BREAKUP OF DROPLETS IN TURBULENT SHEAR FLOW OF DILUTE LIQUID–LIQUID DISPERSIONS

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The investigation of the simultaneous motion of immiscible liquids in pipes and in various kinds of industrial apparatus is very timely in the practical aspect of the problem, because often it is not so much the actual values of the relevant hydrodynamic parameters that are important as what they ultimately govern, namely the most practical operating conditions and the efficiency of a large class of technological processes involved in various branches of industry. The analytical approach to such complex flows is difficult, making it necessary to resort to experiment and similarity methods and to analyze probability situations involving disperse-phase flow elements that vary in space and time [1]. In particular, the implementation of continuous mass transfer, extraction, and other chemical processes in liquid–liquid systems is based on frequently repeated events of aggregation and breakup of droplets of the disperse phase and depends on the configuration and nature of the two-phase flow process [2]. The semiempirical approach to the calculation of the coefficient of fluid friction of dilute emulsions also requires a priori information about the droplet size in the disperse phase [3].

The breakup of droplets of the disperse phase in the motion of liquid–liquid dispersions in turbulent pipe flow takes place under the dynamic and shear stresses of the continuous phase. According to the theory developed by Kolmogorov [4] and Hinze [5], the maximum diameter of breakup-stable droplets is related to the average flow velocity by the expression $d_* \sim U^{-1.2}$. To date, however, the interpretation of the experimental data on the basis of the Kolmogorov–Hinze model has failed to provide a reliable dependence of the droplet sizes on the conditions of pipe flow of dispersions with different physicochemical properties [6, 7]. For example, a different law, $d_* \sim U^{-2.5}$, which departs significantly from the conventionally accepted form, has been established, initially on the basis of experimental data in [8] and later in [9, 10].

In this study we attempt to generalize Kolmogorov's theory in such a way as to refine existing notions of the complex processes involved in the breakup of droplets in nonuniform turbulent flow of a mutually immiscible liquid, on the basis of published empirical material, and to eradicate the conflicts between the theoretical conjectures and the experimentally observed laws. The latter objective is essential to the tenable selection of mathematical models of droplet breakup for use in numerical and analytical studies of disperse systems [11, 12].

1. Droplet Breakup in Uniform Turbulent Flow

Despire the complexity and statistical nature of the process of breakup of droplets by turbulent flow of a mutually immiscible liquid, the hypothesis that the local structure of the flow is the principal factor affecting the maximum diameter of breakup-stable droplets [4] has been extremely fruitful. On the basis of dimensional-analytic considerations, this means that droplets larger than the turbulence microscale λ_0 are predominantly acted upon by inertial forces. Otherwise breakup is instigated by viscous shear stresses. Assuming that droplet stability is associated primarily with interphase tension, we write the fundamental relations in the form

$$\rho_c\overline{v^2} \approx \sigma/d_*, \qquad d_* > \lambda_0; \tag{1.1}$$

$$\mu_c \partial v/\partial r \approx \sigma/d_*, \quad d_* < \lambda_0, \tag{1.2}$$

where ρ_c and μ_c are the density and dynamic viscosity of the continuous phase, σ is the interphase tension, and $\overline{v^2}$, $\partial v/\partial r$ are the mean-square value and gradient of the eddy velocities.

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In the inertial interval of the domain condition (1.2) holds only for a very high rate of mixing in a special apparatus and will not be considered here.

In the inertial interval of the domain of universal static equilibrium, i.e., in the interval specified by the relation $\lambda_0 < d_* < L$, where L is the turbulence macroscale characterizing the upper limit of the spectrum of eddy diameters, we have the relation $\overline{v^2} = 2.0(\varepsilon d_*)^{2/3}$ [4]. The validity of this relation is supported by experimental work [13]. Here ε denotes the energy dissipation rate per unit mass of liquid. Relation (1.1) is therefore reducible to the familiar form

$$d_* (\rho_c/\sigma)^{3/5}\varepsilon^{2/5} = c, \tag{1.3}$$

where c is a constant to be determined experimentally.

It must be noted, however, that the validity of expression (1.3) has been fully verified only for the conditions of turbulent mixing by propeller-type and vane-wheel turbines in mixing vessels with deflectors [14]. The high turbulence intensity of the flow, in some cases 50-60% [15, 16], ensures the breakup of droplets with sizes up to the inertial interval of the spectrum of eddy velocities, where the contribution of viscous forces is insignificant.

