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ELECTRIC FIELD DEPENDENCE OF THE LIMITING CONCENTRATION OF AN EDF-SYSTEM

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Calculation results are reported for the dependence of the limiting concentration of microparticles under electrodynamic fluidization on the electric field intensity. The influence of the electrophysical properties of the surface layer of conducting particles on this dependence is discussed.

In [1] it is shown that for electrodynamic fluidization (EDF) of microparticles with finite conductivity their limiting concentration, with regard for the recombination mechanism of concentration limitation, is determined by τ_c/τ_σ, where τ_c is the time of contact of microparticles in collisions and $\tau_\sigma = \varepsilon\varepsilon_0/\sigma$ is the relaxation time of the charge. For microparticles having a high conductivity with $\tau_c/\tau_\sigma >> 1$ the limiting concentration is independent of σ and E and is $n_{cr} = 2n_{in}$. For microparticles with a relatively low conductivity and $\tau_c/\tau_\sigma << 1$

$$n_{cr} = 2n_{in}/(\tau_c/\tau_\sigma)\,. \tag{1}$$

In [1], for this case a dependence of n_{cr} on particle size is determined, from a comparison of Eq. (1) with experimental data the parameter σ is evaluated, and the conclusion is made that the electrophysical characteristics of the surface layer of metal particles is the governing factor for their limiting concentration.

We will investigate the electric field dependence of the limiting concentration of EDF-particles and its features determined by the properties of the surface layer of conducting microparticles.

Analysis of Eq. (1) at constant conductivity of the surface layer of microparticles shows that the limiting concentration of EDF-particles depends on E only through the parameter τ_c, whose value is determined by the relative velocity of colliding particles $\tilde{V}$ [2]:

$$\tau_c = 4.53\left[\frac{\sqrt{2}\,\delta m}{\sqrt{r\tilde{V}}}\right]^{2/5} \sim \tilde{V}^{-1/5} \tag{2}$$

or

$$\tau_c/\tau_\sigma = \tau_c\,(V_{q_{\max}})/\tau_\sigma\,(\tilde{V}/V_{q_{\max}})^{-1/5}\,, \tag{3}$$

where $V_{q_{\max}} = q_{\max}E/s$ is the velocity of microparticles with the charge $q_{\max}$; $\delta = (1-\mu^2)/\pi E_0$ is a constant of the material. In order to determine $\tilde{V}$, it is necessary to consider the special features of particle charge distribution for this case. As is shown in [1], the distribution function has a gap at $q = 0$ whose width is dependent on the ratio τ_c/τ_σ. For $\tau_c/\tau_\sigma << 1$, on attaining the limiting concentration the majority of particles are in the two charge states $\pm\bar{q}$ separated by a gap with the width

$$\Delta = \frac{1}{2}\,q_{\max}\,(\tau_c/\tau_\sigma)^2\,. \tag{4}$$

Therefore assuming $\bar{q} \simeq \Delta/2$, we obtain

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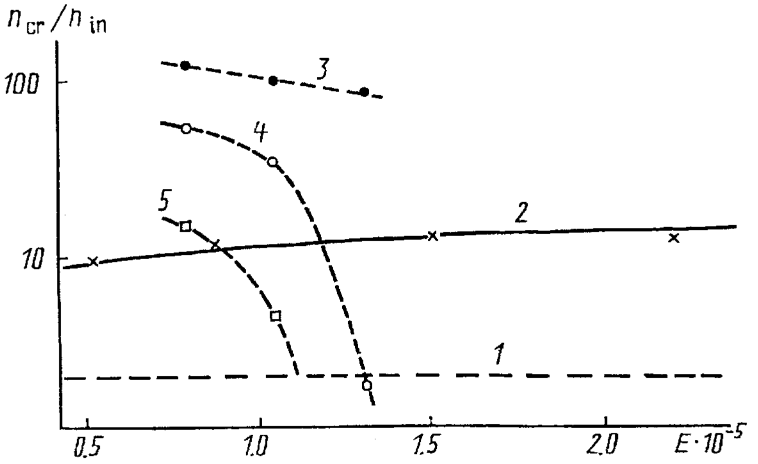

