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ELECTRIFICATION IN TUBE FLOW OF ORGANIC LIQUIDS  
WITH AN ADMIXTURE OF STRONG ELECTROLYTE

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The electrification of weakly conducting organic liquids is investigated within the framework of continuum mechanics [1]. When such liquids (e.g., hydrocarbons) flow through tubes they acquire an electric charge [2-5]. This leads to the risk of electric discharges and explosions [4]. The conductivity of the considered liquids is due to a small amount of impurity electrolyte, whose molecules dissociate into positive and negative ions. According to recent ideas [2-5], the electrification of organic liquids is due to electrochemical reactions occurring on the tube walls, as a result of which the positive or negative ions of the impurity electrolyte are converted to neutral molecules. The problem of electrification of a liquid in the case of a completely dissociated electrolyte was examined in [5], where the effect of the electric field was ignored. The electrification of a weakly conducting liquid, in the case where the impurity electrolyte dissociates slightly and the dissociation can be regarded as an equilibrium reaction, was investigated in [6].

In this paper we examine the problem of electrification of an initially unchanged weakly conducting organic liquid in laminar flow in a metal tube, where the impurity electrolyte molecules are completely dissociated, and the electric field produced has a significant effect on electrification. The diffusion coefficients and charge numbers of the positive and negative ions are assumed to be equal. In addition, for definiteness we assume that only negative ions are involved in the electrochemical reactions on the tube walls, and the neutral molecules formed are present in excess in comparison with ions. The solution obtained can easily be extended to the case of arbitrary ion charge numbers and electrochemical reactions involving ions of both kinds on the tube walls.

1. The system of differential equations and boundary conditions at the tube entrance B and on the wall S, describing the electrification of an organic liquid, has the form

$$\begin{aligned} \operatorname{div} \left( n_{\pm} \mathbf{u} \pm \frac{ezD}{kT} n_{\pm} \mathbf{E} - DVn_{\pm} \right) &= 0, \\ \varepsilon \operatorname{div} \mathbf{E} &= 4\pi ez (n_{+} - n_{-}), \operatorname{rot} \mathbf{E} = 0, n_{\pm}|_B = n^0, \\ \left( \left[ \frac{ezD}{kT} n_{+} \mathbf{E} - DVn_{+} \right] \cdot \mathbf{v} \right) \Big|_S &= 0, \left( \left[ -\frac{ezD}{kT} n_{-} \mathbf{E} - DVn_{-} \right] \cdot \mathbf{v} \right) \Big|_S = K (n_{-} - n_{\omega}) \Big|_S, \end{aligned} \quad (1.1)$$

where  $n_{\pm}$  is the concentration of positive and negative ions;  $\mathbf{u}$ , liquid velocity;  $e$ , proton charge;  $z$ ,  $D$ , charge number and diffusion coefficient of the ions;  $k$ , Boltzmann constant;  $T$ , temperature of the liquid, which is assumed to be constant;  $\mathbf{E}$ , electric field;  $\varepsilon$ , dielectric constant of the liquid;  $n^0$ , concentration of positive or negative ions at the tube entrance;  $n_{\omega}$ , equilibrium concentration of negative ions at the tube wall, which is attained at the end of an infinitely long tube;  $K$ , constant of electrochemical neutralization of negative ions;  $\mathbf{v}$ , normal to the inside surface of the tube.

The last boundary condition (1.1) is fulfilled in the case where the neutral molecules formed by neutralization of negative ions on the tube walls are present in the solution in concentration  $n_s$ , which is much greater than the ion concentration. We then have the relation