2. Nonuniform Pipe Flow

The formal application of relation (1.3) to the breakup of droplets by turbulent pipe flow ignores some of its essential features. For example, while it is generally known that turbulent flow is locally isotropic over a large part of the pipe cross section, the turbulence intensity is 3-4% [13]. Moreover, the logarithmic profile of the average velocity in the wall zone is dictated not only by the dynamic forces, but also by the action of high shear stresses on the droplets.

Under pipe-flow conditions the energy dissipation rate per unit mass of liquid is expressed by the balance relation $\pi D^2 l \rho_c \varepsilon/4 = \tau_w \pi D l U$, in which the frictional stress at the wall $\tau_w = \lambda\rho_c U/8$; λ is the coefficient of fluid friction, calculated according to the Blasius formula; D and l are the diameter and length of the pipe; and U is the flow velocity averaged over the mass flow rate. The final relation has the form

$$\varepsilon = \lambda U^3/2D.$$

Substituting the expression for ε and making certain transformations in accordance with [6], we can express relation (1.3) in terms of two dimensionless groups of parameters:

$$\lambda\,\mathrm{We} = 0.93\,(\sqrt{\lambda}D/d_*)^{2/3}, \tag{2.1}$$

where the dimensionless Weber number $\mathrm{We} = \rho_c d_* U^2/\sigma$ and the constant of proportionality is evaluated for c = 0.725 in accordance with the recommendations of [5].

Data from experimental studies [6-9] are shown in Fig. 1 in the dimensionless coordinates $\lambda\mathrm{We}$ and $\sqrt{\lambda}D/d_*$, which follow from the Kolmogorov–Hinze breakup model in the form (2.1) applied to the conditions of turbulent pipe flow of a dilute liquid–liquid dispersion. This relation has been found to correspond well to the data of [6] (points 14) and to some of the data of [7] (points 2). Line II represents the end results of processing the data of [7] for two disperse systems: kerosene and transformer oil with water (points 1 and 2) on the basis of relation (1.3): $d_{95}/D = 4.0(\rho_c DU^2/\sigma)^{-3/5}$. After transformation to the customary variables we have

$$\lambda\mathrm{We} = 10.08\lambda^{2/3}(\sqrt{\lambda}D/d_{95})^{2/3}.$$

Here the constant of proportionality is equal to 0.743 and 1.18 for $\lambda = 0.02$ and 0.04, respectively; these values practically coincide with relation (2.1), which is represented by line I in Fig. 1.

However, the second part of the data of [7] (points 1), along with the data of [9] (points 3-5) and [8] (points 6-13), correlates with lines having an altogether different slope: –0.03 (in [6] the line is erroneously written in the form $\lambda\mathrm{We} = 5.52$):

$$\lambda\,\mathrm{We} = c_0\,(\sqrt{\lambda}D/d_*)^{-0.3},$$

where c_0 is a dimensionless function of the viscosities of the phases in the disperse system. It can be verified that the relations between the fundamental parameters, namely the

