ISSN 1063-7850, Technical Physics Letters, 2006, Vol. 32, No. 7, pp. 634–637. © Pleiades Publishing, Inc., 2006.
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Anisotropic Instability of Dielectric Liquids and Decay to Vapor–Liquid System in Strong Electric Fields

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Received January 19, 2006

Abstract—The results of a linear analysis for the stability of a homogeneous dielectric liquid with respect to density perturbations in a uniform electric field are presented. The electric field increases the instability increment for the stratification along the field and decreases this increment in the transverse direction. Thus, a strong electric field can induce anisotropic decay into liquid and vapor phases for a fluid that is initially both in a labile state and in a metastable or stable state. Theoretical calculations are confirmed by the results of numerical modeling of the fluid dynamics. The new regions of a low-density phase have the form of narrow cylindrical channels oriented along the field. This new mechanism of gas phase formation in strong local electric fields probably plays a key role in the inception and ultra-fast propagation of streamers during the breakdown in dielectric liquids in a nanosecond range.

PACS numbers: 47.65.+a, 64.70.Fx, 47.17.–k, 47.20.Hw, 64.10.+h

DOI: 10.1134/S106378500607025X

The volume force acting upon a charged dielectric liquid in an electric field is determined by the well-known Helmholtz formula [1]

$$\mathbf{F} = q\mathbf{E} - \frac{E^2}{8\pi}\nabla\varepsilon + \frac{1}{8\pi}\nabla\left[E^2\rho\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T\right], \quad (1)$$

where $\mathbf{E}$ is the electric field, ε is the permittivity, and ρ is the density. The second and third terms in Eq. (1) describe the effect of electric field on polarization-induced charges in the inhomogeneous medium and the action of electrostrictive forces, respectively.

For gases and liquids with weakly polarizable molecules, the permittivity is a linear function of the density:

$$\varepsilon = 1 + 3\alpha\rho, \quad (2)$$

where $\alpha = 4\pi\beta/(3m)$, β is the polarizability, and m is the molecular mass. Nonpolar liquids obey the Clausius–Mosotti law [2, 3]

$$\varepsilon = 1 + 3\alpha\rho/(1 - \alpha\rho), \quad (3)$$

whereas polar liquids obey the Onsager–Kirkwood–Fröhlich relation [2]

$$B\varepsilon\rho = (\varepsilon - \varepsilon_\infty)(2\varepsilon + \varepsilon_\infty), \quad (4)$$

where $B = g\mu_v^2 4\pi(\varepsilon_\infty + 2)^2/(9mkT)$, $\varepsilon_\infty = n^2$, n is the high-frequency refractive index, μ_v is the dipole moment of a molecule in the gas phase, and g is the average correlation factor characterizing the short-range orientation order.

The stability of a homogeneous dielectric liquid with respect to liquid–vapor stratification in an electric field is determined by the equation of state (more precisely, by the $\partial p/\partial\rho$ value), the dielectric properties (ε, $(\partial\varepsilon/\partial\rho)_T$, $(\partial^2\varepsilon/\partial\rho^2)_T$), and the field strength E. We have performed a standard linear stability analysis of the Euler equations [4]

$$\frac{\partial\rho}{\partial t} + \mathrm{div}(\rho u) = 0, \quad \frac{\partial(\rho\mathbf{u})}{\partial t} + \nabla(p + \rho u^2) = F, \quad (5)$$

for a dielectric liquid in an electric field in the simplest isothermal case. Obviously, isothermal changes in the state of a fluid must be accompanied by the corresponding supply and removal of heat.

Let a dc voltage be applied between two plane horizontal electrodes, so that only the vertical projection of the electric field vector E_z is nonzero. We consider the growth of small one-dimensional perturbations in the density and velocity corresponding to stratification along the field,

$$\rho = \rho_0 + A_0\exp(\gamma t)\exp(i2\pi x/\lambda),$$
$$u = C_0\exp(\gamma t)\exp(i2\pi x/\lambda) \quad (6)$$

and in the transverse direction,

$$\begin{aligned}\rho &= \rho_0 + A_0\exp(\gamma t)\exp(i2\pi z/\lambda),\\ u &= C_0\exp(\gamma t)\exp(i2\pi z/\lambda),\end{aligned} \tag{7}$$

where λ is the wavelength, γ is the instability increment, ρ_0 is the average density, and A_0, C_0 are the initial amplitudes of perturbations in density and velocity, respectively.

