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MECHANISM OF GROWTH AND SEPARATION OF AXISYMMETRIC DROPS AND BUBBLES

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A practical solution is obtained for the axisymmetric problem of quasistatic drop and bubble growth.

In practice, processes of mass transfer and heat exchange between drop-forming liquid and gaseous disperse media depend on the interaction of the two media. The bubbling of gases through liquids is a technique widely used in chemical engineering while air conditioning technology is based on producing a liquid spray in a vapor–air mixture [1-11]. The interaction of two media on the developed surface of packings finds application in various branches of industry. The interaction of the two media depends not only on the temperature difference, the partial pressure difference and the area of the contact surface but also on a number of other factors, such as capillary, surface and hydrodynamic effects. If the relative motion of the media in direct contact is due to the action of gravitational forces, then the influence of the velocity factor on the mass-transfer process is conditioned by the physical properties of the media and can be intensified by developing the contact surface. Accordingly, the question of the formation of the contact surface and the capillary effects on that surface are of considerable significance in connection with the solution of engineering problems.

Let us consider an axisymmetric drop of liquid suspended over a calibrated orifice of sufficiently small diameter in a plate of solid material. To this there corresponds a gas bubble formed over the orifice in a device made in the form of an individual nozzle, plate, or capillary. The diameter of the orifice is so selected that the velocity head of the liquid or gas is balanced by the resistance to the motion of the drop or bubble (Fig. 1). Then the total energy of the object at any moment of time $t \leqslant t_{sep}$ is composed of the surface energy and the energy of the force of gravity for drops or the energy of the buoyancy force for gas bubbles. Accordingly, the energy functional takes the form

$$W(t) = \int_D \left\{\sigma_{12} - \sigma_{10} + \sigma_{20}\left(\sqrt{1 + p^2 + q^2} - \frac{z^2}{2a^2}\right)\right\} dxdy + \lambda \int z dxdy, \quad (1)$$

where $z = z(x, y, t)$ is the equation of the surface and $p = \partial z/\partial x$; $q = \partial z/\partial y$; $a^2 = \sigma_{20}/n \quad (\rho_l - \rho_g)g$ is a certain parameter with the dimension of the square of length, since ρ_l and ρ_g are the densities of the liquid and the gas, respectively; σ is the surface tension at the interface of the corresponding phases; λ is the Lagrange multiplier taking into account the invariability of the volume at time t in seeking the extremal of functional (1).

The axisymmetry makes it possible to go over from the two-dimensional domain to the plane problem by making the substitution $r = \sqrt{x^2 + y^2}$. Then (1) takes the form

$$W(t) = 2\pi \int_0^r \left\{\sigma_{12} - \sigma_{10} + \sigma_{20}\left(\sqrt{1 + z_r'^2} + \lambda z\right)\right\} r dr. \quad (2)$$

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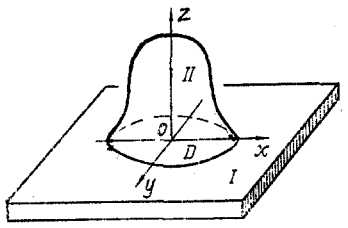

Fig. 1. "Drop" on plate.

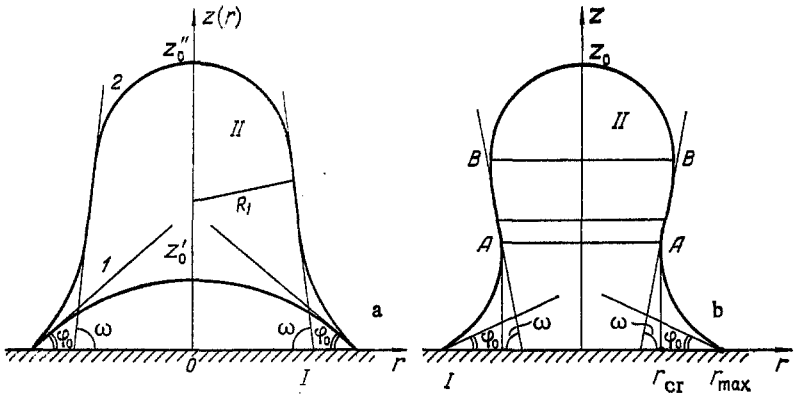

Fig. 2. Meridional section through "drop": 1) first stage; 2) second (a) and third (b) stages.

