EFFECT OF CAPILLARY PHENOMENA ON THE RHEOLOGICAL PROPERTIES OF EMULSIONS

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The rheological properties of moderately concentrated emulsions are determined with allowance for surface tension. Cases of low and high surface tension at the phase boundary of the emulsion are analyzed.

The rheological properties of emulsions have begun to be intensively studied in recent years. The effective viscosity of dilute emulsions is usually determined from the classical Taylor formula [1]. Different generalizations of this formula for concentrated systems are discussed in [2]. However, the Taylor model pertains only to the case when surface tension α is large enough so as to prevent any change in the form of the drops. This condition may be violated in practice, which, as will be shown below, may lead not only to a change in effective viscosity, but also to the creating of non-Newtonian properties in the emulsion. The authors of [3, 4] analyzed the effects which develop due to capillary phenomena on the surface of drops of emulsions in solutions of surfactants. In the present investigation, we study the effect of capillary phenomena due to comparability of capillary forces and viscous stresses in the dispersion medium on the effective viscosity of emulsions and the form of their rheological equation of state.

The relationship between the deviatoric part of the hydrodynamic stress σ and the tensor of the strain rates e is given in the form

$$\boldsymbol{\sigma} = 2\eta \mathbf{e}, \tag{1}$$

where η is the effective viscosity of the emulsion. This quantity can be calculated from the relation [5]

$$2(\eta - \eta_0) e_{ij} = \frac{3a^3\rho}{4\pi}\Big[\int\limits_{|\mathbf{r}|=a} r_i n_l \sigma_{lj}^{+} d\mathbf{r} - \eta_0 \int\limits_{|\mathbf{r}|=a} (n_i v_j^{+} + n_j v_i^{+})\, d\mathbf{r}\Big], \quad i,\ j,\ l = x,\ y,\ z \tag{2}$$

(summation is performed over repeating indices).

Integration is performed in (2) over the surface of a certain sample drop. In determining σ^+ and v^+ on the surface of this drop, it is necessary to consider the presence of surrounding drops. The complexity of such a problem was noted in [3, 4, 6, 7]. In light of this, various schematizations of the hydrodynamic flow around an isolated drop have been used. In the case of limitingly dilute systems ($\rho \ll 1$), the mutual effects of the drops are ignored and it is assumed that the sample drop in located in an infinite volume of the dispersion medium. It is further assumed that the velocity of the flow at an infinite distance from this drop coincides with the mean velocity of the emulsion in the region being examined. For mixtures with a moderate concentration of dispersed liquid particles ($\rho \le 0.2$-0.25), we can use a scheme in which an isolated spherical drop is assumed to be immersed in a homogeneous hypothetical medium whose properties coincide with the properties of the effective medium as a whole [8]. In the present study, we perform our calculations by means of the latter scheme.

If we assume that the equilibrium theory of surface tension is applicable to the phenomenon we are examining, then the only effect of surface tension α will be discontinuity of the stress at the interface [9]. Thus, at each point on the surface of the sphere

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$$-p^* + \sigma_{rr}^* = -p^+ + \sigma_{rr}^+ + P_\alpha, \quad P_\alpha = \alpha\left(\frac{1}{R_1} + \frac{1}{R_2}\right), \tag{3}$$

where R_1 and R_2 are principal radii of curvature of the interface; p^* and p^+ are the pressures inside and outside the spherical drop, respectively.

With allowance for (3), the problem of determining the characteristics of the hydrodynamic field near the sample drop for moderately concentrated emulsions can be formulated as:

$$\begin{gathered}
-\nabla p^+ + \eta\Delta \mathbf{v}^+ = 0, \quad \operatorname{div} \mathbf{v}^+ = 0, \quad r \geqslant a, \\
-\nabla p^* + \eta_1\Delta \mathbf{v}^* = 0, \quad \operatorname{div} \mathbf{v}^* = 0, \quad r \leqslant a, \\
v_n^* = v_n^+, \quad v_t^* = v_t^+, \quad -p^* + \sigma_n^* = -p^+ + \sigma_n^+ + P_\alpha, \\
\sigma_t^* = \sigma_t^+, \quad r = a, \quad \mathbf{v}^*, \quad p^* < \infty, \quad \mathbf{v}^+ \to \mathbf{v}, \quad r \to \infty.
\end{gathered} \tag{4}$$

The formulation of (4) differs from the well-known formulations in that no provision is made here for satisfaction of the condition $v_n^* = v_n^+ = 0$ at $r = a$.

