# ELECTROHYDRODYNAMIC INSTABILITY OF A WEAKLY CONDUCTIVE LIQUID LOCATED BETWEEN SPHERICAL ELECTRODES IN THE PRESENCE OF WEAK INJECTION

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High-voltage conductivity of liquid dielectrics may be produced by charge injection from electrodes [1-6]. The charges flowing from the electrodes have the same sign as the electrode polarity. The electrode region becomes charged, causing Coulomb forces to appear, directed in the direction away from the electrode. There thus develops an unstable equilibrium state, and at sufficiently high electric field intensities the liquid is set in motion [3-6]. The present study will obtain criteria for the stability of equilibrium of a weakly conductive liquid located between two spherical electrodes for cases of unipolar injection from the outer or inner electrodes.

1. Formulation of the Problem. Let the electrode radii be $R_1$, $R_2$ ($R_1 < R_2$) with an applied potential difference $U = \text{const}$. The space charge $q = n_2 - n_1$ is formed in the liquid due to injection of charges from the electrodes, so that conductivity may be expressed in the form $\sigma = u_1 n_1 + u_2 n_2$, where $n_1$, $n_2$ are the values of the space-charge densities injected by the inner and outer electrodes, respectively, and $u_1$, $u_2$ are the charge mobilities. We thus assume that the injection current is significantly higher than the current produced by impurities and dissociation [1-3].

The motion of the weakly conductive polarizable incompressible liquid is described by the electrohydrodynamics equations [6, 7]

$$\begin{gathered}\rho(\partial \mathbf{v}/\partial\tau + (\mathbf{v}\nabla)\mathbf{v}) = -\nabla p + \eta\Delta\mathbf{v} + q\mathbf{E},\ \operatorname{div}\mathbf{v} = 0,\\ \operatorname{div}\varepsilon\mathbf{E} = 4\pi q,\ \operatorname{rot}\mathbf{E} = 0,\ \partial n_i/\partial\tau + \operatorname{div}\mathbf{j}_i = 0,\\ \mathbf{j}_i = (-1)^i n_i u_i \mathbf{E} + n_i\mathbf{v}\ (i = 1, 2),\end{gathered} \tag{1.1}$$

where $\mathbf{j}_1$, $\mathbf{j}_2$ are the respective densities of the currents produced by the injected charges; $\eta$, dynamic viscosity; and p, total pressure [7].

The boundary conditions for Eq. (1.1) have the form

$$\mathbf{v}|_{r=R_1,R_2} = 0,\ \varphi = 0,\ |\mathbf{j}_1| = j_1 \ \text{at}\ r = R_1;\ \varphi = U,\ |\mathbf{j}_2| = j_2\ \text{at}\ r = R_2. \tag{1.2}$$

Equality to zero of the velocity is a consequence of the adhesion condition; the remaining conditions follow from specification of the potential and injected current densities on the electrodes.

2. Equilibrium State. The boundary-value problem describing the equilibrium state consists of system (1.1) at $\mathbf{v} \equiv 0$, $\partial/\partial\tau \equiv 0$ and boundary conditions (1.2). We introduce a spherical coordinate system ($r$, $\theta$, $\varphi_1$), with origin at the center of symmetry. The solution describing the equilibrium state will be sought in the form

$$\mathbf{E}_0 = (E_0(r), 0, 0),\ n_{i0} = n_{i0}(r). \tag{2.1}$$

Here and below we consider index i as taking on the values of 1 and 2.

Placing (2.1) in the equilibrium equation and using (1.2), we obtain a solution in the form

$$E_0 = -\frac{aR_1^2}{r^2}\sqrt{1 + \frac{8\pi D r^3}{3\varepsilon a^2 R_1^4}},\quad E_0 = -\frac{d\varphi_0}{dr},\quad n_{i0} = -\frac{j_i R_i^2}{u_i r^2 E_0},\quad D \equiv \frac{j_1 R_1^2 u_2 - j_2 R_2^2 u_1}{u_1 u_2}, \tag{2.2}$$

where the constant $a$ is determined from boundary conditions (1.2) for the potential $\varphi_0$. We note that the constant $a$ has the dimensions of electric field intensity, and in the absence of injection ($j_i = 0$) then $a \equiv E_1 = UR_2/[R_1(R_2 - R_1)]$. Thus, if we assume that the injection is weak

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$$\mu_i \equiv \frac{4\pi D_i}{3\varepsilon E_1^2 R_1} \ll 1,\ D_i \equiv \frac{j_i R_i^2}{u_i}, \tag{2.3}$$

then Eq. (2.2) may be simplified by expanding in a series in the small parameter $\mu = \mu_1 - \mu_2$ and retaining terms linear in $\mu$

$$E_0 = -\frac{E_1 R_1^2}{r^2}\left(1 + \mu \frac{r^3}{R_1^3}\right),\ n_{i0} = \frac{D_i}{|E_1| R_i^2}\left(1 - \mu \frac{r^3}{R_1^3}\right). \tag{2.4}$$