We write the expression for the variation of the functional (2)

$$\delta W(t) = (F - z'F_{z'})\,\delta_r|_0^r + F_{z'}\delta_z|_0^r + \int_0^r \left\{F_z - \frac{d}{dr}(F_{z'})\right\}\delta_z dx, \tag{3}$$

where $F\{r_1 z(r),\ z_r'(r)\}$ is the integrand expression in (2).

The vanishing of the variation gives the Neumann condition for the contact angles

$$\cos\varphi_0 = \frac{\sigma_{10} - \sigma_{12}}{\sigma_{20}} \tag{4}$$

and the Euler equation for the contour of the drop

$$\frac{z''(r)}{(1+z'^2)^{3/2}} + \frac{z'(r)}{r\sqrt{1+z_r'^2}} = -\frac{z(r)}{a^2} + \lambda. \tag{5}$$

Introducing the parameter $\varphi = \arctan z'(r)$, we write (5) in the form

$$\frac{d}{dr}(\sin\varphi) + \frac{\sin\varphi}{r} = -\frac{z(r)}{a^2} + \lambda, \tag{6}$$

where $k\pi - \pi/2 \leqslant \varphi \leqslant k\pi + \pi/2;\ k = 0, 1, 2, 3\ldots$

Since finding the exact solution of Eq. (4) presents considerable mathematical difficulties, it is necessary to establish the upper and lower bounds of the exact solution. The upper bound is given in [12]. To find the lower bound we express (4) in the form

$$\frac{a}{R} + \frac{a}{R_1} = -\int \frac{R}{a}\sin\varphi d\varphi + \lambda a, \tag{7}$$

where R and R_1 are the radii of curvature of the "drop."

For sufficiently small angles $\varphi = \arctan z'(r)$ and large radii of revolution of the points $\{r_1 z(r)\}$, Eq. (7) describes the two-dimensional "drop" [13]. Solving it, in this case we have

$$R = \pm\frac{\sqrt{2}}{2}\,a\,\frac{1}{\sqrt{L+\cos\varphi}}\begin{cases} r = \pm\dfrac{\sqrt{2}}{2}\,a\displaystyle\int_0^{\varphi}\frac{\cos\varphi}{\sqrt{L+\cos\varphi}}\,a\varphi, \\ z = \lambda a^2 \pm \sqrt{2}\,a\sqrt{L+\cos\varphi}, \end{cases} \tag{8}$$

where $\lambda = \dfrac{\sqrt{2}}{2}\sqrt{L+\cos\varphi_0}$; φ_0 is the value of the contact angle, and the constant of integration L is determined from the expression

$$\frac{z_0}{\sqrt{2}a} = \sqrt{1+L} \mp \sqrt{L+\cos\varphi_0}. \tag{9}$$

From (7) for determining the constant L we obtain

$$L = \frac{\left[\frac{z_0^2}{2a^2} - (1 + \cos\varphi)\right]^2 - 4\cos\varphi_0}{4\frac{z_0^2}{2a^2}} = -\cos\omega, \tag{10}$$

where $\omega = \arccos L$.

On the basis of the above, we can divide the process of growth and separation of axisymmetric drops and bubbles into three successive stages.

First Stage. The drop or bubble grows, the contact angle increasing to the equilibrium value and the contact radius to its maximum; for a drop the maximum of the contact angle

$$\varphi_0^{\max} = \arccos\frac{\sigma_{sv} - \sigma_{sl}}{\sigma_{lv}},$$

and for a gas bubble

$$\varphi_0^{\max} = \arccos\frac{\sigma_{sl} - \sigma_{sv}}{\sigma_{lv}},$$

where σ_{sl}, σ_{sv}, and σ_{lv} are the surface tensions at the solid–liquid, solid–gas, and liquid–gas interfaces, respectively. The growth of the drop or bubble with increase in contact radius will continue until the sum of the surface energy and the force of gravity directed along the axis of symmetry is equal to zero, the curvature of the contour at the points $(r_{max}, 0)$ is also equal to zero and $\omega = \varphi_0$, $\lambda = 0$. In the first stage the maximum height to which the drop or bubble rises will be

$$z_0^{\max} = \sqrt{2}\,a\sqrt{1 - \cos\varphi_0^{\max}}. \tag{11}$$

The maximum volume is calculated by integrating Eq. (6) for $\varphi_0 = \varphi_0^{\max}$:

