Journal of Engineering Physics and Thermophysics, Vol. 79, No. 5, 2006

PULSATING MOTION OF A LIQUID IN CAPILLARIES UNDER THE INFLUENCE OF A FORCE FIELD

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UDC 532.685:537.228

The substantial influence of an inhomogeneous electric field on the processes of mass exchange in capillaries that are a pore-structure model has been found. It has been established that the electromigration of nonequilibrium films and capillary-liquid columns to the evaporation surface is observed in the region of large field gradients.

The process of internal mass exchange in different branches of engineering and technology can be optimized by creating conditions that ensure the approach of a liquid to the surface of a dispersed material, the formation of the gradients of surface tension of the liquid filling the pores in porous samples, and the separation of small moisture droplets in the initial step of dehydration. Variable or inhomogeneous external conditions of mass exchange can be created by a force field, which opens up the possibility of developing and improving moisture-removal methods not connected with the conversion of moisture into a vapor [1].

The interaction of porous materials with moisture frequently occurs under the influence of electric fields of natural and industrial origin. An inhomogeneous electric field (IEF) may substantially influence the dynamics of mass exchange in capillary-porous bodies due to the force action on both the microvolume of a liquid dielectric and the dipole molecules of a vapor [2, 3].

As is well known, systems of differential equations with allowance for the basic regularities of the thermodynamics of irreversible processes are used for description of nonstationary processes of heat and mass transfer. Practical calculations here involve great difficulties caused by the actual nonlinearity of differential equations and the absence of reliable data on thermal-moisture characteristics and phase transitions. Therefore, capillary models of different degrees of complexity are used for investigation of the distinctive features of the mode of mass exchange in a porous medium.

We consider the problem of motion of a liquid in an individual cylindrical capillary of radius R under the influence of a variable pressure gradient. For the nonstationary laminar motion of a viscous fluid along the z axis of the cylindrical capillary, the Navier–Stokes equation has the form

$$\frac{\partial \vartheta}{\partial t} - \nu \left(\frac{\partial \vartheta}{\partial r^2} + \frac{1}{r} \frac{\partial \vartheta}{\partial r} \right) = \frac{1}{\rho} f(t) , \tag{1}$$

where in the general case we have

$$-\frac{\partial p}{\partial z} = f(t) .$$

In integrating Eq. (1), we use the boundary condition

$$\vartheta = 0 \quad \text{for} \quad r = R \tag{2}$$

and the initial condition

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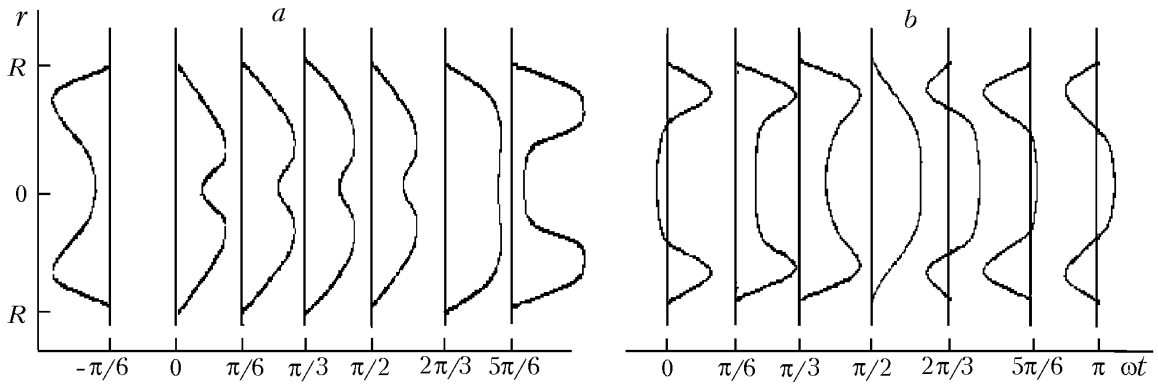

Fig. 1. Change in the velocity diagrams with time in motion of a capillary liquid in a pulsating IEF of commercial frequency: a) $R\sqrt{\omega/\nu}$ = 1.77; b) 8.85.