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$$\left( \left[ -\frac{ezD}{kT} n_- \mathbf{E} - D \nabla n_- \right] \cdot \mathbf{v} \right)_S = K n_- - K_s n_s \equiv K (n_- - n_w),$$

in which  $K_S$  is the constant of the reaction that is the reverse of ion neutralization. Since, according to the condition  $n_S \gg n_-$ , even in the case of complete neutralization of all the negative ions the relative change in concentration  $n_S$  will be small, and it can be neglected. Hence, the quantity  $n_w \equiv K_S n_S / K$  in relations (1.1) is assumed to be constant. It is obviously equal to the negative-ion concentration at the tube walls in the equilibrium case, when  $K n_- = K_S n_S$ . Thus, according to the last relation in (1.1), there is a flux of negative ions, which decreases with increasing distance from the tube entrance, towards the tube wall, where they lose their charge. The neutral molecules formed pass into the solution and are removed by the flow of liquid, which ensures the steady-state nature of the electrification of the liquid in the tube, as is actually observed [2-4]. Electrification mechanisms leading to adsorption of neutral molecules on the tube walls will not, as we know [2-3], cause steady-state electrification. In a motionless liquid in a metal container an electrochemical reaction involving negative ions will also occur on the walls, of course. In this case, however, the ions will eventually acquire an equilibrium distribution, when their flux to the walls becomes zero. Immediately next to the walls  $n_- = K_S n_S / K$ , and at a distance from the walls much greater than the Debye radius  $\rho_d$  of the liquid, we have  $n_- = n^0$ . The electric field and charge density will be nonzero practically only at the walls of the container, in a layer of thickness  $\sim \rho_d$ . If  $K \gg K_S$ , then  $n_S \gg n_-$ . When the liquid from a container with characteristic dimension much greater than  $\rho_d$  is pumped through a narrow tube, the ion concentration at the tube entrance can be regarded as equal to the ion concentration in the bulk of the liquid in the container ( $n^0$ ). As a result, the negative ions lose their charge on the tube walls. With increase in distance from the tube entrance their concentration at the walls will tend to equilibrium, i.e., to  $K_S n_S / K \equiv n_w$ , and the electrification of the liquid will decrease.

From experimental data [2-4] for the electrification and conductivity of organic liquids we can estimate the maximum relative change in the concentration of ions involved in the reaction on the wall. In many cases it is much less than unity. The value of  $n_w$  contained in the boundary condition on the tube wall will differ little from the value of  $n^0$

$$\left| \frac{n_w - n^0}{n^0} \right| \gg 1$$

and in the considered problem there appears a small parameter

$$\gamma \equiv -\frac{n_w - n^0}{n^0} = -\frac{K_s n_s}{K n^0} + 1, \quad |\gamma| \ll 1,$$

determined by the constants  $K_S$ ,  $n_S$ ,  $K$ , and  $n^0$ , which characterize the physicochemical properties of the liquid undergoing electrification.

The effect of electrification of organic liquids on their flow can usually be neglected, and the characteristic settling length of the Poiseuille distribution of the liquid velocity in the tube is much smaller than the characteristic length  $L_0$  of the initial part of the tube, where the electrification of the liquid mainly occurs. Hence, for the liquid velocity  $u$  we can use the Poiseuille formula.

We introduce a cylindrical coordinate system  $(x, r, \theta)$ , whose  $x$  axis coincides with the tube axis. Converting to dimensionless variables in (1.1)

$$x^* = \frac{x}{R}, \quad r^* = \frac{r}{R}, \quad u^* = \frac{u}{u^0}, \quad \mathbf{E}^* = \frac{ezR}{kT} \mathbf{E}, \quad n_{\pm}^* = \frac{n_{\pm}}{n^0},$$

expressing the quantities  $n_{\pm}^*$ ,  $\mathbf{E}^*$  in terms of the small parameter  $\gamma$  ( $n_{\pm}^* = 1 + \gamma n_{\pm}^1$ ,  $\mathbf{E}^* = \gamma \mathbf{E}^1$ ) and linearizing Eq. (1.1), we find

$$\begin{aligned} \text{Pe} (1 - r^{*2}) \frac{\partial n_{\pm}^1}{\partial x^*} + \text{div}^* (\pm \mathbf{E}^1 - \nabla^* n_{\pm}^1) &= 0, \\ \text{div}^* \mathbf{E}^1 &= \frac{1}{2 \text{De}^2} (n_+^1 - n_-^1), \quad \text{rot}^* \mathbf{E}^1 = 0, \quad n_{\pm}^1|_B = 0, \\ [E_r^1 - (n_+^1)']|_S = 0, \quad [-E_r^1 - (n_-^1)']|_S &= K^* (n_-^1 + 1)|_S, \\ \text{Pe} = \frac{u^0 R}{D}, \quad K^* = \frac{KR}{D}, \quad \text{De} = \frac{1}{R} \sqrt{\frac{ekT}{8\pi e^2 n^0}}, \end{aligned}$$