Fig. 1. Limiting concentration n_{cr}/n_{in} (rel. units) as a function of the electric field intensity E (V/m) for different materials: 1) theoretical dependence at $\tau_c/\tau_\sigma \gg 1$; 2) iron powder with a particle size of $r = 6$ μm; points, data of [3]; curve, according to Eq. (7); 3, 4, 5) stainless steel, bronze with $r = 7$ μm, and graphite with $r = 3.15$ μm, respectively [5].

$$\tilde{V} = 2\frac{\bar{q}E}{s} = 2V_{q_{\max}}\frac{\bar{q}}{q_{\max}} = \frac{1}{2}(\tau_c/\tau_\sigma)^2 V_{q_{\max}}. \tag{5}$$

Substitution of (5) into (3) gives the ratio τ_c/τ_σ expressed in terms of the time of contact τ_c $(V_{q_{\max}})$ at $\tilde{V} = V_{q_{\max}}$

$$\tau_c/\tau_\sigma = 2^{1/7}\,[\tau_c\,(V_{q_{\max}})/\tau_\sigma]^{5/7}. \tag{6}$$

Then, according to (1) the limiting concentration of the EDF-particles may be expressed as

$$n_{cr}/n_{in} = (E/E_1)^{2/7}, \tag{7}$$

where, with regard for the independence of the ratio $E/q_{\max}$ from the electric field, E_1 is determined as

$$E_1 = (2.27/\tau_\sigma)^{5/2}\,\delta m \left[\frac{sE}{rq_{\max}}\right]^{1/2}. \tag{8}$$

Thus, the limiting concentration of microparticles is proportional to $n_{cr} \sim E^{2/7}$ and it will rather slowly increase with electric field intensity. Correspondingly, at the same E the limiting concentration of particles will depend on their size as $n_{cr} \sim r^{-22/7}$ which virtually does not differ from the earlier obtained dependence $n_{cr} \sim r^{-3}$ [1].

Figure 1 shows the limiting concentrations obtained for iron powder particles with r = 6 μm [3] for $E = (0.5-2.2)\cdot 10^5$ V/m. In this electric field intensity range the limiting concentration is seen to have almost a two-fold increase. In [3], this growth is explained by involvement of increasingly fine particles, contained in the initial material, in the fluidization process as the electric field intensity increases. However, it is easy to estimate that for microparticles close in size the electric field intensity at which particles rise from the lower electrode, is smaller by an order of magnitude than the values used in [3]. This means that in this case the increase in the limiting concentration with the electric field intensity is caused by an increase in the charge of the microparticles and their relative velocity and a corresponding decrease in the time of contact.

To calculate (7) (Fig. 1), we have used the following parameters: $\mu = 0.28$; $E_0 = 20.6\cdot 10^{10}$ N/m; $d = 10^{-2}$ m. Assuming $\varepsilon \simeq 10$ [4], we obtain for the oxide layer $\sigma \simeq 5.7\cdot 10^{-4}$ S/m. This value is quite consistent with data of [4], where $\sigma \simeq 10^{-3}-10^{-5}$ S/m for iron oxides.

In deriving (7) it has been assumed that the charge recombination in collisions of particles proceeds with electrophysical characteristics of the material of the particles that are independent of the electric field. However other situations may also take place in fluidization of various materials. Figure 1 gives experimental data on the limiting concentration at different electric field intensities for fluidization of powders of stainless steel, bronze, and graphite [5]. As in the previous case, considering that the initial materials possess good conductivity, we may explain the relatively high limiting concentrations only by the effect of the surface layer particles. However, unlike [3], with increase in the electric field intensity the limiting concentration decreases to $n_{\mathrm{cr}}/n_{\mathrm{in}} \simeq 2$, and for different materials this decrease is different. We now consider the possible reasons for this phenomenon.

