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# MODEL OF FILM FLOW IN A DEAD-END CONIC CAPILLARY

**N. P. Migun**[a] **and A. I. Shnip**[b]

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*A physical theory of formation and growth of a liquid column in the dead end of a conic capillary submerged in a liquid has been constructed. It has been established that the mechanism of their phenomenon is based on the film flow (stabilized by disjoining pressure) over capillary walls because of the difference of pressures in two liquid columns due to capillary forces. The main qualitative features of the process following from the theory, namely, the constancy of the liquid flow to the vertex with time and the linear dependence of the inverse specific flow on the angle of opening of capillaries, have been confirmed experimentally.*

It is well known that a dead-end conic capillary submerged in certain liquids is filled not only on the source side of its inlet but also on the source side of its dead end, i.e., the capillary tip (vertex) having no direct contact with the liquid [1]. This phenomenon is illustrated in Fig. 1, where filling, with water, of a glass conic capillary with a radius of the mouth of 40 μm and a length of 450 μm submerged in water is shown. In a number of cases, by the moment of completion of the filling of a conic capillary with liquid, the volume of the near-vertex column is substantially larger than the volume of the column at the inlet to the capillary, which means the predominant filling of the channel on the source side of the dead end.

In [1–4], the assumption of the prevailing role of the processes of evaporation and condensation on two menisci of dissimilar curvatures and the diffusion of a vapor between them in the phenomenon of bilateral filling of conic capillaries with liquids has been made on the basis of the available experimental data. However, in [5], a significant disagreement between the rate of growth of the liquid column in the vertex of a capillary measured experimentally and the rate calculated within the framework of the theory (based on the processes of evaporation, condensation, and diffusion in the capillary channel) has been established. Thus, the problem of searching for an alternative mechanism explaining this phenomenon exists.

**Construction of the Model of Film Liquid Flow to the Vertex of a Capillary.** As the mechanism explaining the appearance and growth of a liquid column at the vertex of a capillary, we consider the flow of a liquid over a thin near-wall wetting film (this film invariably exists by virtue of the presence of disjoining pressure) under the action of the pressure difference in the inlet and near-vertex columns of the liquid due to capillary forces on menisci of dissimilar curvatures. Since, as is shown in [5], the diffusion flows of a liquid vapor are at least two orders of magnitude smaller than the liquid flows actually observed in experiments on bilateral filling, in what follows we will disregard evaporation and condensation.

The evaluations show that in typical cases characteristic of the phenomenon in question the thickness of the wetting film is of the order of several tens of nanometers, which is significantly smaller than the radius of the capillary. Owing to this, one can disregard the curvature of the underlying surface in calculating hydrodynamics and employ it instead of the curvature of the film surface in calculating capillary forces, since they differ insignificantly.

The notion of disjoining pressure in the thin liquid films on the surface of solid bodies was introduced for the first time by Deryagin [6, 7]. The presence of this pressure is due to the fact that at the interface of two adjacent phases, there are transition layers within which the properties of the liquid are different from bulk ones. Such layers exist at both the gas–liquid boundary and the liquid–solid body boundary. If the film on the surface of a solid body in a gas atmosphere is so thin that the transition layers on the two boundaries of it overlap, the stressed state in the

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[a] Institute of Applied Physics, National Academy of Sciences of Belarus, Minsk, Belarus; [b] A. V. Luikov Heat and Mass Transfer Institute, National Academy of Sciences of Belarus, Minsk, Belarus. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 75, No. 6, pp. 145–150, November–December, 2002. Original article submitted April 18, 2002.