3. Study of Stability. We will consider the stability of the equilibrium state described by Eq. (2.4) with respect to infinitely small perturbations [8]. Representing the perturbed state by a velocity $e^{\lambda\tau}\mathbf{v}(\mathbf{r})$, pressure $p_0 + e^{\lambda\tau}p(\mathbf{r})$, electric field intensity $\mathbf{E}_0 + e^{\lambda\tau}\mathbf{e}(\mathbf{r})$, and space charges $n_{i0} + e^{\lambda\tau}q_i(\mathbf{r})$, with consideration of Eq. (2.3) for the perturbation, we obtain the following system of linearized equations:

$$\begin{gathered}\lambda\rho\mathbf{v} = -\nabla p + \eta\Delta\mathbf{v} - (q_2 - q_1) E_1 R_1^2/r^2 \mathbf{e}_r^0,\ \operatorname{div}\mathbf{v} = 0,\\ \lambda q_i - (-1)^i u_i E_1 R_1^2/r^2 \cdot dq_i/dr + dn_{i0}/dr \cdot v_r = 0.\end{gathered} \tag{3.1}$$

Using Eq. (2.3), it can be shown [4] that the boundary conditions for system (3.1) have the form

$$\mathbf{v}|_{r=R_i} = 0,\ q_i|_{r=R_i} = 0. \tag{3.2}$$

We will study the stability with respect to monotonic-type perturbation ($\lambda$ a real number). Then on the stability boundary ($\lambda = 0$) Eq. (3.1) and boundary conditions (3.2) define a boundary problem for eigenvalues of $E_1$. If we seek the eigenfunctions in the form

$$\begin{gathered}v_r = v_l(r)\, T_{0n}^l(\pi/2 - \varphi_1, \theta, 0),\\ v\pm = \mp\frac{1}{\sqrt{2}}(v_{\varphi_1} \pm i v_\theta) = v^\pm(r)\, T_{\pm 1,n}^l(\pi/2 - \varphi_1, \theta, 0),\\ p = p_l(r)\, T_{0n}^l(\pi/2 - \varphi_1, \theta, 0),\ q_i = q_{il}(r)\, T_{0n}^l(\pi/2 - \varphi_1, \theta, 0),\end{gathered}$$

where $T_{nm}^l(\varphi_1, \theta, \varphi_2)\,(-l \leqslant n, m \leqslant l;\ l = 1, 2, 3, \ldots)$ are generalized spherical functions, then in analogy with [9] it can be shown that the critical intensity $E_{1*}$, at which the liquid loses stability, is defined by the following eigenvalue problem for $E_1$

$$\eta D_l^2 v_1 = -l(l+1) E_1 R_1^2/r^3 \cdot (q_{2l} - q_{1l}), \tag{3.3}$$

$$dq_{il}/dr = (-1)^i r/(E_1 R_1^2 u_i) \cdot dn_{i0}/dr \cdot v_1; \tag{3.4}$$

$$v_1 = dv_1/dr = 0,\ q_{il} = 0 \text{ at } r = R_i,$$

where

$$v_1 = r v_l;\ D_l \equiv \frac{d^2}{dr^2} + \frac{2}{r}\frac{d}{dr} - \frac{l(l+1)}{r^2}.$$

We transform to the dimensionless variables $t = r/R_1$, $v = v_1\rho/\eta$, $\gamma_2^0 = q_{2l}\frac{\rho R_1^3 u_2^2 E_1^2}{3\mu\eta j_2 R_2^2}$, $\gamma_1^0 = q_{1l}\frac{\rho R_1 u_1^2 E_1^2}{3\mu\eta j_1}$ and perform a substitution $t = 1/s$, $w(s) \equiv v(1/s)$, $\gamma_i = \gamma_i^0(1/s)$. Then the problem of Eqs. (3.3), (3.4) will be equivalent to the following boundary-value problem for eigenvalues of $K_1$, if the value $K_2$ is specified, and for $K_2$, if $K_1$ values are specified:

$$sL_l s^4 L_l w = 4\pi l(l+1)(K_1\gamma_1 - K_2\gamma_2),\ d\gamma_i/ds = (-1)^i w s^3; \tag{3.5}$$

$$w = dw/ds = 0,\ \gamma_i = 0 \text{ at } s = h_i, \tag{3.6}$$

where $L_l \equiv \frac{d^2}{ds^2} - \frac{l(l+1)}{s^2};\ h_1 \equiv h = \frac{R_1}{R_2};\ h_2 = 1;\ K_1 = \frac{j_1 R_1 (j_1 R_1^2 u_2 - j_2 R_2^2 u_1)}{\varepsilon |E_1|^3 u_1^3 u_2 \eta};\ K_2 = \frac{j_2 R_2^2 (j_1 R_1^2 u_2 - j_2 R_2^2 u_1)}{\varepsilon |E_1|^3 R_1 u_1 u_2^3 \eta}$. We will consider two cases of unipolar injection: from the inner electrode ($j_2 = 0$); from the outer electrode ($j_1 = 0$). In these cases Eqs. (3.5), (3.6) may be reduced to a problem of determining characteristic values $\lambda_i$ of integral equations with positive integrands