where  $u^0$  is the velocity of the liquid on the tube axis;  $Pe$  is the Péclet number;  $De$  is the dimensionless Debye radius of the liquid; the dash denotes the derivative with respect to  $r^*$ .

Introducing the new variables  $q^* = n_+^1 - n_-^1$ ,  $n^* = n_+^1 + n_-^1$  we finally obtain the following system of equations and boundary conditions:

$$\begin{aligned} Pe(1-r^{*2})\frac{\partial q^*}{\partial x^*} - \Delta^* q^* + \frac{q^*}{De^2} &= 0, \\ Pe(1-r^{*2})\frac{\partial n^*}{\partial x^*} - \Delta^* n^* &= 0, \quad \text{div}^* E^1 = \frac{q^*}{2De^2}, \\ q^*|_{x^*=0} &= 0, \quad n^*|_{x^*=0} = 0, \quad \left[ E_r^1 - \frac{1}{2}(n^{*'} + q^{*'}) \right] \Big|_{r^*=1} = 0, \\ \left[ -E_r^1 - \frac{1}{2}(n^{*'} - q^{*'}) \right] \Big|_{r^*=1} &= K^* \left[ \frac{1}{2}(n^* - q^*) + 1 \right] \Big|_{r^*=1}. \end{aligned} \quad (1.2)$$

The electric charge density  $q$  is connected with its dimensionless value  $q^*$  by the equality  $q = e\gamma q^* n^0$ . The quantity  $L_0$  introduced above is obviously the same as the characteristic settling length for equilibrium ion concentrations in the tube (when  $x \gg L_0$  the ion concentration can be regarded as independent of  $x$ ). Since usually  $L_0^* \gg \min(1, De)$ , then in the operators  $\Delta^*$ ,  $\text{div}^*$  the derivatives with respect to  $x^*$  are small in comparison with the derivatives with respect to  $r^*$ , and they can be neglected. The electric current  $J$  flowing through the tube cross section and its dimensionless value  $J^*$  are calculated from the formulas

$$J = 2\pi \int_0^R q u_x r dr, \quad J^* = J/(e\gamma u^0 R^2 n^0) = 2\pi \int_0^1 q^* (1-r^{*2}) r^* dr^*. \quad (1.3)$$

2. To solve the problem (1.2) we use the Laplace transform. Converting from the functions  $q^*(x^*, r^*)$ ,  $n^*(x^*, r^*)$ ,  $E_r^1(x_1^*, r^*)$  to their images  $Q(p, r^*)$ ,  $N(p, r^*)$ ,  $Y(p, r^*)$  ( $p$  is a complex variable) we find that the latter satisfy the following system of equations and boundary conditions:

$$\begin{aligned} Q'' + Q'/r^* - [pPe(1-r^{*2}) + De^{-2}]Q &= 0, \\ N'' + N'/r^* + pPe(1-r^{*2})N &= 0, \quad (r^*Y)'/r^* = Q/2De^2; \end{aligned} \quad (2.1)$$

$$\left[ Y - \frac{1}{2}(N' + Q') \right] \Big|_{r^*=1} = 0, \quad \left[ Y + \frac{1}{2}(N' - Q') + \frac{K^*}{2}(N - Q) \right] \Big|_{r^*=1} = -\frac{K^*}{p}. \quad (2.2)$$

The substitution  $Z = \alpha r^{*2}$ ,  $\alpha = (-pPe)^{1/2}$ ,  $Q(r^*) = V(Z) \exp(-Z/2)$  brings the first equation of (2.1) to the form

$$ZV'' + (1-Z)V' - aV = 0, \quad a = 1/2 - (1/4)(\alpha - \alpha^{-1}De^{-2}). \quad (2.3)$$