The volume force acting upon the liquid with perturbations described by Eqs. (6) can be written as

$$F_x = \frac{E_0^2\rho}{8\pi}\left(\frac{\partial^2\varepsilon}{\partial\rho^2}\right)_T\frac{\partial\rho}{\partial x} = K_x\frac{\partial\rho}{\partial x}, \tag{8}$$

where E_0 is the uniform electric field strength. For the perturbations described by Eqs. (7) we have

$$F_z = \frac{D_0^2\rho}{8\pi\varepsilon^2}\left(\left(\frac{\partial^2\varepsilon}{\partial\rho^2}\right)_T - \frac{2}{\varepsilon}\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T^2\right)\frac{\partial\rho}{\partial z} = K_z\frac{\partial\rho}{\partial z}, \tag{9}$$

where D_0 is the electric displacement, which is constant in space for the perturbations under consideration. The instability increment in both cases is given by the formula

$$\gamma = \frac{2\pi}{\lambda}\sqrt{-\frac{\partial p}{\partial\rho} + K}. \tag{10}$$

A similar expression for the instability increment can be obtained for the Euler equations describing the growth of two-dimensional (2D) perturbations of the following type (cylindrical channels parallel to the z axis):

$$\begin{aligned}\rho &= \rho_0 + A_0\exp(\gamma t)\exp(i2\pi x/\lambda)\exp(i2\pi y/\lambda),\\ u_x &= C_0\exp(\gamma t)\exp(i2\pi x/\lambda)\exp(i2\pi y/\lambda),\\ u_y &= Q_0\exp(\gamma t)\exp(i2\pi x/\lambda)\exp(i2\pi y/\lambda),\end{aligned} \tag{11}$$

provided that K_y is described by a relation analogous to Eq. (8). In the 2D case, the increment is greater by a factor of $\sqrt{2}$ than the value for the perturbations described by Eq. (6).

The derivative $\partial p/\partial\rho$ is determined by the equation of state. In the isothermal case, it is necessary to use the quantity $(\partial p/\partial\rho)_T$, which is positive for stable and metastable states. In the forbidden region where $(\partial p/\partial\rho)_T < 0$, the fluid cannot exist and the so-called spinodal decomposition takes place [5].

In a similar way, the growth of perturbations (6) was considered for one-dimensional Navier–Stokes equations [4],

$$\begin{aligned}\frac{\partial\rho}{\partial t} + \frac{\partial(\rho u)}{\partial x} &= 0,\\ \frac{\partial(\rho u)}{\partial t} + \frac{\partial(p+\rho u^2)}{\partial x} &= F_x + (4/3\mu + \xi)\frac{\partial^2 u}{\partial x^2},\end{aligned} \tag{12}$$

where μ is the dynamic viscosity and ξ is the second viscosity coefficient.

The instability increment (10) exhibits infinite growth with decreasing perturbation wavelength. For viscous fluids, the instability increment for small perturbations can be written as

$$\gamma = \frac{2\pi}{\lambda}\left(\sqrt{(\pi b/\lambda)^2 + K - \partial p/\partial\rho} - \pi b/\lambda\right) \tag{13}$$

and tends to a constant value $\gamma_{\max} \approx (K - \partial p/\partial\rho)/b$ for the wavelength shorter than $\lambda_* \sim 2\pi b/\sqrt{K - \partial p/\partial\rho}$ (instability increment is limited by the viscous forces); here, $b = (4/3\mu + \xi)/\rho_0$. Thus, the viscosity does not change the boundaries of instability on the $\tilde{T} - \tilde{\rho}$ diagram as compared to the case described by Eq. (10).

Expressions for the instability increment of small perturbations along and across the electric field for a particular fluid are determined by the relationship between the permittivity and the density. The expressions for K are different in the cases of stratification along and across the field, which means that the instability increment depends on the orientation of perturbations. Equations (8) and (9) indicate that $K_z < K_x$ in all cases.

For nonpolar liquids obeying relation (3), we have

$$K_x = \frac{E_0^2}{12\pi}(\varepsilon - 1)^2\frac{(\varepsilon + 2)}{3\rho}. \tag{14}$$

For polar liquids, the analytical expression is rather complicated. More reliable results are provided by Eqs. (8) and (9) with experimental values of $(\partial^2\varepsilon/\partial\rho^2)_T$, which are usually positive. For both polar and nonpolar liquids, we have $K_x > 0$, which implies that the electric field increases the instability increment of perturbations of the type described by Eq. (6). At the same time, in all cases considered $K_z < 0$, so that the stability of a fluid with respect to stratification across the field increases. Thus, for $K_x > (\partial p/\partial\rho)_T$, a homogeneous fluid exhibits anisotropic decomposition into a two-phase system of cylindrical vapor channels situated in the liquid and oriented along the field.

