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SPREADING OF A LIQUID DROPLET OVER A SOLID HORIZONTAL SURFACE

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A study is made of the kinetics of flow of the finite volume of an electrorheological liquid on a horizontal substrate.

The spreading of liquids is of importance in many physicochemical phenomena and technological processes. A liquid droplet spreading over a solid surface represents a convenient object of investigation in this field. However, it turns out to be quite difficult to describe this phenomenon because of the necessity of taking into account many different physical factors which influence the spreading of an actual droplet. In particular, of importance is correct representation of the dynamics of the processes near the boundaries of the solid, gaseous, and liquid phases. A number of approaches to the construction of a physicomathematical model of spreading of a droplet over a solid horizontal surface have been proposed at present [1–4]. The common drawback of the existing models is the fact that each of them describes just individual stages of the process of spreading rather than the entire process. Furthermore, theory ensures only a qualitative agreement with experiment, while quantitative results differ significantly. As we believe, this is due to failure to take into account the influence of the gravity forces on the kinetics of spreading of the liquid droplet over the solid horizontal surface. Below we have described the process of spreading of the droplet and have obtained relations which are in good agreement with experimental data.

The results of the experiments [4, 5] show that in spreading, a liquid droplet has the shape shown schematically in Fig. 1. Most (the central part) of the droplet holds a shape close to a spherical segment and is bounded by a convex surface with a boundary angle θ. A meniscus zone having a concave surface with a boundary angle close to the equilibrium angle θ_c is formed at the periphery of the droplet. Such a form of the exterior droplet surface indicates a uniform distribution of the pressure in the main (central) part and its redistribution in the meniscus zone, which is responsible for spreading.

Let us make some simplifications. In the process of spreading of the droplet, we will concentrate our attention on the characteristic properties of the motion of the meniscus zone relative to the solid surface, i.e., we consider the central part of the droplet to be immobile as compared to the moving meniscus. The pressure of the liquid at the inlet of the meniscus is assumed to be equal to the pressure in the central part of the droplet:

$$p = \frac{2\sigma}{r_1} \sin \theta \,. \tag{1}$$

The pressure near the external boundary of the meniscus and the solid surface is considered to be close to the equilibrium value:

$$p_c = \frac{2\sigma}{r_2} \sin \theta_c \,. \tag{2}$$

In the equations of motion of the meniscus zone, we disregard the local derivative of velocity with respect to time as compared to the viscous term and ignore inertial effects. Then the motion of the liquid in the region of the meniscus

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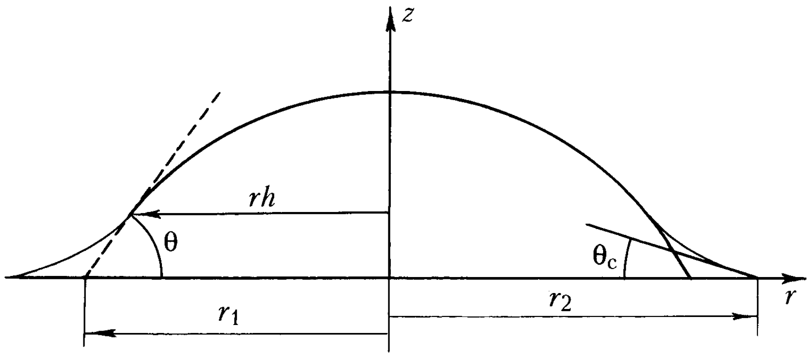

Fig. 1. Scheme of spreading of a droplet.

in the cylindrical coordinate system whose Z axis is perpendicular to the solid surface and coincides with the axis of symmetry is described by the system of equations

$$-\frac{\partial p}{\partial r}+\mu\frac{\partial^2 u}{\partial z^2}=0\,,\quad \frac{\partial p}{\partial z}=0 \tag{3}$$

with boundary conditions

$$u=0 \quad \text{for} \quad z=0\,,\quad \frac{\partial u}{\partial z}=0 \quad \text{for} \quad z=h\,. \tag{4}$$

From the second relation of system (3) it follows that $p = p(r)$; upon integration of the first equation of the system with account for boundary conditions (4) we find

$$u=-\frac{h^2}{\mu}\frac{\partial p}{\partial r}\left(\frac{z}{h}-\frac{z^2}{h^2}\right).$$

Whence for the flow rate of the liquid in the meniscus zone we obtain

$$Q=\int_0^h 2\pi r u dr=-\frac{2\pi r h^3}{3\mu}\frac{\partial p}{\partial r}\,.$$

As a result, the average velocity of motion of the meniscus zone is described by the relation

$$u=\frac{\partial r}{\partial t}=\frac{Q}{S}=\frac{Q}{2\pi r h}=-\frac{h^2(r)}{3\mu}\frac{\partial p}{\partial r}\,, \tag{5}$$

which contains two unknown quantities: $h(r)$ and $\partial p/\partial r$. To determine them we use the following considerations. From relations (1) and (2) it is easy to obtain

$$-\frac{\partial p}{\partial r}\approx\frac{p-p_c}{r_2-r_1}=\frac{2\sigma}{r_1(r_2-r_1)}\left(\sin\theta-\frac{r_1}{r_2}\sin\theta_c\right).$$