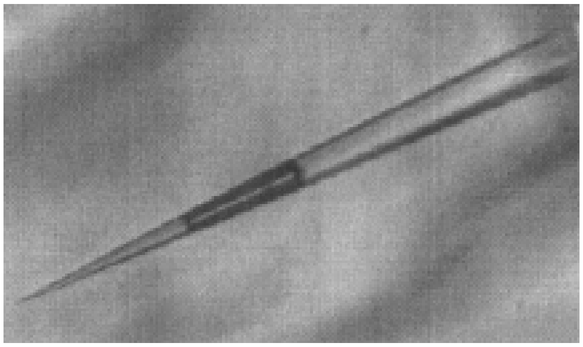


Fig. 1. Filling of a glass conic capillary (of radius 40 μm and length 450 μm) with a "Pion" indicator liquid; the capillary is submerged in this liquid.

film becomes anisotropic: the component of the stress tensor normal to the surface remains equal to the pressure of the gas (if the gas–liquid interface is plane), whereas the tangential component is a function of the distance from the surface and hence the thickness of the film. The difference of the normal component of the pressure tensor in the film $p_{\mathrm{n}}$ and the pressure in the bulk phase of the liquid $p_0$ determines the disjoining pressure of the film $\Pi = p_{\mathrm{n}} - p_0$. The dependence of this pressure on the film thickness $h$, i.e., the isotherm $\Pi(h)$, is the basic characteristic of a thin liquid film.

According to the definition of disjoining pressure, we can write

$$p_0 = p_{\mathrm{n}} - \Pi \,. \tag{1}$$

If the film is out of equilibrium and we have flow in it, the tangential component of the pressure $p_{\mathrm{t}}$ directed along the radial coordinate must change along the film (the gradient of this pressure is a driving force); consequently, it does not coincide with the pressure in the bulk phase. By analogy with (1), for the moving film we have

$$p_{\mathrm{t}} = p_{\mathrm{n}} - \Pi \,. \tag{2}$$

If the film surface is plane, the normal component of the pressure $p_{\mathrm{n}}$ is in equilibrium with the pressure in the gas phase above the film $p_{\mathrm{g}}$. In the case of a curved surface, $p_{\mathrm{n}}$ differs from $p_{\mathrm{g}}$ by the value of the capillary pressure:

$$p_{\mathrm{n}} = p_{\mathrm{g}} + \sigma K \,. \tag{3}$$

The thickness of the film $h$ is negligibly small as compared to its length $l$ (in our case $h/l \sim 10^{-4}$); therefore, we can, first, consider the problem as a local one in which the dependence of the film thickness on the longitudinal coordinate has a parametric form and, second, disregard the curvature of the underlying surface in analyzing the flow field. With allowance for this approximation, the equation of flow of the film in rectangular coordinates with transverse and longitudinal coordinates $y$ and $z$ will be written in the form

$$\mu \frac{\partial^2 u}{\partial y^2} = - \frac{\partial p_i}{\partial z} \tag{4}$$

with the boundary conditions

$$u = 0 \quad \text{for} \quad y = 0 \,, \quad \frac{\partial u}{\partial y} = 0 \quad \text{for} \quad y = h \,. \tag{5}$$

The solution of this problem has the form

$$u\,(y, z) = - \frac{1}{2\mu} \frac{\partial p_{\mathrm{t}}}{\partial z} \left[ - (y - h\,(z))^2 + h\,(z)^2 \right] . \tag{6}$$

Integrating (6) for $y$ going from 0 to $h$, we obtain the volumetric flow rate of the liquid in the film per unit length of its perimeter

$$q(z) = -\frac{h^2(z)}{3\mu}\frac{\partial p_t}{\partial z} \tag{7}$$

or with account for (2) and (3) we obtain

$$q(z) = -\frac{h^2(z)}{3\mu}\left(\sigma\frac{\partial K}{\partial z} - \frac{\partial \Pi}{\partial z}\right). \tag{8}$$

Let us apply (8) to the case of film flow in a conic capillary. Here the role of $z$ is played by the coordinate $r$ of a spherical coordinate system whose origin is brought into coincidence with the vertex of the capillary (see the figure in [5]). Using (8), we find the total rate of liquid flow through the film $Q$. For this purpose, we multiply the specific flow rate (per unit length of the perimeter) by the cross-section length. As a result, the total flow of liquid through the entire cross section of the film $Q$ is written as

$$Q = -\frac{2\pi\tau r h^3(r)}{3\mu}\frac{d}{dr}\left(-\Pi(h(r)) + \sigma K(r)\right), \tag{9}$$

where $\tau = \frac{r_0}{R_c}$.