For weakly polarizable molecules obeying the gas-type law (2), we have $K_x = 0$. Therefore, the instability in this case (as well as in the absence of an electric

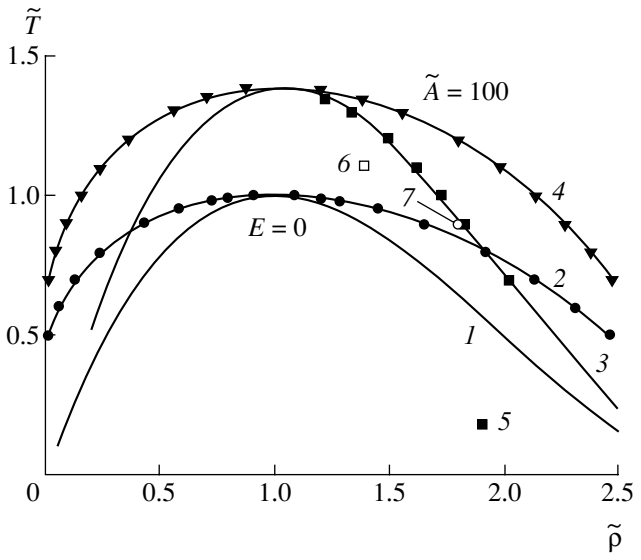

Fig. 1. The T–ρ diagrams showing (*1*, *3*) theoretical spinodals (17) for the van der Waals equation (16) and (*2*, *4*) the coexistence curves calculated (*1*, *2*) in the absence of an electric field and (*3*, *4*) in a uniform electric field for $\tilde{A}$ = 100; (*5*) points of spinodal obtained in hydrodynamic calculations using the LBE method; (*6*, *7*) states initially occurring above the critical point ($\tilde{\rho}_0 = 1.4$; $\tilde{T} = 1.1$) and in the region of stable liquid ($\tilde{\rho}_0 = 1.8$; $\tilde{T} = 0.9$), respectively.

field) is still possible only in the region of forbidden states corresponding to $(\partial p/\partial\rho)_T < 0$. Note that, even in this case, the instability is anisotropic: the stability for the stratification across the field increases because the coefficient $K_z = -D_0^2(\varepsilon - 1)^2(4\pi\varepsilon^3\rho)$ is negative.

As is known, an electric field shifts the critical point both in temperature and density [1]. The equation of spinodal $(\partial p/\partial\rho)_T = K_x$ (hydrodynamic stability) obtained using formula (10) exactly coincides with the boundary of thermodynamic stability for dielectric liquids as determined in [1]:

$$(\partial p/\partial\rho)_T = \frac{E_0^2\rho}{8\pi}\left(\frac{\partial^2\varepsilon}{\partial\rho^2}\right)_T. \qquad (15)$$

At the same time, the classical monograph [1] did not take into account the possibility that the instability can be anisotropic, which leads to stratification.

As an example, let us consider a liquid obeying the van der Waals equation of state. In dimensionless variables, this equation can be written as

$$\tilde{p} = 8\tilde{T}\tilde{\rho}/(3 - \tilde{\rho}) - 3\tilde{\rho}^2. \qquad (16)$$

For this model equation of state, an expression for the spinodal of a nonpolar dielectric liquid with the permittivity (3) can be written in the explicit form as

$$\tilde{T} = \frac{\tilde{\rho}(3 - \tilde{\rho})^2}{4}\left(1 + \tilde{A}\frac{(\alpha\rho_{cr})^2}{(1 - \alpha\rho_{cr}\tilde{\rho})^3}\right), \qquad (17)$$

where the second term in parentheses describes a temperature shift of the critical point and $\tilde{A} = E_0^2/(8\pi p_{cr})$ is the dimensionless magnitude of the electric field squared.

Figure 1 shows the T–ρ plots representing the spinodals for $E = 0$ (curve *1*) and $\tilde{A} = 100$ (curve *3*) calculated for the values of parameters corresponding to argon ($T_{cr} = 151$ K, $\rho_{cr} = 531$ kg/m^3, $p_{cr} = 4.86$ MPa, $\alpha\rho_{cr} = 0.057$). The shift of the critical point in density $\Delta\tilde{\rho}_{cr} \sim 2(\alpha\rho_{cr})\Delta\tilde{T}$, is a small due to the smallness of parameter $\alpha\rho_{cr}$.