In the state of pure shear, the velocity of the external flow can be given in the form $v_x = Ex$, $v_y = -Ey$, $v_z = 0$, $E = \text{const}$, i.e., the nontrivial components of the tensor E are $e_{xx} = E$, $e_{yy} = -E$.

We introduce a spherical coordinate system with its origin at the center of the drop. Then, analogous to [10], the components of the mean velocity of the medium in pure shear are written in the form

$$v_r = Er\sin^2\theta\cos 2\varphi, \quad v_\theta = Er\sin\theta\cos\theta\cos 2\varphi, \quad v_\varphi = -Er\sin\theta\sin 2\varphi. \tag{5}$$

The general solution of the equation div v = 0, with an angular part that coincides with the angular part of (5), has the form

$$\begin{gathered}
v_r = \left(A_1 r - 3A_2 r^3 + \frac{3A_3}{r^4} + 3\frac{A_4}{r^2}\right)\sin^2\theta\cos 2\varphi, \\
v_\theta = \left(A_1 r - 5A_2 r^3 - \frac{2A_3}{r^4}\right)\sin\theta\cos\theta\cos 2\varphi, \\
v_\varphi = -\left(A_1 r - 5A_2 r^3 - \frac{2A_3}{r^4}\right)\sin\theta\sin 2\varphi,
\end{gathered} \tag{6}$$

where A_i ($i = 1, 2, 3, 4$) are unknown coefficients subject to determination; r, θ, φ are spherical coordinates. Equations (6) are written in general form and are valid for an inhomogeneous medium. They do not apply to the effective medium.

The radial components of (6) can be written separately for a spherical drop and for the region outside it. Considering the boundedness of v at $r \to 0$ and $r \to \infty$ in accordance with conditions (4), in Eqs. (6) in the region inside the drop we need to ignore the terms containing the coefficients A_3 and A_4. The terms with A_2 should be absent outside the drop in this case. Then

$$\begin{gathered}
v_r^* = A_1 r - 3A_2 r^3, \quad v_\theta^* = A_1 r - 5A_2 r^3, \quad v_\varphi^* = -v_\theta^*, \\
v_r^+ = Er - \frac{2B_3}{r^4} + 3\frac{B_4}{r^2}, \quad v_\theta^+ = Er - \frac{2B_3}{r^4}, \quad v_\varphi^+ = -v_\theta^+,
\end{gathered} \tag{7}$$

where B_3 and B_4 are the same coefficients A_3 and A_4.

In accordance with (4), conditions of continuity of the quantities $\sigma_{r\theta}$, $\sigma_{r\varphi}$, v_r, v_θ, ν, must be satisfied at the phase boundary. It can be shown that conditions of continuity of $\sigma_{r\varphi}$, v_φ, as well as $\sigma_{r\theta}$, v_θ lead to the same equation at $r = a$. In accordance with (3), the normal stress meanwhile undergoes a discontinuity connected with the presence of surface tension. The stresses σ_{rr} and $\sigma_{r\theta}$ inside and outside the particle are determined on the basis of expressions for the stress tensor in spherical coordinates [11]. The surface pressure, figuring in (3), can be determined through the radial velocity of the fluid on the surface of the sphere in Fourier transforms. This can be done by means of the formula [11]

$$P_{\alpha} = -\frac{\alpha}{i\omega}\left[2v_r + \frac{1}{\sin^2\theta}\frac{\partial^2 v_r}{\partial\varphi^2} + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial v_r}{\partial\theta}\right)\right], \tag{8}$$

where $i\omega$ ($i = \sqrt{-1}$) in the Fourier transforms replaces the operator $\partial/\partial t$.