By virtue of the stationarity of the process, the flow $Q$ must be equal in all the cross sections of the film, i.e., be independent of $r$.

For the disjoining-pressure isotherm we take the following analytical approximation [7]:

$$\Pi(h) = \frac{A}{h^3}. \tag{10}$$

The constant $A$ will be determined by approximating experimental data by dependence (10). With allowance for the remarks made above, we express the curvature of the film surface $K(r)$ in (9) as the curvature of the underlying surface:

$$K = -\frac{1}{\tau r}. \tag{11}$$

With account for (10) and (11) we can transform expression (9) to the form

$$-\left(\frac{3\mu Q}{2\pi}\frac{1}{h^3(r)} + \frac{\sigma}{r}\right)\frac{h^4(r)}{3A\tau r} = \frac{dh(r)}{dr}. \tag{12}$$

This relation should be considered as a differential equation for $h$ which must be solved in the interval $R_2 \le r \le R_1$ with boundary conditions relating the pressure in the film $p_t$ at the boundaries of the menisci to the pressure of the liquid in the corresponding column, i.e.,

$$p_t(R_1) = p_g - \frac{2\sigma}{\tau R_1}, \quad p_t(R_2) = p_g - \frac{2\sigma}{\tau R_2}. \tag{13}$$

Here the quantity $p_t(r)$ with account for (2), (3), and (10) for the disjoining pressure $\Pi$ can be expressed in terms of $h(r)$, and in such a form they can be used as the boundary conditions for (12). Since (12) is an equation of first order, it has only one integration constant. Consequently, boundary conditions (13) are sufficient to determine both the integration constant and the constant $Q$, which is unknown as yet and which we seek to determine.

Let us assume that the solution of (12) is such that it leads to the distribution of the pressure in the film $p_t$, which can be described by the dependence

$$p_t(r) = a + \frac{b}{r} . \tag{14}$$

We determine the constants $a$ and $b$ using (13). As a result, (14) is transformed to the form

$$p_t(r) = p_g - \frac{2\sigma}{\tau r} . \tag{15}$$

Expressing $p_t$ using (2) and (3) and taking account of (10) and (15), we find

$$h(r) = \left( \frac{A\tau}{\sigma} r \right)^{1/3} . \tag{16}$$

The distribution of the film thickness along the capillary (16) satisfies boundary conditions (13). At the same time, by substituting (16) into (12) we assure ourselves that (16) is its solution if $Q$ is expressed as follows:

$$Q = -\frac{4\pi A\tau}{3\mu} . \tag{17}$$

This confirms the validity of dependence (14) and yields the expression for the sought liquid flow through the film. Here the minus sign means that the liquid flow in the film is directed toward the vertex of the capillary, which does occur in actual practice. As we see, similarly to the case of a diffusion vapor flow [5], the liquid flow to the dead end of the capillary is independent of the position of the menisci $R_1$ and $R_2$, i.e., it remains constant with time.

Using the expression for the liquid flow, we can construct the time dependence of the length of the liquid column at the vertex of the capillary:

$$Q = -\pi (\tau R_1)^2 \frac{dR_1}{dt} . \tag{18}$$

With account for the condition $R_1 = 0$ at $t = 0$, we find

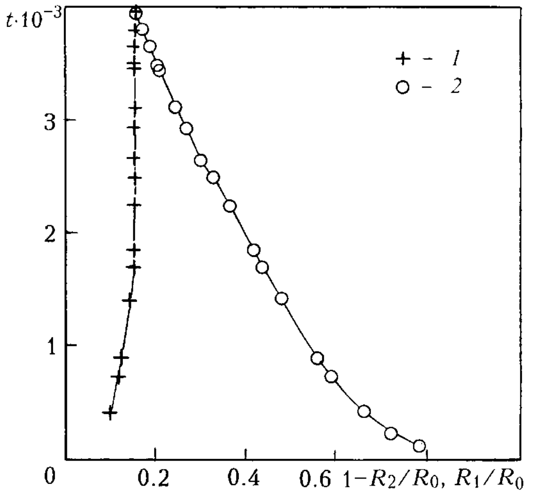
$$Qt = -\frac{\pi\tau^2 R_1^3}{3} , \tag{19}$$

whence we obtain, using (18), the relationship between the length of the liquid column at the vertex of the capillary $R_1$ and the time $t$:

$$t = \frac{\tau\mu}{4A} R_1^3 . \tag{20}$$

**Comparison with Experiment.** We investigated experimentally the filling of conic capillaries of radii $3.5\ \mu\text{m} < r_0 < 50\ \mu\text{m}$ and lengths $2 \cdot 10^2\ \mu\text{m} < R_c < 1 \cdot 10^3\ \mu\text{m}$ with doubly distilled water and ethanol. The experiments were conducted at a constant temperature of the liquid and the capillary and an ambient temperature of 22 to $25^{\circ}$C. The conic capillaries of the indicated size were manufactured by drawing from cylindrical glass intermediate products in the flame of a gas burner. Freshly drawn capillaries were placed in a cell filled with the working fluid.

The gas closed in the conic-capillary channel penetrates into the liquid as a result of its dissolution and diffusion through two liquid–gas interfaces representing two menisci. In some cases (Fig. 2), the channel of a conic capillary is filled predominantly on the source side of its vertex. The figure shows the dynamics of growth of the inlet and near-vertex water columns. In submerging the capillary of radius $r_0 = 14.6\ \mu\text{m}$ and length $R_c = 310\ \mu\text{m}$ in doubly distilled water, the volume of the main (at the inlet to the channel) liquid column (of length $(R_0 - R_1)$) ceases to in-



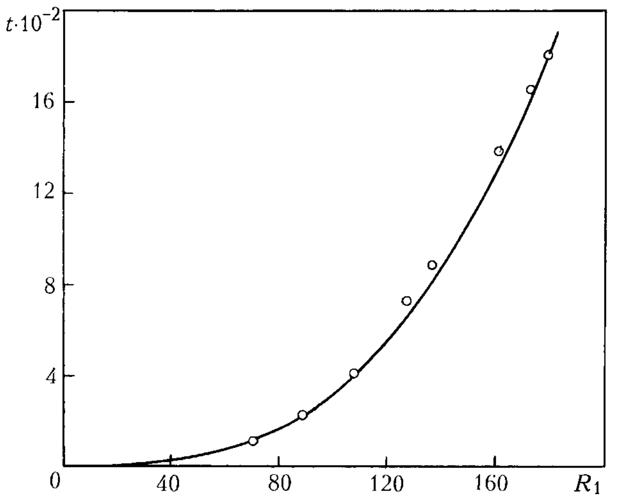
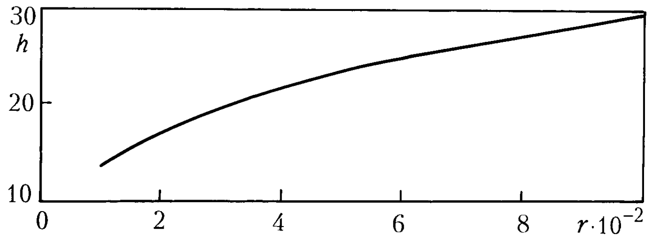


Fig. 2. Kinetics of bilateral filling of a conic capillary of length $R_c$ = 310 μm and radius $r_0$ = 15 μm with water at a temperature of 22°C: 1) inlet column; 2) near-vertex column. $t$, sec.