We take into account that the meniscus zone r_2-r_1 is much smaller than the main part of the droplet r_1, i.e., $r_1/r_2\approx 1$. Finally, we have

$$-\frac{\partial p}{\partial r}=\frac{2\sigma}{r_1(r_2-r_1)}(\sin\theta-\sin\theta_c)\,.$$

Transformation of formula (5) for the average velocity of motion of the meniscus yields

$$\bar{u} = \frac{\partial r_1}{\partial t} = \frac{2\sigma h^2 (r)}{3\mu r_1 (r_2 - r_1)} (\sin\theta - \sin\theta_c) . \tag{6}$$

Let us find the height of the droplet $h_1 = h(r_1)$ at the entry to the meniscus zone. The tangential friction stress is equal to zero on the free surface of the meniscus (see (4)). Therefore, the meniscus shape is determined only by the surface tension of the liquid σ and the gravity force ρg. Then to find the shape of the meniscus surface we can employ the equation of a static meniscus

$$\frac{d^2 r_h}{dz^2}\left[1 + \left(\frac{dr_h}{dz}\right)^2\right]^{-3/2} = \frac{\rho g z}{\sigma} \tag{7}$$

with boundary conditions

$$\frac{dr_h}{dz} = -\operatorname{ctan}\theta_c \quad \text{for} \quad z = 0 , \quad \frac{dr_h}{dz} = -\operatorname{ctan}\theta \quad \text{for} \quad z = h_1 .$$

Single integration of (7) yields

$$\frac{dr_h}{dz}\left[1 + \left(\frac{dr_h}{dz}\right)^2\right]^{-1/2} = \frac{\rho g z^2}{2\sigma} + C ,$$

whence, with account for the boundary conditions, we represent the height of the droplet at the entry to the meniscus zone as

$$h_1 = \left(\frac{2\sigma}{\rho g}\right)^{1/2} (\cos\theta_c - \cos\theta)^{1/2} . \tag{8}$$

The quantity h_1 is the characteristic dimension of the meniscus zone; therefore, we assume that

$$r_2 - r_1 = ah , \tag{9}$$

where the proportionality factor a can be obtained either by numerical solution of Eq. (7) or from experimental data. We substitute relations (8) and (9) into formula (6). As a result, the kinetic equation describing the process of spreading of the liquid droplet over the solid horizontal surface takes the form

$$\frac{dr_1}{dt} = \frac{2\sigma}{3a\mu}\left(\frac{2\sigma}{\rho g}\right)^{1/2} \frac{1}{r_1} (\sin\theta - \sin\theta_c) (\cos\theta_c - \cos\theta)^{1/2} . \tag{10}$$

The relationship between the boundary angle θ of the spherical segment (main part of the droplet) and the radius of its base r_1 will be determined from the trigonometric equality

$$\frac{6V}{\pi r_1^3} = 3\tan\frac{\theta}{2} + \tan^3\frac{\theta}{2} , \tag{11}$$

where $V = m/\rho$. The system of equations (10) and (11) describes the kinetics of spreading of the droplet in the region of both the obtuse boundary angles ($\theta > \pi/2$) and the acute angles ($\theta < \pi/2$) in a unified context. To assure ourselves that this is true we introduce the dimensionless variables $R = r_1/r_b$ (dimensionless radius, where $r_b = (3V/2\pi)^{1/3}$ is

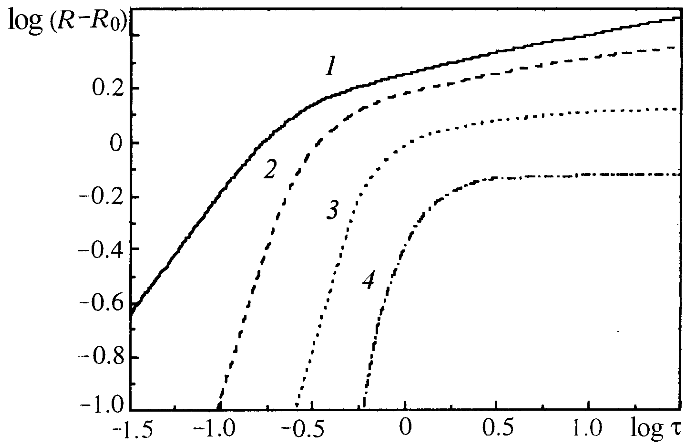

Fig. 2. Kinetics of spreading of a droplet: 1) θ_c = 0; 2) 0.174; 3) 0.523; 4) 0.872 rad.

