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# INJECTION CURRENT AND THE FORMATION OF BUBBLES IN STRONG, VERY NONUNIFORM ELECTRIC FIELDS

**S. M. Korobeinikov**

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*The flow problem of a nonstationary current upon injection from a spherical electrode is solved. It is shown that the field distribution can have a maximum that is displaced in the direction from the electrode with the velocity of the injected-charge front. The formation of bubbles in pre-breakdown electric fields is analyzed and it is shown that the bubbles can form in dielectric liquids without ionization under the action of high voltage.*

**Introduction.** An analysis of nonstationary injection currents is important not only for studying the structure of dielectric and semiconducting materials [1], but also for estimating the near-electrode electric fields, energy release, and pressure in the near-electrode region under the action of strong electric fields on the dielectrics.

It is difficult to find a general solution of the problem, because the equations are nonlinear; therefore, the problem was solved earlier for partial cases of a nonstationary current limited by a volume charge injected from a flat [1] or spherical electrode [2] connected to a constant-voltage source. It was assumed that at the initial moment of time, the volume charge is photoinjection-induced at the electrode; otherwise the electrode is regarded as a reservoir electrode. These assumptions are not true in the case of a nonstationary current in the liquid under the action of a strong electric field. Many experiments (see, e.g., [3, 4]) showed that the dependence of the current on the voltage $I(U)$ in nonpolar liquids of the type of hexane, tetramethylsilane, and cyclohexane for electrodes with a strongly nonuniform field has three characteristic sites; a linear dependence occurs at low voltage; when a certain voltage is reached, the current increases by two or three orders of magnitude; with the voltage further increased, the dependence $I(U)$ takes the form $I \sim U^2$. The first site corresponds to Ohm's law, the second site to the field emission of the charge carriers from the electrode, and the third site to the space-charge-limited current (SCLC). In nonpolar liquids, the transition to the SCLC regime occurs at the comparatively low local strength $E_l \leqslant 10^7$ V/cm. We note that the specific feature of the SCLC regime is the weak dependence of the near-electrode strength on voltage.

Korobeinikov et al. [5] studied the electric-field distributions in the near-electrode region in experiments with electro-optical registration of the pre-breakdown electric fields in nitrobenzene [5]. It was shown that after exceeding a certain threshold voltage, the spatial distribution of the electric field at a needle-shaped cathode has a maximum that is displaced from the electrode. The field strength remains constant irrespective of the voltage near the electrode.

**Model.** In modeling the above-described behavior of the current and the pre-breakdown field, we consider that as the voltage increases, the near-electrode strength first grows proportionally to the voltage (ohmic regime), then the SCLC regime occurs, and the near-electrode strength remains at the level of strength that corresponds to the transition to this regime. The transient regime is not considered.

To determine the injected charge, it is necessary to solve a system of equations of electrodynamics with appropriate initial and boundary conditions.

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The method of solving the equations that was developed by us allows one to solve the problem for an arbitrary law of emission and for electrode systems having symmetry, i.e., for planes, cylinders, and spheres [6]. We restrict ourselves to the case of a spherical electrode with the simple law of emission, which is the most important for practice. In calculating the volume charges, the pressures, and the energy release, the contribution from the first two sites of the volt–ampere characteristic can be ignored; therefore, we assume that the emission begins when a certain threshold strength $E_i$ is reached and that the electrode becomes a reservoir electrode when this strength is exceeded. For the solution, information on the mobility of charge carriers $\mu$ is needed; without taking into account its dependence on the time and the field strength, we consider this mobility hydrodynamic [7].

