4. In order to reduce heat inflow from the surrounding medium the unit is insulated with a layer of thermal insulation.

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