A characteristic property of stainless steel is the presence of a thin surface layer consisting of dopant oxides [6]. Therefore, one may expect that when differently charged particles collide, charge recombination is accomplished by tunneling through a double layer of an oxide-insulator. For the symmetrical metal-insulator-metal structure the tunnel current is [7]:

$$J_t = \frac{e}{2\pi h t^2}\left\{\left(\Phi_0 - \frac{eU_t}{2}\right)\exp\left[-\frac{4\pi t}{h}(2m_{\mathrm{ef}})^{1/2}\left(\Phi_0 - \frac{eU_t}{2}\right)^{1/2}\right] - \right.$$

$$\left. - \left(\Phi_0 + \frac{eU_t}{2}\right)\exp\left[-\frac{4\pi t}{h}(2m_{\mathrm{ef}})^{1/2}\left(\Phi_0 + \frac{eU_t}{2}\right)^{1/2}\right]\right\}, \tag{9}$$

where Φ_0 is the height of the potential barrier, t is the thickness of the barrier, and U_t is the potential difference across the barrier. With increase in this difference, the tunnel current increases highly. Thus, in collisions charge recombination proceeds at a rate dependent on the potential difference of the microparticles. For engineering estimation of this rate we assume that $J_t = \sigma_t U_t/t$ and $\tau_\sigma = \varepsilon_t\varepsilon_0/\sigma_t$. Use of the effective conductivity σ_t is motivated here by the circumstance that for $\tau_c/\tau_\sigma \ll 1$ the quantities U_t and J_t change only slightly in collisions of microparticles. Since the EDF-system at the limiting concentration represents two groups of particles with the charges $\pm\bar{q}$ and the gap width $\Delta \simeq 2\bar{q}$ also in this case, we may write the expression for the current of the EDF-system as

$$\bar{J} = 2\frac{n_{\mathrm{cr}}}{2}\bar{q}\bar{V}, \quad \bar{V} = \frac{\bar{q}E}{s}. \tag{10}$$

Then the potential difference of the colliding particles is

$$U_t = U_d\frac{\bar{q}}{q_{\max}}. \tag{11}$$

Thus, to calculate U_t, it is necessary to determine the charge $\bar{q}$, which is easy to do using experimental data on the field dependence of the current density for this material.

Using Eqs. (10) and (11) we obtain

$$\bar{q}/q_{\max} = (\bar{J}/J_{q_{\max}})^{1/2}, \quad J_{q_{\max}} = n_{\mathrm{cr}}\, q_{\max}^2 \frac{E}{s}. \tag{12}$$

If we measure simultaneously the limiting concentration and the current density, we may check the correctness of the assumptions by determining the width of the charge gap:

$$\Delta = \frac{1}{2}q_{\max}(\tau_c/\tau_\sigma)^2 = \frac{1}{2}q_{\max}\left(\frac{2n_{\mathrm{in}}}{n_{\mathrm{cr}}}\right)^2 = 2q_{\max}\left(\frac{n_{\mathrm{cr}}}{n_{\mathrm{in}}}\right)^{-2}, \tag{13}$$

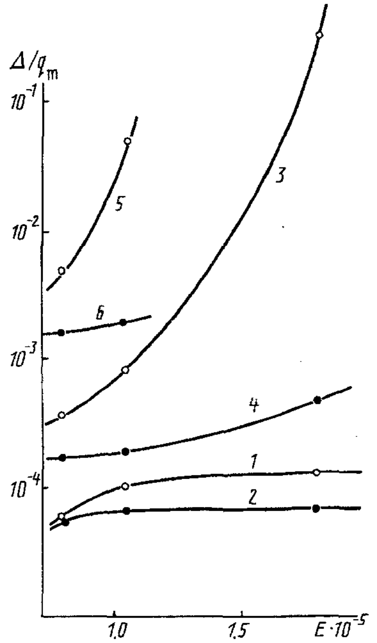

Fig. 2. Field dependence of the relative width of the charge gap Δ/q_m (rel. units) calculated from data of [5] using Eqs. (13) (1, 3, 5) and (14) (2, 4, 6) for stainless steel (1, 2), bronze (3, 4), graphite (5, 6). E, V/m.

$$\Delta = 2\bar{q} = 2q_\mathrm{max}\,(\bar{J}/J_{q_\mathrm{max}})^{1/2}, \tag{14}$$

where n_cr and $\bar{J}$ are taken from experiment.