$$\vartheta = \vartheta_0 (r) \quad \text{for} \quad t = 0 . \tag{3}$$

For the steady-state pulsating motion corresponding to the harmonic law of change in the pressure gradient

$$f(t) = \rho A \cos \omega t ,$$

only boundary condition (2) is preserved. In the presence of a pulsating inhomogeneous electric field, the value of the periodic force $f(t)$ acting on the liquid dielectric can be found with allowance for the ponderomotive force [4] from the expression

$$f(t) = \frac{\varepsilon_0 (\varepsilon - 1)}{2} \nabla E^2 (t) . \tag{4}$$

Carrying out the replacement

$$\vartheta (r, t) = F (r, t) + \frac{A}{\omega} \sin \omega t , \tag{5}$$

in (1), we obtain the heat-conduction equation

$$\frac{\partial F}{\partial \tau} = \frac{\partial^2 F}{\partial r^2} + \frac{1}{r} \frac{\partial F}{\partial r} \tag{6}$$

with the boundary condition

$$F (R, \tau) = - \frac{A}{\omega} \sin \frac{\omega}{\nu} \tau , \tag{7}$$

where $\tau = \nu t$ is a new variable.

Thus, the problem formulated is equivalent to the problem of propagation of heat in an infinite cylinder on whose surface the temperature changes according to the law (7). The general solution of Eq. (6) (it is given, e.g., in [5]) makes it possible to obtain an expression for the velocity distribution $\vartheta(r, t)$, which contains the real Kelvin functions:

$$\vartheta (r, t) = \frac{A}{\omega} \left\{ f_1 \left[\text{bei} \left(R \sqrt{\frac{\omega}{\nu}} \right), \text{bei} \left(r \sqrt{\frac{\omega}{\nu}} \right), \text{ber} \left(R \sqrt{\frac{\omega}{\nu}} \right), \text{ber} \left(r \sqrt{\frac{\omega}{\nu}} \right) \right] \right. \sin \omega t +$$

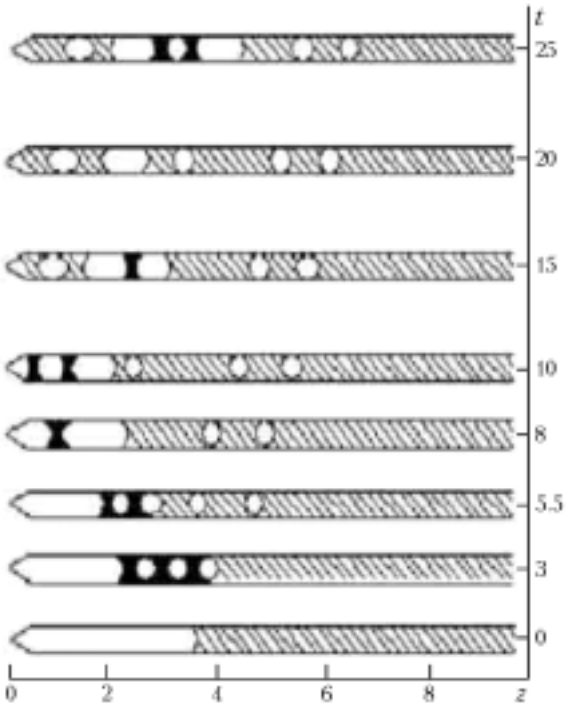

Fig. 2. Dynamics of moisture exchange in the sealed part of a capillary (R = 41.8 μm) under the influence of an IEF of strength $E = 2.7\cdot10^6$ V/m ($\nabla E = 2.1\cdot10^9$ V/m^2) at T = 294 K. t, h; z, mm.

$$+f_2\left[\mathrm{bei}\left(R\sqrt{\frac{\omega}{\nu}}\right),\mathrm{bei}\left(r\sqrt{\frac{\omega}{\nu}}\right),\mathrm{ber}\left(R\sqrt{\frac{\omega}{\nu}}\right),\mathrm{ber}\left(r\sqrt{\frac{\omega}{\nu}}\right)\right]\cos\omega t\Bigg\}. \qquad (8)$$

Equation (1), describing the straight-line, steady-state motion of a viscous incompressible fluid, contains no convective terms. It has been shown in [6] that inertial effects can be disregarded in solving capillary-absorption problems.