The equation is a confluent hypergeometric equation [7]. The regular zero solution of this equation is a Kummer function [7]

$$\Phi(a, 1, Z) = 1 + \frac{a}{1} \frac{Z}{1!} + \frac{a(a+1)}{1 \cdot (1+1)} \frac{Z^2}{2!} + \dots$$

Thus, for  $Q$  we have

$$Q = A(p)G_a(p, r^*), \quad G_a \equiv \Phi(a, 1, \alpha r^{*2}) \exp(-\alpha r^{*2}/2),$$

and in a similar way we find the functions  $N(p, r^*)$ ,  $Y(p, r^*)$

$$\begin{aligned} N &= B(p)G_b(p, r^*), \quad G_b \equiv \Phi(b, 1, \alpha r^{*2}) \exp(-\alpha r^{*2}/2), \\ Y &= \frac{1}{2De^2} A(p)H(p, r^*), \quad H \equiv \frac{1}{r^*} \int_0^{r^*} G_a(p, r^*) r^* dr^*, \quad b \equiv \frac{1}{2} - \frac{\alpha}{4}. \end{aligned}$$

The coefficients  $A$  and  $B$  are found from the boundary conditions (2.2):

$$\begin{aligned} A &= 2p^{-1} [G_b'/F] \Big|_{r^*=1}, \quad B = -2p^{-1} [(G_a' - H/De^2)/F] \Big|_{r^*=1}, \\ F &= [(G_a G_b)' + 2G_a' G_b'/K^* - (H/De^2)(G_b + 2G_b'/K^*)] \Big|_{r^*=1}. \end{aligned}$$

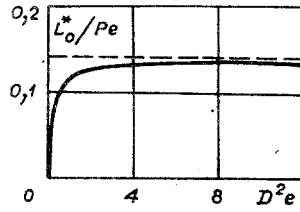


Fig. 1

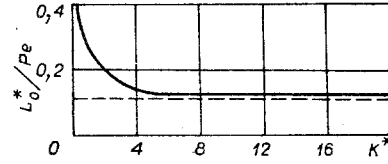


Fig. 2

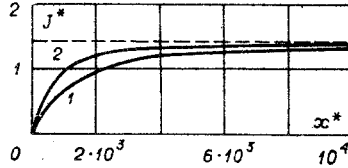


Fig. 3

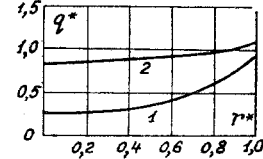


Fig. 4

The function  $Q(p)$  has poles at the point  $p = 0$  and points  $p_k \neq 0$  ( $k = 1, 2, 3, \dots$ ), which are zeros of the function  $F(p)$ ;  $0 > \text{Re}(p_1) > \text{Re}(p_2) > \dots$ . Using the expansion theorem [8] and calculating the residues of the function  $Q(p) \exp(px^*)$ , we obtain the following expression for the electric charge density  $q$

$$q = \gamma e z n^0 q^* = \gamma e z n^0 \left\{ \frac{2I_0(r^*/De)}{I_0(1/De) + 8De^2 I_2(1/De)} + \sum_k \frac{1}{(n_k - 1)!} \lim_{p \rightarrow p_k} \frac{d^{n_k-1}}{dp^{n_k-1}} \left[ (p - p_k)^{n_k} Q(p, r^*) \exp(px^*) \right] \right\}, \quad (2.4)$$

where  $I_0, I_2$  are modified Bessel functions of zero and second order;  $n_k$  is the order of the pole  $p_k$ .

There are similar expressions for the functions  $n^*(x^*, r^*)$ ,  $E_r^1(x^*, r^*)$ .

3. We introduce the function

$$\lambda(De, K^*) \equiv \left[ \text{Pe} \min_k \text{Re}(-p_k) \right]^{-1} \equiv \left[ \min_k \text{Re}(a_k^2) \right]^{-1}.$$

If  $x^* \gg \lambda \text{Pe}$  all the terms of the sum in relation (2.4) are small. Hence, when  $x^* \gg \lambda \text{Pe}$  the electric charge density in the tube depends weakly on  $x^*$  and electrification practically ceases. The characteristic electrification length introduced above can then be determined by the equality  $L_0 = \lambda \text{Pe} R$ .