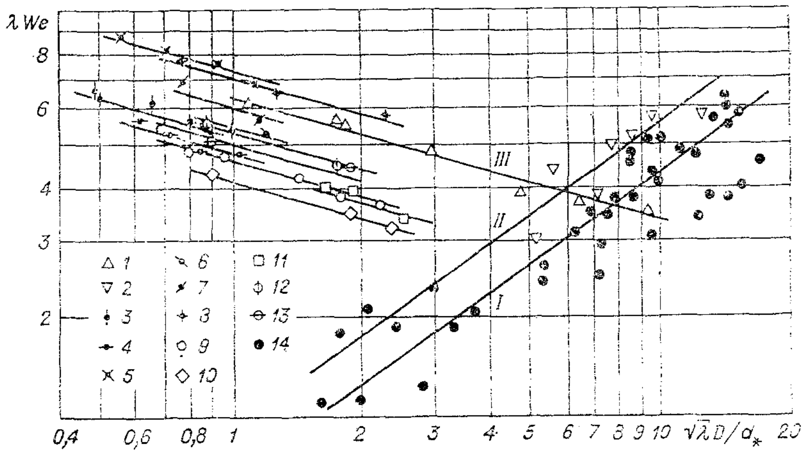

Fig. 1

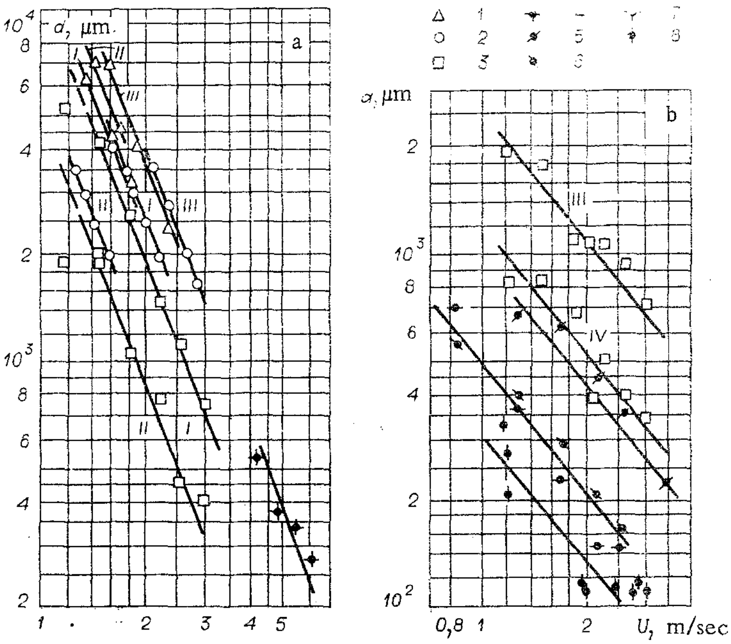

Fig. 2

average flow velocity, the interphase tension, the pipe diameter, and the maximum diameter of stable droplets, practically coincide with the empirical correlation postulated in [8]:

$$\mathrm{We}(\mu_c U/\sigma)^{0.5} = c_1[1 + 0.7(\mu_d U/\sigma)^{0.7}],$$

where μ_d is the viscosity of the disperse phase. Processing of the experimental results for D = 3.81 cm yields the constant c_1 = 38 [8]. Repetition of the experiments for D = 1.27 cm resulted in the constant c_1 = 43, which confirms the relation $d_* \sim D^{0.1}$ [9].

The fact that the doubtful [6, 7] relation $d_* \sim U^{-2.5}$ is not a consequence of erroneous processing of the experimental results [17] is illustrated by data from several studies, shown in Fig. 2. Points 1 are taken from [8], points 2 from [4], 3 from [7], 4 from [11], 5-7 from [6], and 8 from [18]. Although the procedures used to measure the maximum diameter of stable droplets differed, this fact does not affect the general character of the functional relationship as long as the disperse phase has the same kind of droplet-size distribution function. The volume/surface diameter d_{32}, the diameter d_{95} of the droplets comprising 95% of the volume of the disperse phase, and other such quantities reflect the distribution function in generalized form and differ only by the proportionality constants. This conjecture is corroborated by lines I and III, plotted for the maximum diameter d_*, and by lines II and IV for d_{32} [7] (the relationship of these diameters is expressed as $d_* = 2.1 d_{32}$).

The points from [8], approximated by lines I-III, correspond to disperse-phase viscosities of 7.2, 14.1, and 23.1 mPa·sec and a continuous-phase viscosity of 0.97 mPa·sec. Lines I-III also identify points corresponding to the dispersion in water of droplets with viscosities of 0.62, 0.88, and 28.1 mPa·sec [9], while lines I, II and III, IV represent dispersions of water in kerosene and of water in transformer oil with viscosities of the organic phase equal to 1.82 and 16-18 mPa·sec. It is important to note that the data of Fig. 2a correspond to the correlation equation (2.2), and the data of Fig. 2b to (2.1).

An attempt to make a generalization, analogous to that in Fig. 1, of the experimental data of the above-cited papers in terms of the dimensionless groups given in [8] $We(\mu_c U/\sigma)$ and $\mu_d U/\sigma$ proved unsuccessful. This is evidence of the universality of Kolmogorov's droplet breakup theory [4]. However, although the nature of the breakup process may correspond to relation (2.1) in special cases, it is obvious from an examination of the data in Figs. 1 and 2 that the extent of interaction of nonuniform turbulent flow with disperse-phase droplets is far greater than is allowable within the bounds of its local structure.