Some of the calculated velocity diagrams at different instants of time are given as an example in Fig. 1 for capillaries of radii R = 100 μm ($R\sqrt{\omega/\nu}$ = 1.77) (a) and R = 500 μm ($R\sqrt{\omega/\nu}$ = 8.85) (b). The value of the cyclic frequency of pulsations of the field strength taken in the calculations corresponds to the commercial-frequency voltage. The position of the curves proves that return flows occur in the capillary for the considered fluctuations of the pressure gradient. Also, we observe a considerable deviation of the meniscus profile from a parabolic form; this deviation is caused by the wall layers leading the layers near the capillary axis.

The solutions obtained have been confirmed in our investigations of the kinetics of flow of water films on the walls of a cylindrical capillary under the influence of an inhomogeneous pulsating electric field. For this purpose, we used quartz capillaries with radii of 10–50 μm obtained and prepared for experiments according to the procedure of the Institute of Physical Chemistry of the Russian Academy of Sciences [7]. The dielectric constant of the utilized quartz with a content of 99.99% SiO_2 was 3.75. The conicity of the capillaries did not exceed 10^{-5}.

We used degassed doubly distilled water in the investigations. The first distillation was carried out in a laboratory distillation setup, whereas the second one was performed in a quartz vessel. The specific electrical conductivity of the freshly prepared water was $0.5\cdot10^{-6}$ $\Omega^{-1}\cdot$cm^{-1}. Prior to filling the capillaries, we degassed the water by boiling in a quartz flask for a long time. A constant temperature during the experiments was monitored by thin copper-constantan thermocouples placed at different points near the capillary. The junctions of the thermocouples were shielded by grounded metal-foil cases from electrostatic strays. Temperature fluctuations along the capillary length did not exceed ±0.5 K. The position of the liquid menisci was determined with a KM-6 microcathetometer accurate to ±5 μm.

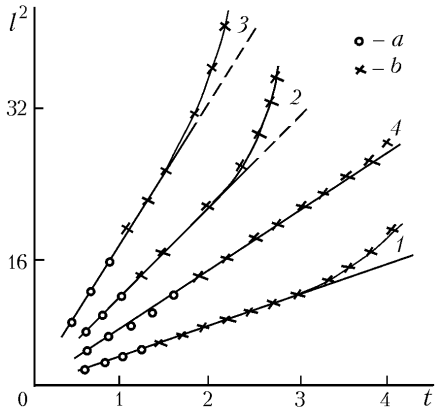

Fig. 3. Results of observations of the absorption of ethylene glycol (1–3) and transformer oil (4) into cylindrical capillaries without a field (a) and in an IEF (b): 1) R = 7.5, U = 12.4, and d = 2.2; 2) 32.1, 11.8, and 2.6; 3) 37.2, 11.4, and 2.6; 4) 25.1 μm, 10.6 kV, and 2.1 cm. l^2, cm^2; t, min.

An inhomogeneous pulsating field of commercial frequency (positive harmonics) was created with a point-plate electrode system. The plane electrode was arranged perpendicularly to the capillary under study. A capillary partially filled with liquid and sealed was placed in the field on one axis with a needle electrode. The right side of the capillary was entirely filled with water, whereas the liquid-free left side was taken to the needle electrode.

First, the stretching of a polymolecular film on the capillary walls under the influence of the gradient field occurred. Once the effective thickness of the wetting film had attained a certain value, the film became nonequilibrium. The arising fluctuations of the film thickness and the deformation of the liquid meniscus contributed to the joining of the film, i.e., to the formation of secondary liquid columns (Fig. 2). The length of the bridge columns gradually increased due to the additional feeding from the basic column under the action of the field. This meant that the liquid was pumped to the region of a larger inhomogeneity of the field via the wetting film. Subsequently, the secondary liquid columns became the source for additional feeding of new bridges that (see the figure) were formed in the vacant part of the capillary. Vapor-mixture bubbles entrapped by the menisci of the liquid moved deep into the capillary opposite to the liquid flow.

Flow of nonequilibrium films in an inhomogeneous electric field of high strength ($E = 2.7 \cdot 10^6$ V/m and $\nabla E = 1.2 \cdot 10^9$ V/m^2) and for high values of the relative humidity of air ($\varphi \geq 0.80$) was observed in experiments on evaporation of water from thin capillaries [8]. Analogous results have been obtained in experiments with polar ethylene glycol and butyl alcohol. At the same time, no influence of the inhomogeneous electric field ($E \leq 3 \cdot 10^6$ V/m and $\nabla E \leq 1.3 \cdot 10^8$ V/m^2) on the evaporation and flow of nonpolar benzene films was found.