$$V_I^{\max} = 2\pi a^2 r_{\max} |\sin\varphi_0^{\max}|. \tag{12}$$

Second Stage. The growth of the drop or bubble proceeds at the expense of changes in its surface. The radius and the contact angle remain unchanged and equal to r_{max} and $\varphi_0^{\max}$. The inflection of the contour curve, i.e., the vanishing of the curvature and the reversal of its sign, occurs at the points where $|\varphi| = \omega$, $\lambda \neq 0$ (Fig. 2a):

$$z_{cr} = \sqrt{2}a\sqrt{\cos\varphi_0^{\max} - \cos\omega};$$

$$r_{cr} = \pm\frac{\sqrt{2}}{2}\,a\int_0^{\omega}\frac{\cos\varphi}{\sqrt{\cos\varphi - \cos\omega}}\,d\varphi. \tag{13}$$

We will call the section of the "drop" with radius r_{cr} the critical section $z = z_{cr}$ and write the equations of the contour

$$\begin{cases} z = \sqrt{2}\,a\{\sqrt{\cos\varphi_0 - \cos\omega} + \sqrt{\cos\varphi - \cos\omega}\}, & z \geqslant z_{cr} \\ r = \pm\dfrac{\sqrt{2}}{2}\,a\displaystyle\int_0^{\varphi}\frac{\cos\varphi}{\sqrt{\cos\varphi - \cos\omega}}\,a\varphi, & |\varphi| \leqslant |\omega|; \end{cases} \tag{14}$$

$$\begin{cases} z = \sqrt{2}\,a\{\sqrt{\cos\varphi_0 - \cos\omega} - \sqrt{\cos\varphi - \cos\omega}\}, & 0 \leqslant z \leqslant z_{cr} \\ r = \pm\dfrac{\sqrt{2}}{2}\,a\left\{\displaystyle\int_0^{\omega}\frac{\cos\varphi\,d\varphi}{\sqrt{\cos\varphi - \cos\omega}} + \int_{\varphi}^{\omega}\frac{\cos\varphi\,d\varphi}{\sqrt{\cos\varphi - \cos\omega}}\right\}, & |\varphi_0^{\max}| \leqslant |\varphi| \leqslant |\omega|. \end{cases}$$

By integrating Eq. (6) we calculate the total volume of the "drop":

$$V_2 = 2\pi r_{cr} a^2 \sin\omega + \pi r_{cr}^2 z_{cr} + \sqrt{2}\,\pi a/r^2\sqrt{\cos\varphi - \cos\omega} - \sqrt{2}\arcsin\varphi\Big|_{\varphi=\omega}^{\varphi=\varphi_0^{\max}}. \tag{15}$$

The critical volume, i.e., the volume for $z \geqslant z_{cr}$, we take equal to

$$V_{cr} = 2\pi r_{cr} a^2 \sin\varphi. \tag{16}$$

Separation of the "drop" can also occur in the second stage, even when the energy functional (2) is positive, i.e., when the forces of cohesion hold the "drop" at the surface of the solid. In this case for the critical section we require satisfaction of the necessary condition of onset of separation of the "drop"

$$n(\rho_l - \rho_g) g V_{cr} = 2\pi\sigma_{20} r_{cr}, \tag{17}$$

where n is a number showing by how many times the force of gravity exceeds the force of surface tension. Condition (17), together with expression (16), makes it possible to determine the precise onset of separation, i.e., the start of the third stage, when $|\sin\omega| = 1$ and $|\omega| = 90°$, which is confirmed by the experimental data [14].

Third Stage. The growth of the media contact surface proceeds not only as a result of increase in the mass of the object, but also as a result of its separation, the separation rate being limited only by the viscosity of the liquid. If the total energy functional (2) is nonnegative, the radius and contact angle will remain the same as in the second stage (Fig. 2b). In order to describe the contour by means of Eqs. (8) we find the constant of integration L from relation (10). We note that in the third stage its sign is reversed; at the point $\varphi = \operatorname{arctg}(\pm\infty)$ the angle φ has a jump equal to π radians. The points $|\varphi| = \omega$ and $|\varphi| = \pi - \omega$ are called the points of inflection; accordingly, the contour equations take the form

$$\begin{cases} z = \sqrt{2}\, a\{\sqrt{\cos\varphi_0 + \cos\omega} + \sqrt{\cos\varphi + \cos\omega}\}, \ 0 \leqslant |\varphi| \leqslant \pi - \omega, \\ r = \pm\dfrac{\sqrt{2}}{2}\, a \displaystyle\int_0^{|\varphi|} \frac{\cos\varphi}{\sqrt{\cos\varphi + \cos\omega}}\, d\varphi, \ 0 \leqslant |\varphi| \leqslant \pi/2, \end{cases}$$