Fig. 3. Approximation of the experimental data on the kinetics of filling of a conic capillary of length $R_c$ = 310 μm and radius $r_0$ = 15 μm with water by dependence (20). $t$, sec, $R_1$, μm.



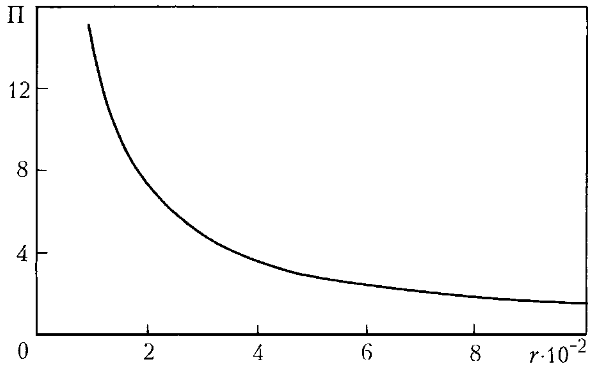


Fig. 4. Calculation of the film-thickness profile $h(r)$ along the capillary wall from formula (17). $h$, nm; $r$, μm.

Fig. 5. Calculation of the disjoining-pressure profile $\Pi(r)$ along the wall from formulas (10) and (16). Π, bar; $r$, μm.

crease. The filling of the channel with the liquid is completed due to the further growth of the near-vertex column (of length $R_1$). Thus, 61.5% of the volume of the liquid in the channel, by the moment of its total filling, arrives from the channel vertex having no direct contact with the liquid from the outside (Fig. 2).

In Fig. 3, the experimental data of Fig. 2 for the near-vertex liquid column are approximated by a dependence of the form (20), in which the value of the constant $A$ is selected so as to obtain the best agreement with these data. The value of this constant turned out to be equal to $A_e = 3.8\cdot10^{-20}$ J.

The result obtained reflects, first, the fact that the experimental data are in good agreement with dependence (20), since the liquid flow to the vertex remains constant with time. Second, the value of the constant $A_e$ corresponds in order of magnitude to the Hamaker constant describing one of the main components of the disjoining pressure.

Figure 4 shows the profile of the film thickness along the capillary wall calculated from formula (17) with the use of the above value of the constant $A_e$ and $\tau = 0.047$, while Fig. 5 shows the profile of the disjoining pressure $\Pi(r)$ calculated from (10) with account for (16) for the same case. The isotherm of the disjoining pressure calculated from formula (10) with the same value of $A_e$ (determined from experiment) is constructed in Fig. 6. The results of the

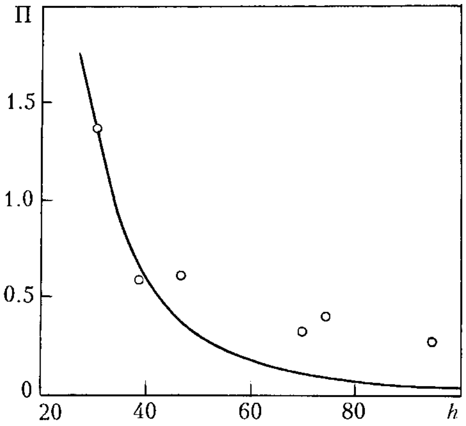


Fig. 6. Calculated curve of the disjoining-pressure isotherm Π($h$) (solid line) and experimental data [7] (points). Π, bar; $h$, nm.