the radius of the base of the droplet when it has the shape of a hemisphere) and $\tau = t\,\dfrac{2\sigma}{3\mu a}\left(\dfrac{2\sigma}{\rho q}\right)^{1/2}\left(\dfrac{2\pi}{3V}\right)^{2/3}$ (dimensionless time). Then we represent the system of equations (10)–(11) in dimensionless form as

$$\frac{dR}{d\tau}=\frac{1}{R}(\sin\theta-\sin\theta_c)(\cos\theta_c-\cos\theta)^{1/2}\,,\quad \frac{4}{R^3}=3\tan\frac{\theta}{2}+\tan^3\frac{\theta}{2}\,. \tag{12}$$

Let us analyze system (12). First we consider the case where the equilibrium boundary angle θ_c is small, i.e., $\theta_c \approx 0$. As a result, in spreading of the droplet in the region of obtuse boundary angles $\theta = \pi - \alpha$, for small α from system (12) we obtain

$$\frac{dR}{d\tau}=\frac{\alpha\sqrt{2}}{R}\,,\quad \frac{4}{R^3}=\left(\frac{\alpha}{2}\right)^3. \tag{13}$$

Integration of (13) with the initial condition R = 0 at τ = 0 leads to the expression

$$R=\frac{2\sqrt{2}}{4^{1/3}}\,\tau\,. \tag{14}$$

In spreading of the droplet in the region of acute boundary angles $\theta < \pi/2$, for low values of θ from (12) we find

$$\frac{dR}{d\tau}=\frac{1}{\sqrt{2}}\frac{\theta^2}{R}\,,\quad \frac{4}{R^3}=\frac{3}{2}\theta\,. \tag{15}$$

Solving this system with the initial condition $R = R_0$ at τ = 0, we obtain

$$R^8-R_0^8=\frac{512}{9\sqrt{2}}\,\tau\,.$$

The last relation for $R^8 >> R_0^8$ enables us to write

$$R=1.587\tau^{1/8}\,. \tag{16}$$

Thus, from formulas (14) and (16) it follows that for $\theta_c \approx 0$ the kinetics of movement of the perimeter of the droplet changes from the dependence $R \sim \tau$ in the region of obtuse boundary angles $\theta > \pi/2$ to the dependence $R \sim \tau^{1/8}$ in the region of acute boundary angles $\theta < \pi/2$. This result has been obtained in the unified context and it is in good agreement with numerous experimental data [2, 4].

Let us turn back to the system of equations (12). It follows that the functional dependence of the kinetics of spreading in the region of both the obtuse and acute boundary angles θ is mainly affected by the equilibrium boundary angle θ_c. Figure 2 shows plots of the kinetics of spreading of the liquid droplet over the solid surface which have been obtained by numerical solution of system (12) for different values of θ_c. Noteworthy is the fact that, when $\sin\theta \le \sin\theta_c$, the liquid droplet remains immobile in both the region of obtuse ($\theta > \pi/2$) and acute ($\theta < \pi/2$) boundary angles but it is spreading when the condition $\sin\theta > \sin\theta_c$ is fulfilled.

In closing, we dwell on evaluation of the proportionality factor *a*. From Eqs. (7) and (8) we can state that it is independent of the viscosity of the liquid μ and the volume of the droplet *V*. This theoretical conclusion is in agreement with experimental data [4]. Moreover, if we assume that *A* is equal to the numerical value of the ratio $|\rho g/\sigma|$, we obtain a satisfactory quantitative agreement between theory and experiment on the kinetics of spreading of the droplet on a solid horizontal surface. Thus, for example, in experimental investigation of the spreading of droplets of polymethylsiloxane liquids with $\theta_c \approx 0$ over horizontal substrates in the region of acute boundary angles θ, the empirical equation $R = A\tau^{1/8}$ [4] has been obtained, which totally coincides with the theoretical formula (16). With a change of 40 times in the viscosity μ and a change of 10 times in the droplet volume *V* the values of the constant *A* varied within A = 1.543–1.603 for different types of solid substrates (lavsan, glass, aluminum). A comparison of these experimental values with theoretical values (A = 1.587 from formula (16)) shows their quite satisfactory agreement.

NOTATION

A, proportionality factor; *C*, constant; *g*, free-fall acceleration; *h*, transverse coordinate of the meniscus surface; *m*, droplet mass; *p*, pressure; *Q*, flow rate; $r_{1,2}$, droplet radius; *r* and *z*, longitudinal and transverse coordinates; r_h, longitudinal coordinate of the meniscus surface; *S*, cross-sectional area of the meniscus; *t*, time; *u*, velocity; *V*, volume of the liquid droplet; α, certain angle; θ, boundary angle; θ_c, equilibrium boundary angle; μ, dynamic viscosity; ρ, density; σ, surface tension. Subscripts: 1 and 2, without a dynamic meniscus and with it; c, constant; b, base; 0, initial value.

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