The influence of the inhomogeneous electric field on mass exchange in capillaries apparently goes beyond the action of the mass force (4). The inhomogeneous polarization of the liquid-free internal capillary surface may play a role in the presence of the inhomogeneous electric field. In particular, it is noted in [9] that investigation of dielectrics among which is quartz is accompanied by experimental difficulties associated with the existence of an electrostatic charge that is formed on the material surface in the process of implantation of ions and electrons in electron-ion bombardment of this surface. Both point and extended charged fragments are formed.

The well-known effort to theoretically investigate liquid flow in dielectric channels disregards the influence of the solid phase on the character of charge distribution in the liquid, which frequently produces results inconsistent with experimental data. At the same time, a dielectric forming channel walls actively adsorbs charges on its surface. According to the evaluations of [10], the potential of the adsorbed charge in organic-glass channels attains several hundred kilovolts for external control fields with strengths of 10 to 15 kV. The interaction of the charged liquid and the charged walls may be quite substantial. This property of dielectrics is the stronger, the lower their conductivity.

Figure 3 shows results of observations of the absorption of ethylene glycol (curves 1–3) and transformer oil (curve 4) in the inhomogeneous electric field. The needle electrode was placed above the capillary mouth, and a hole through which a capillary was passed was made in the plate electrode. The plate electrode was above the level of the

liquid in the vessel. Selection of the liquids was determined by their fairly high viscosity, which slowed down absorption, making it possible to carry out reliable measurements of the positions of the meniscus in the capillary as a function of time. On the other hand, it was desirable to compare the absorption of liquids noticeably differing in values $\varepsilon - 1$, which determine the contribution of the inhomogeneous electric field to capillary impregnation ($\eta = 19 \cdot 10^{-3}$ Pa·sec and $\varepsilon = 38.7$ for ethylene glycol and $\eta = 23 \cdot 10^{-3}$ Pa·sec and $\varepsilon = 2.2$ for transformer oil).

As is seen in Fig. 3, first (with a switched-off field) we observed regular capillary absorption. The linear $l^2(t)$ plots without a field (points a) are in satisfactory agreement with the theory equation for thin capillaries

$$l^2 = \frac{\sigma R \cos\theta}{2\eta} t, \tag{9}$$

which points, in particular, to the constancy of the contact angle θ in each capillary in the studied range of $2.2 \cdot 10^{-3}$ to $6.8–10^{-2}$ cm/sec of the velocities ϑ. At the same time, the θ values for ethylene glycol could vary from 37 to 59° in different capillaries. The contact-angle value calculated from curve 4 corresponding to the absorption of transfer oil is 46°.

With a switched-on field (when the meniscus is in the region of action of the field), the dependence $l^2(t)$ becomes nonlinear for ethylene glycol (points b). The impregnation rate increases in connection with growth in the second term in the equation

$$\vartheta = \frac{R^2}{8\eta l}\left(\Delta p + \int_0^l f(z)\, dz\right), \tag{10}$$

obtained in [11] as a result of solution of the quasistationary problem on motion of a liquid in a cylindrical capillary in joint action of the pressure gradient and the external mass force $f(z)$. In experiments with the field ($\nabla E^2 > 0$) in the absorption step where the meniscus is to enter the region of action of the field, the position of the $l^2(t)$ curves is the same as that in experiments without a field. Subsequently, when the capillary-liquid column enters the inhomogeneous electric field, the experimental points lie higher than the points corresponding to absorption without a field, which is due to the influence of the field on the transfer of liquid mass in the capillary. For the same values of the strength, the influence of the inhomogeneous electric field is stronger in absorption into the capillaries with larger radii. The reason is that the value of $\Delta p = 2\sigma \cos\theta / R$ in expression (10) decreases with growth in R, whereas the second term related to the action of the inhomogeneous electric field is independent of the capillary radius.

The increase in the field is more considerable for large l, i.e., as the liquid meniscus approaches the needle electrode. This is consistent with Eq. (10) and is mainly due to the growth in the integral value of the mass force $f(z)$. A change in the shape of the meniscus of the liquid in an inhomogeneous pulsating electric field may decrease the gradient of capillary pressure. However, its reduction is compensated for with the second term in Eq. (10), whose value increases with the liquid approaching the needle electrode. This is confirmed by the rapid growth in experimental $l^2(t)$ curves for ethylene glycol in the inhomogeneous electric field with a high ∇E^2 gradient.