As is seen (Fig. 2), in the range of electric field intensities used the gap widths determined by the current density and by the limiting concentration differ by less than a factor of two for $\tau_\mathrm{c}/\tau_\sigma \ll 1$. A comparison of the n_cr (E) and $\Delta(E)$ curves in Figs. 1 and 2 shows that the difference between the gap widths determined by (13) and (14) decreases with decrease in the $\tau_\mathrm{c}/\tau_\sigma$ ratio. This means that the expressions used are sufficiently correct if $\tau_\mathrm{c}/\tau_0 \ll 1$. Next, from the same experimental data, by determining σ_t in terms of the limiting concentration using (1), (2), and (5) and calculating U_t by (11) and (12), we obtain the effective conductivity of tunneling as a function of the tunnelling potential difference (Fig. 3). In the calculations we used $\rho = 7.8\cdot 10^3$ kg/m^3, $\mu = 0.28$, $E_0 = 20.6\cdot 10^{10}$ N/m, $\varepsilon_t = 12$ [4].

In plotting the theoretical dependence $\sigma_t(U_t)$ according to (9) the best approximation is attained at $t = 1.4\cdot 10^{-9}$ m, $\Phi_0 = 2.5$ eV for $m_\mathrm{ef} = m_e$. For the metal-insulator-metal system the height of the potential barrier is approximately equal to the halfwidth of the forbidden band of the insulator. For chromium oxide [4] $\varepsilon_g/2 = 1.7$ eV, and therefore the obtained estimate of Φ_0 with an allowance for a probable additional gap, consisting of gas molecules adsorbed by the surface of microparticles, seems quite acceptable. The thickness estimate $t/2 \simeq 7$ Å for the surface layer of microparticles confirms the monomolecular character of the oxide surface layer of stainless steel [6].

The field dependence of the limiting concentration for bronze particles [5] is more abrupt. For instance, when the electric field changes from 1 to $1.3\cdot 10^5$ V/m the value of $n_\mathrm{cr}/n_\mathrm{in}$ falls to $\simeq 2$. Unlike the previous case, the surface layer of particles possesses other properties. Assuming that it contains mainly copper oxides, representing a p-type semiconductor with the resistivity 10^4-10^5 Ω·cm [4] and forming a rectifying contact with the metal [8], we may assert that charge recombination in collisions of particles occurs in a circuit that consists of two opposite series-connected Schottky diodes. Here one of the junctions at the metal-oxide layer interface is always a barrier. In this case the density of the recombination current is determined by thermoelectronic emission over the barrier junction. According to [8], for back bias the density is

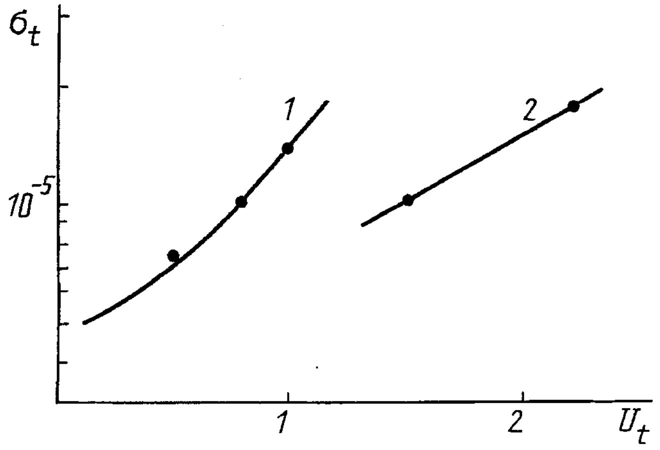

Fig. 3. Effective conductivity of the surface layer σ_t (S/m) as a function of the potential difference of the particles U_t (V) for stainless steel (1) and bronze (2): points, calculation using data of [5]; curves, fitting by Eqs. (9)

$$J_t = AT^2 \exp(-\Phi/kT), \quad \Phi = \Phi_0 - \left[\frac{e^7 (U_c + U_t) N_a}{8\pi^2 \varepsilon_t^3 \varepsilon_0^3} \right]^{1/4}, \quad \Phi_0 = \Phi_s - \Phi_m, \tag{15}$$

where $A = 4\pi e m_{ef} k^2/h^3$ is the Richardson constant; Φ_s, Φ_m are the work function of the semiconductor and the metal, respectively; U_c is the contact difference of potentials; N_a is the concentration of acceptors.