With allowance for (7), the conditions indicated above in (4) lead to a system of equations relative to the unknown coefficients A_1, A_2, B_3, B_4. We will also write these equations in Fourier transforms:

$$\begin{gathered} A_1 - 3A_2 + 3B_3 - 3B_4 = E,\ A_1 - 5A_2 + 2B_3 = E, \\ A_1\eta_1 - 8\eta_1 A_2 - 8B_3\eta - 3\eta B_4 = \eta E,\ A_1\eta_1 + \frac{3}{2}\eta_1 A_2 + \\ + \left(12\eta - \frac{6\alpha}{ai\omega}\right)B_3 + 3\left(3\eta - \frac{2\alpha}{ai\omega}\right)B_4 = \left(\eta + \frac{2\alpha}{ai\omega}\right)E. \end{gathered} \tag{9}$$

In terms of significance, the integral (2) for the determination of η coincides with the integral obtained using the energy-based criterion of the equivalence of homogeneous and heterogeneous media [10]. Here, the Eshelby formula [10] leads to an expression which we can use to determine effective viscosity

$$\eta = \eta_0 + \frac{3\rho}{4\pi a E^2}\int_0^{2\pi}\int_0^{\pi}[-\sigma_{rr}^0 v_r^+ - \sigma_{r\theta}^0 v_\theta^+ - \sigma_{r\varphi}^0 v_\varphi^+ + \sigma_{rr}^+ v_r^0 + \sigma_{r\theta}^+ v_\theta^0 + \sigma_{r\varphi}^+ v_\varphi^0]\sin\theta d\theta d\varphi, \tag{10}$$

where the variables having the superscript 0 pertain to the case of a homogeneous medium consisting entirely of the dispersion liquid with the viscosity η_0:

$$\begin{gathered} v_r^0 = Er\sin^2\theta\cos 2\varphi,\quad v_\theta^0 = Er\sin\theta\cos\theta\cos 2\varphi, \\ v_\varphi^0 = -Er\sin\theta\sin 2\varphi,\quad \sigma_{rr}^0 = 2\eta_0 E\sin^2\theta\cos 2\varphi, \\ \sigma_{r\theta}^0 = 2\eta_0 E\sin\theta\cos\theta\cos 2\varphi,\quad \sigma_{r\varphi}^0 = -2\eta_0 E\sin\theta\sin 2\varphi. \end{gathered}$$

Having determined A_1, A_2, B_3, and B_4 from system (9), we find the corresponding components of stress and velocity. Using these components in the integral (10) leads to the following equation in the Fourier components to determine the effective viscosity of the emulsion:

$$\begin{gathered} \Omega = \Omega_0 + \frac{15}{2}\frac{\Omega(1-\Omega_0)}{1-\Omega}\rho[(1-\Omega)(9{,}5+8\Omega)i\omega + \\ + 2\Lambda_1(2\Omega+5)]/[(4{,}5\Omega+3)(9{,}5+8\Omega)i\omega + 30\Lambda_1(1+\Omega)], \\ \Omega = \eta/\eta_1,\ \Omega_0 = \eta_0/\eta_1,\ \Lambda_1 = \frac{\alpha}{a\eta_1}. \end{gathered} \tag{11}$$

In the absence of surface tension ($\alpha = 0$), the formula is simplified:

$$\Omega = \Omega_0 + \frac{15}{2}\frac{(1-\Omega_0)\Omega}{3+4{,}5\Omega}\rho, \tag{12}$$

while at $\delta << 1$, where $\delta = \omega/\Lambda_1 = \omega a \eta_1/\alpha$, we find from (11) that

$$\Omega = \Omega_0 + \frac{1}{2}\,\frac{\Omega(1-\Omega_0)}{1-\Omega}(5+2\Omega)\,\rho/(1+\Omega). \tag{13}$$

We can use (11) to determine Ω with different values of δ and ρ, while the limiting cases $\delta >> 1$ and $\delta << 1$ are studied by means of equations (12) and (13), respectively.