We have also performed numerical calculations of the evolution of a homogeneous dielectric liquid that is initially at rest in a uniform electric field for the initial random density fluctuations on the order of $\Delta\rho/\rho_0 \sim 10^{-6}$ at the lattice nodes. The fluid dynamics was simulated using the method of lattice Boltzmann equations (LBE) with phase transitions for an arbitrary equation of state [6] and allowance for the action of electric forces (1) on the liquid [7]. The boundary conditions in the x axis direction were periodic; the electrodes were characterized by neutral wetting (contact angle was set to π/2). The electric field distribution was determined by solving the following system of equations:

$$\mathrm{div}(\varepsilon\nabla\varphi) = 0, \quad \mathbf{E} = -\nabla\varphi \qquad (18)$$

with the corresponding boundary conditions $\varphi = 0$ and $\varphi = E_0L_y$ on the lower and upper electrodes, respectively. The calculation area dimensions were 150 × 150 nodes.

For a nonpolar dielectric liquid with the permittivity obeying relation (3), the results of numerical calculations were used to construct the T–ρ diagrams of coexisting phases in the absence of electric field (Fig. 1, curve *2*) and in the initial vertical uniform electric field (curve *4*), as well as a part of the spinodal (points *5*). Both spinodal and binodal exhibited an upward shift, which increased with the field strength. As can be seen from these data (Fig. 1), a sufficiently strong electric field makes possible the anisotropic stratification along the field for a liquid that is initially in a metastable and even in a stable state close to the initial binodal (Fig. 1, states *6* and *7*). The same effects will be observed for polar dielectric liquids.

Indeed, the results of computer simulation showed evidence of a stratification along the uniform electric field for a fluid that was initially in the states both above the critical point (Figs. 2a and 2b) and in the liquid state (Figs. 2c and 2d).

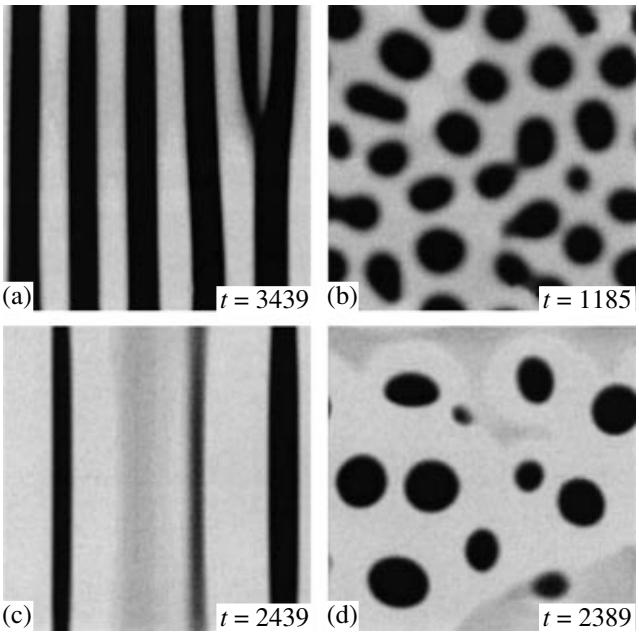

Fig. 2. Patterns illustrating the anisotropic decomposition of a dielectric liquid under the action of a uniform electric field: (a, c) stratification along the initial vertical field; (b, d) development of instability in the *xy* plane perpendicular to the field. Dark regions correspond to lower density of the fluid; (a, b) $\tilde{\rho}_0 = 1.4$; $\tilde{T} = 1.1$ (state *6* in Fig. 1); (c, d) $\tilde{\rho}_0 = 1.8$; $\tilde{T} = 0.9$ (state *7* in Fig. 1); $\tilde{A} = 100$.

The instability is developed in the form of channels of approximately cylindrical shape, which expand generating compression waves (Fig. 2d) up to the values corresponding to the curve of coexisting phases in the presence of an electric field; this is a cooperative effect in the theory of nucleation [8].

The main drawback of all preceding investigations devoted to the behavior of dielectrics in strong electric fields (see, e.g., [9]) was that the possibility of nucleation was considered only for spherical or ellipsoidal bubbles. The possibility of anisotropic instabilities already at the initial stage (immediately upon the electric field application) was ignored.

Experimental observation of the above-described principally new type of electrohydrodynamic instability of a dielectric liquid in a pure form is not a simple task, because electric breakdown of low-density channels will take place in a strong electric field simultaneously with the development of instability in the liquid phase. Apparently, these phenomena can be more readily separated for the initial states slightly above the critical point in relatively weak electric fields.

During the breakdown of dielectric liquids in strong electric fields (with local field strengths reaching 1–100 MV/cm for various liquids), the anisotropic instability described above is probably the key mechanism of the inception of streamer structures, their rapid propagation (at a velocity exceeding 100 km/s [10]) in the form of thin filaments oriented predominantly along the local electric field (Figs. 2a and 2c), and their further branching in the course of propagation.

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Translated by P. Pozdeev