We write the initial system of equations, which includes the continuity, Poisson, and total-current and total-voltage equations, in dimensionless form:

$$\frac{1}{r'^2}\frac{\partial}{\partial r'}(r'^2E'\rho') + \frac{\partial \rho'}{\partial t'} = 0, \qquad \frac{1}{r'^2}\frac{\partial}{\partial r'}(r'^2E') = \rho',$$
$$\frac{\partial E'}{\partial t'} + \rho' E' = J', \qquad \int_1^\infty E'(r',t')\,dr' = U'(t'). \tag{1}$$

The transition from dimensionless to dimensional quantities is performed according to the following formulas:

$$J = \frac{\varepsilon_0 \varepsilon U^2 \mu}{r_0^3} J', \quad t = \frac{r_0^2}{\mu U} t', \quad \rho = \frac{\varepsilon_0 \varepsilon U}{r_0^2}\rho', \quad r = r_0 r'.$$

Here $J$ is the current density, $U$ is the amplitude of the voltage pulse, $r_0$ is the electrode radius, $\varepsilon$ is the dielectric permeability of the liquid, and $\varepsilon_0$ is the dielectric constant. The hyperbolic system (1) is solved by the method of characteristics [8]. To do this, we substitute the second equation into the first one:

$$\frac{\partial \rho'}{\partial t'} + \frac{\partial \rho'}{\partial r'}E' + \rho'^2 = 0.$$

The curves $dr'(t',t'_0)/dt' = E'$, where $t'_0$ is the moment at which the characteristic leaves the electrode (injection of each portion of the charge), are the characteristics of this equation. We can obtain

$$\frac{d\rho'}{dt'}(t',t'_0) = -\rho'^2, \qquad \frac{d(E'r'^2)}{dt'}(t',t'_0) = J'r'^2$$

from the conditions on the curve or

$$r'(t',t'_0) = 1 + 3(l(t') - l(t'_0) - Q(t'_0)(t' - t'_0))^{1/3},$$
$$\rho'(r'(t',t'_0)) = \rho'_0/(1 + (t' - t'_0)\rho'_0), \quad E'(r'(t',t'_0)) = (E'_i + Q(t') - Q(t'_0))/r'^2 \tag{2}$$

after the integration. Here $\rho'_0$ is the density of the volume charge in the near-electrode region at the moment $t'_0$, $Q(t') = \int J'(t')\,dt'$, and $l(t') = \int Q(t')\,dt'$. Substituting (2) into the total-voltage equation, after transformations we obtain

$$Q(t') = U'(t') - E'_i + \int_0^{t'} \frac{Q(t')[E'_i + (t' - t'_0)\,dQ/dt'_0]}{\{1 + 3[l(t') - l(t'_0) - Q(t'_0)(t' - t'_0)]\}^{4/3}}\,dt'_0. \tag{3}$$

For quite short voltage pulses, Eq. (3) can be solved by the method of successive approximations. We use $Q'_0 = U(t') - E'_i$ as a zero approximation. The physical meaning of the zero approximation is clear — this is a charge of geometrical capacity. The physical meaning of $Q_1$ and subsequent approximations is to increase the capacity by means of the volume emission charge. Choosing the criterion of convergence of the iterative process $(Q_n - Q_{n-1})/Q_n < \delta$ ($\delta$ is the specified small number), we find the solution $Q(t')$.

Figure 1 shows the field-strength distribution in the vicinity of the injecting spherical electrode at various moments of time. One can see that, with time, the minimum of the field strength in the volume-charge zone does not remain at the electrode surfaces in the case of flat electrodes [1]. For large moments

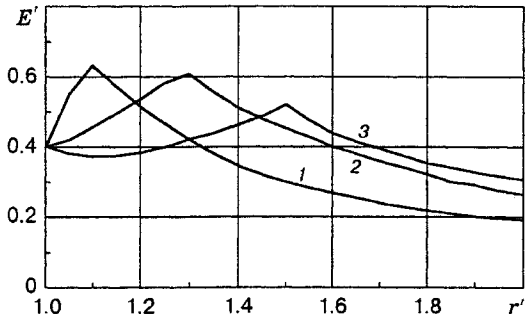


Fig. 1. Distribution of the electric-field strength upon injection of the charge carriers for $t' = 0.3$ (1), 0.6 (2), and 1 (3).

of time, the volume charge is distributed over the gap so that the maximum of strength is restored at the electrode.