The increase the current density, in recombination, with the field is attributable here to the Schottky effect. Figure 3 shows $\sigma_t(U_t)$ calculated using data from [5] with $\rho = 8.7 \cdot 10^3$ kg/m^3, $\mu = 0.35$, $E_0 = 9.8 \cdot 10^{10}$ N/m for electric fields for which $\tau_c/\tau_\sigma = (2n_{in}/n_{cr}) << 1$. The theoretical dependence $\sigma_t(U_t)$ calculated by (15) is also given there.

The best approximation at $\varepsilon_t = 7.5$ [4], $T = 300$ K, and $m_{ef} = m_e$ is given by $t = 1.1 \cdot 10^{-8}$ m, $\Phi_0 = 0.85$ eV, $eU_c = 0.6$ eV, $N_a = 5.7 \cdot 10^{19}$ cm^{-3}. Hence we obtain $\Phi_s = 5.25$ eV at $\Phi_m = 4.4$ eV [9], which is consistent with data on the work function for copper oxides, namely, 4.9-5.3 eV [9]. A high concentration of acceptors is also typical of copper oxides [4] due to the available vacancies. The thickness of the oxide film is estimated as $t/2 = 50$ Å.

In [5] data are also reported for fluidization of a graphite powder with a particle size of $r = 3.15$ μm (see Fig. 1), whose limiting concentration tends to decrease as the field grows. This may be explained by the influence of the adsorbed layer of gas molecules on the charge recombination process. In [10], it is noted that with an increase of the concentration of adsorbed atoms of oxygen the conductivity decreases abruptly and the work function increases for graphite, thus indicating the appearance of charged chemisorption in an acceptor form. Therefore in this case charge recombination is also accomplished by tunnelling through the layer of adsorbed atoms.

Unfortunately, under the necessary condition $\tau_c/\tau_\sigma << 1$ for analysis we may take only one point at $E = 0.78 \cdot 10^5$ V/m. The corresponding value of U_t is 14 W at $\rho = 2.2 \cdot 10^3$ kg/m^3, $\mu = 0.4$, $E_0 = 2 \cdot 10^{10}$ N/m. Assuming the barrier height to be equal to the work function for graphite $\Phi_0 = 5$ eV and applying the Fowler-Nordgame formula for the density of the tunnel current for the case $\Phi_0 < eU_t$ [7] at $\varepsilon_t = 1$, $m_{ef} = m_e$, we obtain the thickness of the adsorbed layer $t/2 = 12$ Å.

It is obvious that data [3, 5] produced by studies of technical applications of the EDF-process are insufficient for all-round checking of the procedure of analysis of the surface structure of microparticles. This refers, first of all, to the choice of the materials to be studied. Therefore, in order to explore the field dependence of the EDF-system it is desirable to employ structures with well-known properties such as Al-Al_2O_3, Si-SiO_2, etc., in which the thickness of the oxide layer may be monitored in a wider electric field intensity range. Theoretically, it is necessary to directly calculate $n_{cr}(E)$ with different mechanisms of charge recombination in collisions of microparticles. Investigations of contact phenomena [7] run into the problem of creating symmetrical tunnelling transitions, which explains the poor reproducibility of results in the case of punching-in the current. In this connection the total symmetry of the charge recombination process in collisions of microparticles in the EDF-process and, correspondingly, the symmetrical structure formed in contact of microparticles are noteworthy. In addition, the relatively small time of contact prevents punching-in of the recombination current.

To sum up, measurement of the field dependence of the limiting concentration and the current of an EDF-system permits one to obtain characteristics of the surface layer of fluidized microparticles.

CONCLUSION

The mechanism by which charge recombination proceeds in collisions of microparticles exerts a decisive influence on the field dependence of the limiting concentration.

NOTATION

r, particle radius; d, interelectrode distance; $\beta = 4\pi r^2$, scattering section; $n_{\text{in}} = 1/\beta d$, initial concentration; τ_c, contact time; τ_σ, relaxation time of a charge; σ, conductivity; ε, ε_0, dielectric permittivity; E, electric field intensity; μ, Poisson coefficient; E_0, Young's modulus; q_m, maximum charge of a particle; $s = 6\pi\eta r$, resistance of a medium per unit velocity; η, medium viscosity; m, particle mass; m_{ef}, effective electron mass.

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