The experiments have shown that the influence of the field on the rate of absorption of ethylene glycol begins to manifest itself for $E \geq 1.2 \cdot 10^5$ V/m and $\nabla E \geq 1.3 \cdot 10^7$ V/m^2. At the same time, no influence of the inhomogeneous electric field ($E \leq 2.2 \cdot 10^6$ V/m and $\nabla E \leq 8.5 \cdot 10^8$ V/m^2) on the rate of absorption of the nonpolar transformer oil in capillaries of radii of 3 to 45 μm (Fig. 3, curve 4) has been found, which is due to the low value of the dielectric constant of the oil.

As the liquid moves to the needle electrode where the values of E and ∇E are noticeably higher, we observe the fragmentation of a capillary-liquid column. This is consistent with the solution of Eq. (1), which shows the presence of return flows (Fig. 1) contributing to the discontinuity of the liquid column and to the formation of the bubbles of the vapor–air mixture. After short vibrations, the fragments of liquid columns formed escape from the capillary mouth. The reason is that the wetting liquid film becomes stretched on the capillary walls because of the grown inhomogeneity, which leads to an increase in the cosine of the advancing contact wetting angle $\cos\theta_A$. Indeed, the calculations of the velocity of movement of a capillary-liquid column of length l [12] from the formula

$$\vartheta = \frac{R^2}{8\eta}\left(\frac{\varepsilon_0(\varepsilon-1)}{2}\nabla E^2 - \frac{2\sigma}{lR}(1-\cos\theta_A)\right)$$

show that the value of the first term becomes dominant when $\theta_A > 75^o$.

With the occurrence of a corona discharge, the surface of the volume and film liquid and the capillary walls are saturated with ions. The meniscus and the volume and film liquid are acted upon (in different directions) by the forces of electrostatic pressure, changed surface tension, and electric wind; these forces improve the wettability of the charged interior capillary surface [13] and exert an appreciable influence on the velocity of motion of the liquid column in it. Analogous effects with the fragmentation of a capillary liquid and the electromigration of the liquid in films under the action of an inhomogeneous electric field were observed in experiments on investigation of the absorption of a liquid into horizontal macrocapillaries [14].

In the calculations carried out, we have used the average values of E and ∇E because of the absence of the actual values of these quantities in quartz capillaries. To refine the solutions we must also investigate the influence of the inhomogeneous electric field on the contact wetting angle. It is of interest to evaluate the role of capillary waves on the meniscus surface and of fluctuations of the thickness of a wetting liquid film [15], which contribute to the instability of the liquid film. In this connection, a study of the phenomena of generation and intense development of self-oscillations of capillary-liquid columns in a high-gradient inhomogeneous electric field deserves attention.

Thus, constant and variable inhomogeneous electric fields may substantially influence mass exchange in capillaries that are a pore-structure model. The use of force fields is the most efficient in cases where liquids with a fairly large dielectric constant are utilized and where regular mass-exchange processes are slow. The results obtained show that the gradient fields may be used for acceleration of moistening, capillary impregnation, or drying of dispersed materials, which makes the duration of the above processes much shorter. This leads to a reduction in the energy consumption and an improvement in the efficiency of different technological processes.

NOTATION

A, amplitude value of the mass force, $N\cdot m^2/kg$; d, distance between the electrodes, m; E, electric-field strength, V/m; l, length of a capillary-liquid column, m; p, pressure of the liquid, Pa; R, radius of a capillary, m; r, radial coordinate; T, temperature, K; t, time, sec; U, electric-field voltage, V; z, vertical coordinate; ε_0 and ε, dielectric constants of vacuum and the liquid; η, dynamic coefficient of viscosity of the liquid, Pa·sec; θ, contact wetting angle, deg; ϑ, velocity of movement of the meniscus, m/sec; ν, kinematic coefficient of viscosity of the liquid, m^2/sec; ρ, density of the liquid, kg/m^3; σ, surface tension of the liquid, N/m; φ, relative humidity of air; ω, cyclic frequency, Hz.

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