One can estimate the heating of the liquid $\Delta T$ and the pressure drop $\Delta P$ in the near-electrode region under pre-breakdown conditions from the dimensionless quantity $Q(t')$ [5]:

$$\Delta T = \frac{\varepsilon_0 \varepsilon E_0^2}{c_p d} Q(t') E_i', \qquad \Delta P = \varepsilon_0 \varepsilon E_0^2 \frac{d}{dt}(Q - U'). \tag{4}$$

Here $c_p$ and $d$ are the thermal capacity and density of the liquid and $E_0$ is the field strength in the absence of injection.

**Analysis of Experimental Data.** In analyzing the experimental data, one should take into consideration that it is not known with certainty if the bubbles are the consequence of ionization processes in the liquid, the transformation of a microbubble (micronucleus) to a "macrobubbles," or ionization-free pre-breakdown processes.

The estimates (4) show that, for experimental conditions under which the pre-breakdown processes in nitrobenzene are studied for $E_0 \approx 1$ MV/cm and $E_i' \approx 0.5$ [5], the variation in temperature is negligible ($\Delta T < 1$ K), and a negative pressure equal to about 1 MPa is the determining factor for the formation of bubbles.

In the experiments on measurement of the dependence of the current on the voltage in using the point–plane electrode system with a point radius of 0.5–10, it was established that:

— short current pulses (shorter than 4 nsec) are generated at this strength;

— the charge injected per pulse is determined only by the radius of curvature and does not depend neither on the voltage applied nor the pressure;

— the period of time between the pulses decreases with increase in voltage;

— each pulse produces a bubble at the point and the size and time of life of this bubble depend on the external pressure.

These facts suggest that in nonpolar liquids, the strength at the onset of shock ionization amounts to several megavolts per centimeter, for example, 7 MV/cm for pentane and decane [9].

We consider the formation of bubbles in the near-electrode region owing to injection processes. It is impossible to use directly the calculation results obtained for the nonstationary injection current, because the flow conditions for the current in [9] are almost stationary. As the field strength shows, the flow regime for the current is close to the SCLC regime.

In passing to the SCLC regime, the charge density $\rho$ is estimated to be $\rho \sim \varepsilon_0 \varepsilon E_l / r_l$, where $E_l$ is the field strength near the micropoint of radius $r_l$, at which the current passes to the SCLC regime. Upon emission of the charge carriers in the SCLC regime, the energy release is

$$W \approx \rho \mu E_l^2 t \sim \varepsilon_0 \varepsilon \mu E_l^3 t / r_l. \tag{5}$$

Here, because of the action of Coulomb forces on the volume charge, the pressure near the micropoint is smaller than atmospheric pressure $P_a$, i.e.,

$$P = P_a - \frac{Jr_l}{\mu} \sim P_a - \varepsilon_0 \varepsilon E_l^2. \tag{6}$$

It follows from the estimates (5) and (6) that overheating and cavitation should occur near the micropoint in the microregion for quite a high $E_l$. This means that at a high local strength, microbubbles can also form during the action of a voltage pulse for almost any duration of this pulse.

We analyze the results of [9] as applied to the bubble model [10] with allowance for the emission of charge carriers. We consider that at a field strength of about 7 MV/cm, the electron emission varies from the injection-limited current (ILC) to the SCLC regime. For a point of radius 1 $\mu$m, the volume-charge density is approximately 0.01 C/cm$^3$. If the mobility of charge carriers is $10^{-3}$–$10^{-4}$ cm$^2$/(V $\cdot$ sec), the current density amounts to 7–70 A/cm$^2$, the negative pressure is 7 MPa in the stationary regime, and the energy release reaches 1 kJ/cm$^3$ for 20–200 $\mu$sec. The heat diffusion from the micropoint region decreases the temperature in the near-electrode region but does not affect the level of negative pressure. Therefore, a metatstable region should appear near the micropoint mainly owing to the negative pressure; this leads to the generation of a bubble as a result of homogeneous nucleation. Overheating of the liquid becomes the determining factor in the case of high-mobility charge carriers, and tension in the case of low-mobility charge carriers.