In the case of dilute systems, we use a scheme in which the sample drop is assumed to be surrounded by liquid whose viscosity coincides with the viscosity of the dispersion medium η_0. Here, by repeating the above calculations we obtain a formula to determine the relative effective viscosity of the emulsion in the form

$$\Omega = \Omega_0\left[1 + 5\rho\,\frac{(1-\Omega_0)(9{,}5+8\Omega_0)\,i\omega + 2\Lambda_1(2\Omega_0+5)}{(3\Omega_0+2)(9{,}5+8\Omega_0)\,i\omega + 20\Lambda_1(1+\Omega_0)}\right]. \tag{14}$$

From (14) for $\delta >> 1$ we have

$$\Omega = \Omega_0\,[1 + 5(1-\Omega_0)\,\rho/(3\Omega_0+2)], \tag{15}$$

which coincides in form with the formula in [2, 10] for the effective shear modulus of an elastic medium with spherical elastic inclusions if we take Ω and Ω_0 to mean the relative effective shear modulus μ/μ_1 and the relative shear modulus μ_0/μ_1 of the matrix, respectively.

At $\delta << 1$, it follows from (15) that

$$\Omega = \Omega_0\left[1 + \frac{1}{2}(2\Omega_0+5)\,\rho/(1+\Omega_0)\right], \tag{16}$$

which coincides with the Taylor formula for the effective viscosity of emulsions.

Considering (14) and (1) and using the inverse Fourier transform, we arrive at the following rheological equation of state for the emulsion:

$$\left(1 + \lambda_1^{\sigma}\frac{\partial}{\partial t}\right)\sigma = 2\tilde{\eta}\left(1 + \lambda_1^{e}\frac{\partial}{\partial t}\right)\mathbf{e}, \tag{17}$$

where

$$\lambda_1^{\sigma} = \frac{(3\Omega_0+2)(9{,}5+8\Omega_0)}{20\Lambda_1(1+\Omega_0)},\quad \lambda_1^{e} = \frac{[3\Omega_0+2+5(1-\Omega_0)\rho](9{,}5+8\Omega_0)}{10\Lambda_1[2(1+\Omega_0)+(2\Omega_0+5)\rho]}, \tag{18}$$

$$\tilde{\eta} = \eta_0\left[1 + \frac{1}{2}(2\Omega_0+5)\,\rho/(1+\Omega_0)\right].$$

It can be seen from (17) that when allowance is made for surface tension at the phase boundary of the emulsion, the rheological equation is of a relaxational character. For high-frequency nonsteady processes or small Λ_1, i.e., at $\delta >> 1$, relaxation (17) is replaced by the the relation $\sigma = 2\eta e$, where η is determined as

$$\eta = \eta_0\,[1 + 5\rho(1-\Omega_0)/(3\Omega_0+2)]. \tag{19}$$

In the case of low-frequency processes ($\delta << 1$), rheological equation (17) also changes into the standard steady-state rheological equation, with an effecive viscosity $\tilde{\eta}$, calculated from (18).

The ranges of δ for different systems depends on the characteristics of the liquid drop and the value of frequency ω. For example, for high-viscosity oils with $\eta_1 = 10$ Pa·sec and $\alpha = 25$ mN/m, when the radius of a liquid drop $a = 10^{-4}$ m, we have $\delta = 4 \cdot 10^{-3}\,\omega$. It is evident from this that at $\omega \leq 10^2$ sec^{-1} we can assume that $\delta \ll 1$. In the region of frequencies of tens of kilohertz and higher, $\delta \gg 1$.

In the more general case, we can examine situations in which one of the phases (or both phases) have viscoelastic properties. Such an analysis was performed in [12, 13] for certain types of emulsions and suspensions. Here, using the principle of elastic-viscoelastic analogy, we can immediately write expressions for effective viscosity. For example, let the disperse phase have viscoelastic properties. In the Fourier transforms in this case, we need to replace η_1 by $\eta_1(1+\varepsilon_1^e i\omega)/(1+\varepsilon_1^\sigma i\omega)$. This quantity corresponds to the Oldroyd viscoelastic model [1], where ε_1^σ and ε_1^e are the relaxation and retardation times, respectively. Then, in accordance with (14), we obtain an expression for the effective viscosity of the emulsion in Fourier transforms which is a rational fractional function relative to $i\omega$ (due to the limited scope of this article, the specific form of this expression is omitted here). Performing the inverse Fourier transformation with allowance for (1), we obtain a rheological equation of the form (17) which contains derivatives up to the third order with respect to time relative to σ and e and, accordingly, new characteristic scales of time — relaxation times. The appearance of higher derivatives is due to the relaxation times ε_1^σ and ε_1^e. When they vanish, the additional relaxation times also vanish, i.e., the rheological equation becomes (17).