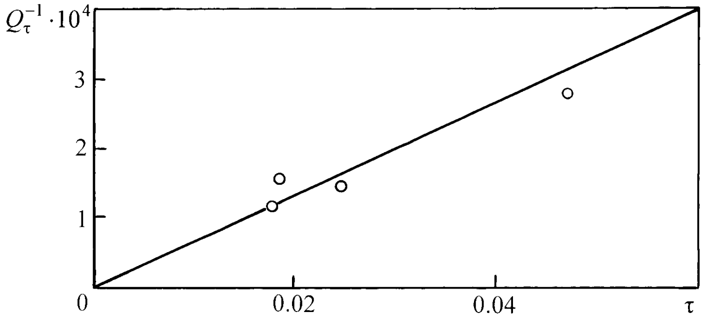


Fig. 7. Inverse specific flow of the liquid $Q_\tau^{-1}$ vs. tangent of the angle of opening of the cone. $Q_\tau^{-1}$, sec/mm$^3$.

experiments on determining the isotherm of the disjoining pressure of water films on silicate (glass) surfaces taken from [7] (Fig. 21, p. 55) are applied on the same plot. It is seen that the analytical curve for Π correlates well with the experimental data in the region of small $h$ and high Π. According to Figs. 4 and 5, it is precisely this region that is characteristic of our experiments.

The experimental data are also in good agreement with the inversely proportional dependence on τ of the quantity

$$Q_\tau = \frac{R_1^3}{t}, \tag{21}$$

which represents the specific flow of the liquid (its triple flow rate per unit solid angle of the capillary) and is predicted by formula (20).

We obtained experimental results for the kinetics of growth of the inlet and near-vertex water columns for conic capillaries of different configuration. The dependence $Q_\tau^{-1}(\tau)$ calculated, according to (20), from the formula

$$Q_\tau^{-1}(\tau) = \frac{t}{R_1^3} = \frac{\mu}{4A_e}\tau$$

is plotted in Fig. 7.

Here use is made of the value of $A_e$ given above. The values of $Q_\tau^{-1}(\tau)$ computed on the basis of (21) from experimental results for different capillaries as $t/R_1^3$ are applied on the same plot. A good correlation with the linear dependence is obvious.

Thus, a comparison of the experimental and calculated results confirms the idea that dead-end capillaries are filled on the source side of the vertex due to the film flow (stabilized by disjoining pressure) over capillary walls because of the difference of pressures in two liquid columns attributed to capillary forces.

## CONCLUSIONS

1. The mechanism of filling of dead-end conic capillaries by the liquid flowing over a thin wetting film which invariably exists by virtue of the presence of disjoining pressure explains the main qualitative features of this phenomenon well and yields quantitative results; these quantitative results with allowance for numerous factors influencing the parameter spread in such experiments are in agreement with experiment.

2. The main qualitative features of the process following from the theory, namely, the constancy of the liquid flow to the vertex and the linear dependence of the inverse specific flux on the angle of opening of capillaries, are confirmed well by experiment.

3. The analytical approximation employed in the theory for the disjoining-pressure isotherm with an undetermined parameter $A$ which is found subsequently from experiments on the kinetics of growth of the near-vertex liquid column is in good agreement with independent experiments on determination of disjoining pressure.

## NOTATION

$t$, time; $p_{\text{n}}$, normal component of the pressure tensor in the film; $p_{\text{t}}$, tangential component of the pressure; $p_0$, pressure in the bulk phase of the liquid; $\Pi$, disjoining pressure of the film; $h$, film thickness; $p_{\text{g}}$, pressure in the gas phase above the film; $\sigma$, coefficient of surface tension; $K$, curvature of the film surface; $y$ and $z$, transverse and longitudinal rectangular coordinates; $r$, radial coordinate (spherical system); $\mu$, coefficient of shear viscosity; $u$, rate; $r_0$, inlet radius of the capillary; $R_{\text{c}}$, capillary length; $R_1$ and $R_2$, coordinates of the menisci; $q$, specific (per unit length of the perimeter of the film) volumetric flow rate of the liquid in the film; $Q$, total volumetric flow rate of the liquid in the film; $Q_\tau$, reduced (to the solid angle) volumetric flow rate of the liquid in the film; $\tau$, tangent of the angle of opening of the capillary; $A$, constant in the analytical approximation of the disjoining-pressure isotherm; $a$ and $b$, integration constants. Subscripts: n and t, normal and tangential components; g, gas; c, capillary; e, experiment; 0, bulk phase.

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