Figure 1 shows a plot of the dimensionless electrification length (divided by the Péclet diffusion number) against the parameter  $De^2$ . It is apparent that an increase in Debye radius initially leads to a great increase in the characteristic electrification length. With further increase in Debye radius the characteristic electrification length changes by a small amount. A plot of  $L_0^*/\text{Pe}$  against the dimensionless rate of the chemical reaction on the wall is shown in Fig. 2 (in Figs. 2-4  $De = 1$ ). The rate of the reaction on the wall has a great effect on the characteristic electrification length of the liquid. Reduction of the rate of the electrochemical reaction on the wall leads to an increase in the characteristic electrification length of the liquid. Figure 3 shows a plot of the dimensionless electric current  $J^*$ , calculated from the formula (1.3), against the dimensionless channel length  $x^*$  (in the calculations we took  $\text{Pe} = 10^4$ ). An increase in tube length leads initially to a great increase in the electric current, and then it becomes insignificant (after the tube length becomes much greater than the electric charge relaxation length). For curves 1, 2 we have  $K^* = 1, \infty$ , respectively. When  $x^* \rightarrow \infty$  the electric current tends to a maximum value

$$J_{\max} = \frac{8\pi\gamma e z n^0 R^2 De^2 I_2(1/De)}{I_0(1/De) + 8De^2 I_2(1/De)}.$$

Plots of the dimensionless electric charge density  $q^*$  against the dimensionless tube radius  $r^*$  for different values of the dimensionless length  $x^*$  are shown in Fig. 4. For curves 1 and 2 we have  $x^* = 1000$  and  $\infty$  ( $K^* = \infty$ ), respectively. The electric charge density has a minimum value on the tube axis and a maximum value on the wall. Near the tube axis the variation of the electric charge density is slower than near the tube wall. With increase in distance from the tube entrance to the considered cross section the profile of the electric

charge density distribution changes and tends to the profile for an infinitely long tube, shown by curve 2 in Fig. 4. This curve is given by the first term on the right-hand side of expression (2.4) for  $q$ .

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#### ELECTRIFICATION OF A METAL BODY IN AN AEROSOL FLOW WITH A SOLID DISPERSE PHASE IN THE PRESENCE OF A CORONA DISCHARGE FROM THE BODY

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The electrification of a metal body in a flow of uncharged monodisperse aerosol with a solid disperse phase is investigated within the framework of continuum mechanics [1]. The corona discharge from the body is taken into account. We consider cases of well-conducting aerosol particles, for which the electric charge relaxation time is much greater than the time of impact with the body. A closed system of equations and boundary conditions describing the electrification of the body is obtained. We determine the main dimensionless parameters affecting the electrification of the body. We obtain expressions for the electrification current, the maximum corona current, the floating charge and potential of the body, the maximum corona overvoltage, and the characteristic time for establishment of the floating charge on the body. The main dimensionless characteristics of electrification of a sphere with a spark gap are calculated.

1. We consider a metal body with a spark gap in a steady flow of uncharged monodisperse aerosol with a solid disperse phase. As is known [2], the aerosol particles are charged by collisions with the body. The body consequently acquires an electric charge that is opposite in sign to the particle charge. This effect is observed when bodies move through clouds, precipitation, and a dust-laden atmosphere [3]. It can be used in electric probes designed for measuring the parameters of aerosol flows [4].

Using the methods of continuum mechanics [1, 5] we will consider the averaged motion of a monodisperse aerosol flow past a body as the interpenetrating motion of two continuous media - gas and aerosol particles. We assume that the concentration of the latter is fairly low and their effect on the gas motion can be neglected. Then, in the investigation of the electrification of bodies the motion of the gas can be regarded as prescribed. The averaged motion of the aerosol particles before collision with the body is described by the following equations:

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