TABLE 1

| $l$ \ $h$ | 0,05 | 0,1 | 0,2 | 0,3 | 0,4 | 0,5 |
|---|---|---|---|---|---|---|
| 2 | $478\cdot10^{-7}$ | $150\cdot10^{-5}$ | $582\cdot10^{-4}$ | 0,639 | | |
| 3 | $501\cdot10^{-7}$ | $148\cdot10^{-5}$ | $505\cdot10^{-4}$ | 0,470 | 2,990 | |
| 4 | $571\cdot10^{-7}$ | $163\cdot10^{-5}$ | $534\cdot10^{-4}$ | 0,445 | 2,465 | 12,76 |
| 5 | | $185\cdot10^{-5}$ | $600\cdot10^{-4}$ | 0,475 | 2,363 | 10,81 |
| 6 | | | | | 2,471 | 10,14 |
| 7 | | | | | 2,703 | 10,18 |

TABLE 2

| $l$ \ $h$ | 0,05 | 0,1 | 0,2 | 0,3 | 0,4 | 0,5 |
|---|---|---|---|---|---|---|
| 2 | $144\cdot10^{-7}$ | $415\cdot10^{-6}$ | $194\cdot10^{-4}$ | 0,264 | | |
| 3 | $200\cdot10^{-7}$ | $505\cdot10^{-6}$ | $181\cdot10^{-4}$ | 0,198 | 1,497 | |
| 4 | $281\cdot10^{-7}$ | $663\cdot10^{-6}$ | $210\cdot10^{-4}$ | 0,195 | 1,250 | |
| 5 | $382\cdot10^{-7}$ | $867\cdot10^{-6}$ | $261\cdot10^{-4}$ | 0,218 | 1,220 | 6,37 |
| 6 | | | | | 1,305 | 6,03 |
| 7 | | | | | 1,468 | 6,12 |

$$w(s) = \lambda_i \int_h^1 G_i(s, t) \frac{w(t)}{t^5} dt, \tag{3.7}$$

where

$$\lambda_i = 4\pi l(l+1) B_i;\ B_1 = \frac{j_1^2 R_1^3}{\varepsilon |E_1|^3 u_1^3 \eta};\ B_2 = \frac{j_2^2 R_2^4}{\varepsilon |E_1|^3 u_2^3 \eta};$$

$$G_1(s, t) = \int_h^t G(s, \xi)\, d\xi; \qquad G_2(s, t) = \int_t^1 G(s, \xi)\, d\xi;$$

$G(s, \xi)$ is Green's operator function, defined by the differential expression $sL_l s^4 L_l$ and boundary conditions $w = dw/ds = 0$ at $s = h_i$.

Calculation of the smallest characteristic values $\lambda_{i_1}$ was performed by the iteration method [9] with a relative error < 0.1%. Tables 1 and 2 present the results of calculating the functions $B_1 = B_1(h, l)$, $B_2 = B_2(h, l)$ for various values of h and $l$. The critical values $B_{i*}$ are determined from the condition $B_{i*} = \min_{l \geq 1} B_i(h, l)$. For $B_i < B_{i*}$ the liquid is in equilibrium, while for $B_i > B_{i*}$ it goes into motion. The calculations also showed that in both cases for h < 0, 1 the critical motions correspond to $l = 2$. With increase in h the corresponding $l$ values increase by one.

We will present some numerical estimates. In polar liquids in the presence of semipermeable membranes, injection may occur even at low voltages [3-5]. The densities of the injected currents can then reach significant values (up to 100 $\mu A/cm^2$ in nitrobenzol [3]). We will estimate the magnitude of the critical current density for the inner electrode, assuming that the electric field intensity $E_1$ varies over the limits $E_1$ = 50-100 kV/cm, with ion mobility of the order of magnitude of $10^{-4}$ $cm^2/V \cdot sec$ [2, 3], viscosity $\eta = 0.2$ P, $\varepsilon = 2$ for $h = 0.1$, $R_2 = 1$ cm. On the stability boundary $K_1 = K_{1*} = 0.0015$, whence after substitution in the expression for $K_1$ of the indicated values we obtain $j = 6.45$-$8.16$ nA/cm, with $\mu_1 = 0.1$-$0.05$. To determine the exact value of the critical electric field intensity near the inner electrode (and also the density of the injected current at the electrode) it is necessary to know the dependence $j = j(E)$ [1-3]. For example, for steel electrodes and well purified n-hexane the cold emission current density dependence on field intensity at the cathode has the form [1]

$$j = aE^2 \exp(-b/E),$$

where $a = 5.1 \cdot 10^{21}$ A/V$^2$, b = $2.66 \cdot 10^5$ V/cm. If we assume [2, 10] that $u_1 = 10^{-4}$ cm$^2$/V·sec, $\eta$ = 0.0029 P, $\varepsilon$ = 2, h = 0.1, $R_2$ = 1 cm, then $E_{1*}$ = 38 MV/cm, with $\mu_1$ = 0.0036.

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