Using the principle of elasto-viscoelastic analogy, we can also proceed on the basis of (11) to study mixtures with phases having more complex rheological properties. Here, η_0 or η_1 (or η_0 and η_1) should be replaced by $\eta_0(i\omega)$ or $\eta_1(i\omega)$. Then, using (11), we can perform numerical calculations to establish the relation $\eta = \eta(i\omega)$. In (1), changing over from the Fourier transforms to the originals, we obtain the rheological equation of the mixture. With approximation of the solution of (11) Ω in the form of a polynomial relative to $i\omega$ or a ration of such polynomials, by changing over to the originals we obtain a linear relaxation equation of the form

$$P\boldsymbol{\sigma} = 2\tilde{\eta}Q\mathbf{e}, \tag{20}$$

where $\tilde{\eta}$ is a certin coefficient which can be represented in the form (18), (19), etc. The quantities P and Q are differential operators of the form

$$P = 1 + \lambda_1^\sigma \frac{\partial}{\partial t} + (\lambda_2^\sigma)^2 \frac{\partial^2}{\partial t^2} + \ldots,\ Q = 1 + \lambda_1^e \frac{\partial}{\partial t} + (\lambda_2^e)^2 \frac{\partial^2}{\partial t^2} + \ldots$$

Numerical calculations of the effective properties for moderately concentrated and dilute emulsions were performed in accordance with Eqs. (11) and (14). Figure 1 shows the change in the real part of Ω for two values of the parameter δ. At small δ, the difference in the change in Re Ω with respect to ρ for dilute and moderately concentrated systems is expressed more sharply compared to the case of large values of δ. In each case, Re Ω is larger for moderately concentrated systems than for dilute systems. In other words, large values of drop surface tension lead to an increase in the difference between the effective viscosities of dilute and concentrated emulsions. This difference is larger, the larger the concentration of drops of the emulsion ρ.

Figure 2 shows the dependence of the real and imaginary parts of Ω for moderately concentrated emulsions on ln δ. It can be seen from the figure that an increase in δ is accompanied by a monotonic decrease in Re Ω. The quantity Im Ω is characterized by convex dynamics. This is analogous to the result obtained previously for dilute emulsions: at small and large ω, the relaxation process can be ignored. Intermediate values of δ cause the rheological model to be of a relaxational nature, i.e., the effect of Im Ω is substantial in this case. The character of the change in Re Ω and Im Ω with respect to δ for dilute emulsions is generally similar to that for moderately concentrated emulsions. However, the values of Re Ω and Im Ω for dilute emulsions are considerably smaller than the corresponding values for moderately concentrated systems. For dilute systems with small δ, the values of Re Ω coincide with the values of Re Ω determined from the Taylor formula. At large δ, they coincide with the values obtained from Eq. (15). For moderately concentrated systems, these limiting values are also greater than the corresponding limiting values of Re Ω for dilute systems.

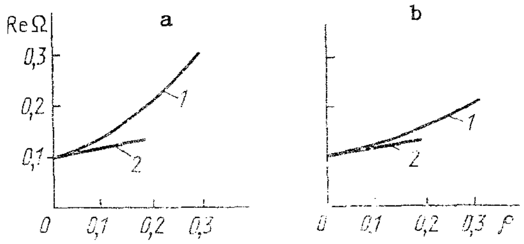

Fig. 1. Dependence of Re Ω on ρ for moderately concentrated (1) and dilute (2) emulsions: $\delta = 10^{-2}$ (a); 10^2 (b).