$$\begin{cases} z = \sqrt{2}\, a\{\sqrt{\cos\varphi_0 + \cos\omega} - \sqrt{\cos\varphi + \cos\omega}\}, \ \varphi_0 \leqslant |\varphi| \leqslant \pi - \omega, \\ r = \pm\dfrac{\sqrt{2}}{2}\, a\left\{\displaystyle\int_0^{\omega} \frac{\cos\varphi d\varphi}{\sqrt{\cos\varphi + \cos\omega}} \pm \int_0^{|\varphi|} \frac{\cos\varphi d\varphi}{\sqrt{\cos\varphi + \cos\omega}}\right\}, \ \varphi_0 \leqslant |\varphi| \leqslant \pi - 0. \end{cases} \tag{18}$$

By analogy with (16), by integrating Eq. (6) we can calculate the volume up to the separation section (A–A) (Fig. 2b):

$$V_{sep} = \sqrt{2}\,\pi a \sqrt{\cos\varphi_0 + \cos\omega}\,(r_B^2 - r_A^2) + 2\pi a^2 (r_B - r_A) + \pi r_B^2 (z_B - z_A), \tag{19}$$

where $\pm r_B$, z_B; $\pm r_A$, z_A are the contour points of sections B–B and A–A, which can be calculated from Eqs. (18). Complete separation occurs when for the separation section the following condition is satisfied:

$$\frac{npgV_{sep}}{\pi r_A^2} = E, \tag{20}$$

where E is the isothermal modulus of elasticity of medium II (see Fig. 1).

Since the modulus of elasticity of most liquids lies in the range 10^9–10^{10} N/m^2, while for gases it is equal to the pressure inside the bubble [15], we may conclude that the bubble separation time will be largely determined by the rate of flow of liquid over the interphase boundary and will depend very little on the properties of the gas. For a suspended drop, on the other hand, the separation time will depend on the flow of liquid inside the drop and be practically independent of the properties of the surrounding gas. Thus, knowing the modulus of elasticity of a given liquid, from relations (20), (19), (18), (14) and (10) we can obtain an estimate of the separation time, more accurate for a drop and less so for a bubble.

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KINETIC THEORY OF DIFFUSION IN LIQUID AND GAS MIXTURES

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Expressions for evaluating diffusion coefficients in real multicomponent solutions are obtained on the basis of the kinetic theory of dense gases and liquids.

For multicomponent mixtures, molecular mass transport is determined from [1] $\mathbf{j} = -\mathbf{D} \cdot \operatorname{grad} \mathbf{y}$, where $\mathbf{D}$ is the diffusion coefficient matrix of multicomponent systems.

Since most mass-exchange processes of separation, mixing, and chemical transformation occur, as a rule, in multicomponent systems, to calculate these processes information is needed on the diffusion coefficient matrix, providing the total pattern of mass transport. There are quantitative and qualitative distinctions between multicomponent and binary diffusion [2, 3]. However, due to the absence of information on $\mathbf{D}$ the available methods of evaluating processes in multicomponent systems operate only with a single coefficient of binary diffusion. This situation restricts the development of mass transfer theory in multicomponent systems and the creation of justified methods of a computational structure.

It must also be noted that experimental data on diffusion coefficients in a wide interval of concentrations and temperatures, necessary for the calculations, are absent even for binary mixtures. Theoretical methods of calculating transport coefficients in liquid mixtures are far from complete. Thus, the more recently developed kinetic theory of multicomponent dense gases and liquids for rigid sphere models [4] with the use of a radial distribution function [3] generally renders the basic transport characteristics in ideal mixtures. For this reason, this theory does not fully include the complexity of molecular interactions, and the agreement between calculated and experimental values of diffusion coefficients in nonideal systems is very poor. Therefore, further development of the kinetic theory must occur in the direction of a refined intermolecular interaction in real solutions.

One of the methods of taking into account the real molecular interaction in liquid mixtures, more precisely the presence of many-particle interactions and real shapes of intermolecular forces, as well as capabilities of formation of associated complexes, is their account in describing collision terms of the kinetic equations, which is still an unresolved problem. Another method, recently developed, consists of using in the solution of the kinetic

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