The rate of bubble growth is determined by the degree of overheating. The velocity of sound and the characteristic velocity of Rayleigh pulsations are the upper and lower estimates of the rate, respectively. The time of bubble growth up to a radius of 1 $\mu$m is estimated to be 1–10 nsec. The volume charge in the near-electrode zone is ejected by the growing bubble; this is manifested in the external electric circuit by the current pulse whose duration is equal to the time of bubble growth. The bubble size is determined by the radius of rounding of the point; therefore, the charge ejected by the bubble from the strong-field zone is determined by the radius of the point as well.

Upon homogeneous nucleation, the frequency $S$ of bubble formation per unit volume is determined by the depth of penetration into the metastable region [11] and can be written in the form

$$S = \left[N \exp\left(-\frac{\lambda}{kT}\right)\left(\frac{2\sigma}{\pi m}\right)^{1/2}\right] \exp\left(\frac{-16\pi\sigma^3}{3kT(P_s(T) - P_a + \varepsilon_0 \varepsilon E_l^2)^2}\right). \tag{7}$$

Here $N$ is the molecular concentration, $m$ is the molecular mass, $\lambda$ is the heat of vaporization, $k$ is the Boltzmann constant, $T$ is the temperature, $\sigma$ is the surface-tension coefficient, and $P_s(T)$ is the pressure of the saturated vapors. An analysis of the dependence (7) shows that the expression in square brackets is a large quantity ($10^{30}$ cm$^3$/sec) that depends weakly on the depth of penetration into the metastable region. The decisive role is played by the second cofactor equal to zero on the line of vapor–liquid equilibrium; this cofactor increases rapidly with distance from the line of equilibrium, reaching unity at the critical point. In turn, the possibility of deep penetration into the metastable region depends strongly on the local strength. This follows from the following considerations. If the negative pressure and overheating occur before the transition of the ILC regime to the SCLC regime, the energy release will depend on the voltage exponentially. In the SCLC regime, the energy release has a cubic dependence on the voltage. The dependence of the frequency of bubble formation on the voltage should have a threshold character in the ILC and SCLC regimes. Therefore, the formation of bubbles should also have a threshold character, depending on the voltage. Here the voltage threshold does not almost depend on the pressure, because the energy release and the negative pressure are determined by emission processes, and even weak changes in the local strength compensate for any real changes in the external pressure. The size of the formed bubble depends on the external pressure. The release of overheating into the bubble gives rise to cooling of the near-electrode region. The bubble is rejected from it owing to the electrohydrodynamic currents and the dielectrophoretic force, is cooled, and collapses according to the known model of Rayleigh pulsations. After that, the process is repeated: the energy release, the negative pressure, the formation of a bubble, etc. The period depends on voltage: the higher the voltage,

the shorter the period of pulse repetition. Here, in practice, the charge stored in the near-electrode region in the SCLC regime for the period does not depend on the voltage applied.

This mechanism also explains the occurrence of bubbles near the micropoint at the surface of flat electrodes in strong electric fields [11]. As a rule, the field increases by a factor of 30–200 near the micropoint [12]. This means that an emission intensity of 10 MV/cm will be reached already at an average strength of 100 kV/cm, i.e., under the usual conditions of breakdown experiments in liquids. In turn, this implies that in almost all the "breakdown" experiments, the ionization-free formation of microbubbles could occur immediately during the action of the voltage pulse.

**Conclusion.** Thus, an analysis of injection currents and the related energy release and negative pressure allows one to explain known experiments on detection of pre-breakdown microbubbles without using the hypothesis of shock ionization in the liquid phase at the pre-breakdown stage.

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