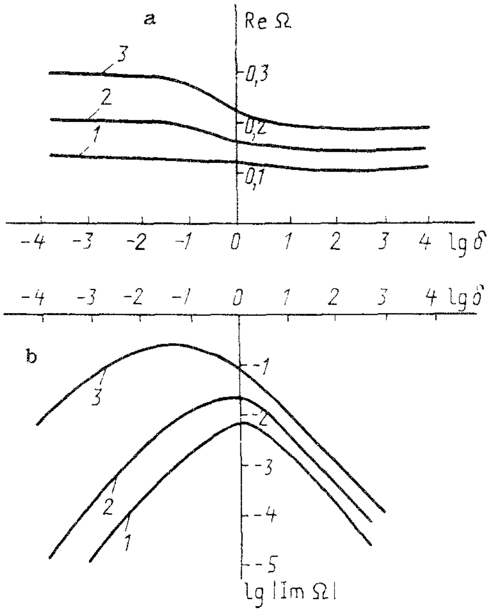

Fig. 2. Dependence of the real part (a) of Ω and the modulus of the imaginary part of Ω (b) on ln δ in moderately concentrated emulsions at ρ = 0.1 (1); 0.2 (2); 0.3 (3).

NOTATION

a, radius of spherical drop; e, strain-rate tensor; n, normal unit vector on the surface of the sphere; p, pressure; P_α, surface pressure; r, position-vector; v, velocity vector; v_n, v_t, normal and tangential components of velocity; v_r, v_θ, v_φ, coordinates of velocity in the spherical coordinate system; α, surface tension at the phase boundary; $\delta = \omega a \eta_1/\alpha$, parameter; ε_1^σ, σ_1^e, relaxation times for stress and the velocity gradient of a viscoelastic drop; η_0, η_1, viscosity of the dispersion medium and disperse phase of the emulsion, respectively; η, effective viscosity of the emulsion; λ_1^σ, λ_2^e, λ_2^σ, λ_2^e, relaxation times; δ, volume concentration of the disperse phase in the emulsion; σ, stress tensor; σ_n, σ_t, normal and tangential components of stress; ω, Fourier transform parameter. The superscript * denotes the region inside a sphere of radius a, while the superscript + denotes the region outside this sphere.

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EXPERIMENTAL STUDY OF HEAT TRANSFER FROM THE WALLS OF A CHANNEL TO A CIRCULATING FLUIDIZED BED

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Results are presented from an experimental study of external heat transfer in a circulating fluidized bed.

Due to the intensive mixing of the dispersed material and the gas and the possibility of controlling their time of contact within a broad range of values, it has been found expedient to use circulating fluidized beds in a number of chemical processes and operations involving drying and combustion. The broader use of such systems is being held up by a lack of reliable data on their aerodynamics and heat transfer characteristics.

An experimental study of external heat transfer in a circulating bed was conducted on a unit at the S, M. Kirov Ural Polytechnic Institute [1]. The unit was closed with regard to the dispersed material and open with regard to the gas. The main component of the unit was a channel made of a steel tube with an inside diameter of 250 mm. The tube was composed of 10 sections placed on top of one another. The length of each section was 950 mm. Transparent windows for visual observation were located in each section. The lowest section of the channel had a gas-distributing grate with a through section of 10%. The grate functioned as an aerodynamic chamber in the given set-up. The finely dispersed material was quartz sand with a mean-mass particle diameter of 287 μm. The fractional composition of the sand is shown in Table 1.

The bulk and true densities of the sand were 1249 and 2640 kg/m^3. In amounts of up to 12.1 tons/h (which corresponded to a change of up to 70 kg/(m^2·sec) in the unit load of material per 1 m^2 of channel cross section), the solid phase was introduced into the unit with the use of a cantilever-type screw feeder with an inside diameter of 144 mm. The feeder was positioned 175 mm above the grate. The delivery of air into the feeder kept it from becoming obstructed in the case of high sand feeds. The bed was fluidized with air having a temperature of 30-60°C. The air was directed through the grate with a filtration velocity w ranging from 5 to 10 m/sec (calculated on the basis of the empty cross section of the channel). Here, the theoretical eddy velocity of the particles was 3.4 m/sec for particles 0.4 mm in diameter and 6 m/sec for particles 0.8 mm in diameter. At the outlet of the channel, the disperse flow was separated in a two-stage dust separator consisting of cyclones and a bag filter. Passing through a system of hoppers and shut-off valves, the particles returned to the air chamber, while the air was released into the atmosphere.

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