3. Mechanism of Droplet Breakup in Nonuniform Turbulent Flow

A relation for the maximum diameter of stable droplets in uniform locally isotropic turbulent flow can be derived theoretically on the basis of an analysis of capillary waves on the surface of a liquid sphere [17]. When the characteristic frequency of the turbulent fluctuations coincides with the natural frequency of the sphere, the droplet becomes unstable. The natural frequencies f_n of a droplet are given by the formula [19]

$$(2\pi f_n)^2 = \frac{8(n-1)n(n+1)(n+2)\sigma}{[(n+1)\rho_d + n\rho_c]\, d_*^3}.$$

The oscillatory motions of the sphere correspond to frequencies beginning with n = 2. Making use of the fact that the turbulent fluctuation frequency $f^0 = \sqrt{\overline{v^2}}/d_*$, we deduce the following from the condition $f_2 = f^0$ for liquid–liquid dispersions with close densities of the phases $\rho_d \approx \rho_c$:

$$\rho_c d_* \overline{v^2}/\sigma = c_2. \tag{3.1}$$

In the inertial interval of the domain of universal static equilibrium [4] relation (3.1) is analogous in form to the well-known expressions (1.3) and (2.1), evincing the adequacy of the model in [17] for the breakup mechanism.

It has been determined by visual observations, however, that in the course of breakup of droplets in pipe flow they are considerably deformed [8, 9, 11]. Under the action of the gradient of the average velocity a sufficiently large drop is elongated and, in the wall zone, acquires a form resembling an ellipsoid of revolution. According to Taylor's theory, the deformation F = (A + B)/(A − B), where A and B are the major and minor axes of the ellipsoid, is related to the physical properties and dispersion flow conditions as follows [19]:

$$F = \frac{Gd\mu_c}{2\sigma}\left(\frac{19\mu_d/\mu_c + 16}{16\mu_d/\mu_c + 16}\right). \tag{3.2}$$

Next, it is reasonable to assume that the diameter of the deformed droplets must be represented in expression (3.1) by an effective value taking into account the increase in the radius of curvature of the surface of the original sphere upon transition to an elliptical shape. To adjust for the change in the droplet diameter we introduce $d_e = d_* f(F)$, where f is a function of the deformation. The average-velocity gradient in expression (3.2) can be written as follows with regard for the logarithmic distribution function in a hydraulically smooth pipe:

$$G = \frac{u(y+d) - u(y)}{d} = \frac{u_*}{\varkappa d}\left[\ln\frac{(y+d)\nu_c}{u_*} - \ln\frac{y\nu_c}{u_*}\right].$$

Assuming that the droplet is located close to the pipe wall, that y ≈ d, and that the dynamic velocity $u_* = \sqrt{\lambda/8}U$, we obtain (correct to within a constant) G = U/d. Thus, the deformation expression (3.2) acquires the form

$$F = \frac{\mu_c U}{\sigma} f\left(\frac{\mu_d}{\mu_c}\right),$$

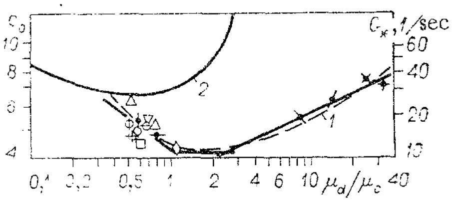

Fig. 3

where the function $f(\mu_d/\mu_c)$ under the conditions of the wall zone of turbulent flow differs from the expression in parentheses in (3.2) and is evaluated according to experimental data.

The form of the function f(F) can be determined on the basis of the following considerations. Deformations by the average-velocity gradient are experienced by large drops having a diameter commensurate with the inertial interval. The upper bound is determined by the turbulence macroscale $L = 0.0074DRe^{0.125}$ [20], where the Reynolds number $Re = UD\rho_c/\mu_c$. Such droplets are acted upon only by the largest eddies, so that under turbulent pipe flow with $d_* \approx L$ it must be assumed that $\bar{v}^2 = u_*^2 \approx \lambda U^2/8$ [20]. Finally, bearing in mind the empirical relation for the investigated conditions $d_* \sim U^{-2.5}\sigma^{1.5}$ [8, 9], on the basis of expression (3.1) we obtain $f(F) \equiv \sqrt{F}$.

We have thus shown that the empirical relation (2.2) describes the resonant oscillations of a deformed sphere with an effective diameter $d_e = d_*\sqrt{F}$ and can be represented by the equation $\rho_c d_e \bar{v}^2/\sigma = c_3$, which has the same form as expressions (3.1). This means that the Kolmogorov–Hinze model, extended to the breakup of deformable droplets, confirms the reliability of the correlation equation (2.2) and can be used to refine the mechanism of their breakup by nonuniform turbulent flow.

4. Influence of Viscosities of the Phases of the Dispersion on the Stability of Deformable Droplets

According to [5], with an increase in the viscosity of the disperse phase the oscillations of a droplet are suppressed, and for sufficiently small values of μ_d the viscosity of the disperse phase does not affect the breakup process. The correlation introduced in [8] reflects this viscosity dependence by the empirical term $1 + 0.7(\mu_d U/\sigma)^{0.7}$, which presumes a constant growth of the stable droplet size with μ_d. However, there are data (cf. [9]) that do not conform to the nature of this dependence (curves I-III in Fig. 2a).

The indicated contradiction can be explained and the form of the functional relation for the viscosity ratio in (2.2) can be sensibly postulated by taking into account the above-determined viscous behavior of the breakup of deformable droplets. Experimental studies of the behavior of droplets in a simple shear field [21] have shown that the breaking value of the velocity gradient has a minimum for a viscosity ratio μ_d/μ_c in the interval from 0.2 to 1.0, in which the droplets break up most easily. Also, irrespective of the type of liquid–liquid system, if the condition $\mu_d/\mu_c > 4$ or $\mu_d/\mu_c < 0.005$ holds, breakup will not occur. In this case the values of the breaking velocity gradient tend asymptotically to infinity.

Figure 3 shows the values of c_0 in relation (2.2) for various ratios μ_d/μ_c (curve 1) according to the data of [7-9]. Curve 2 represents the analogous dependence for the breaking velocity gradient G_* necessary in order to break up droplets in a simple shear field [21]. Although curves 1 and 2 are shifted apart because of the influence of turbulent fluctuations and the departure of the flow in the wall zone of the pipe from simple shear, the similarity of their behavior is further evidence in support of the breakup mechanism adopted in this study. A shortage of experimental material prevents us from determining the analytical form of curve 1 in Fig. 3. For practical calculations the following approximation is proposed:

$$c_0 = 4.27(\mu_d/\mu_c)^{-0.38} \quad \text{for} \quad \mu_d/\mu_c < 1.05; \tag{4.1}$$

$$c_0 = 4.2 \quad \text{for} \quad 1.05 \leqslant \mu_d/\mu_c \leqslant 2.4; \tag{4.2}$$

$$c_0 = 3.45(\mu_d/\mu_c)^{0.22} \quad \text{for} \quad \mu_d/\mu_c > 2.4. \tag{4.3}$$

The validity of expressions (4.1)-(4.3) must be restricted to the domain of variation of the phase velocities in experiments: 0.96 mPa·sec < μ_c < 1.8 mPa·sec; 0.5 mPa·sec < μ_d < 32.1 mPa·sec.

It follows from a comparison of curves 1 and 2 in Fig. 3 that the viscous characteristics of the breakup process can be sensibly included by replacing the parameter $\mu_d U/\sigma$ with μ_d/μ_c for the estimation of c_0 in relation (2.2). This operation also eliminates the scatter of the experimental points for small values of μ_d, which in Fig. 8 of [8] and in Fig. 2 of [9] are clustered about the ordinate axis and in no way reflect the empirical relation (4.1) obtained on the basis of processing of the data of the cited works in the coordinates We and $\sqrt{\lambda}D/d_*$ (see Fig. 1).

With a reduction in the deformability of the droplets the empirical curve of the type of curve 1 in Fig. 3 will doubtless change to the form proposed in [5]: $1 + f(\mu_d U/\sigma)$. The role of the viscous forces and, hence, of μ_c in the inertial interval is small in comparison with the dynamic fluctuation forces. In this case the relation for the maximum diameter of stable droplets in the form (2.2) will go over to the form (2.1). The description of the transient process represents the problem of the resonant oscillations of a liquid droplet subjected to deformation in a shear field; the solution of this problem requires separate treatment.

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