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STATISTICAL DISPERSION MODEL TO DESCRIBE THE PROCESSES IN A HOMOGENIZER OF THE "LIQUID–LIQUID"-TYPE

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A physicomathematical model of dispersion and homogenization in a liquid–liquid medium which is based on the system of equations for the probability density of the size of disperse particles has been developed. The proposed model takes into account the processes of turbulent atomization and cavitation reduction in size and the process of coalescence of the dispersed-phase drops.

The results of theoretical investigation into the processes of dispersion and homogenization of liquid–liquid systems enjoy wide practical application in various industries (milk, paint and varnish, chemical, and cosmetic), since emulsions treated in such a way are more slowly segregated and retain their macroscopic homogeneity for a longer time. Study of the dispersion of these systems is a complex practical and theoretical problem because of the need to take into account the competing processes of atomization and coalescence of disperse components.

Analyzing the available hypotheses and theories [1–4], one can single out the following basic features of the process of homogenization. First, the dispersion of drops is practicable only in the case where they are in a readily deformable liquid state. Second, by means of deformation, the drops must be brought into a thermodynamically unstable state where the process of spontaneous atomization can occur under the action of surface forces which tend to decrease the phase interface. Third, considerable deformation of the drops is possible only for rather high local gradients of velocity or in the presence of cavitation impacts in the medium. Cavitation atomization occurs because of the formation and collapse of gasdynamic nuclei immediately inside the drop on a sharp local decrease and then recovery of the pressure in the flow. With low volume concentrations of dispersion, cavitation impacts mainly destabilize the interface from the outside and are the factor which accompanies turbulence.

An important component of the process of homogenization in liquid–liquid systems is the process of coalescence, which depends on the diffusion rate of disperse particles in the medium and on the structural-mechanical properties of the interface.

Criterion of Atomization of Drops in a Turbulent Flow. Atomization of drops in a turbulent liquid flow occurs in the case where the intensity of turbulent pulsations of the velocity of a dispersing liquid exceeds a certain critical value, which is different for various drop sizes. In other words, for the turbulent field (of prescribed intensity) of the dispersing liquid there is such a size of the liquid drops in the dispersed liquid that the drops with a radius larger than the critical one can be atomized.

Using the data of [5] as the basis, we write the following expression for the critical radius of the drop:

$$a_{\mathrm{cr}} = L^{2/5}\left(\frac{\sigma}{k_{\mathrm{f}}\,\rho}\right)^{3/5}\frac{1}{\Delta u^{6/5}}\,, \tag{1}$$

where σ is the surface tension coefficient of the dispersed liquid, k_{f} is the coefficient of resistance of the moving drop in the liquid, $k_{\mathrm{f}} = 0.5$, and Δu is the characteristic difference in the pulsation velocities of the medium at a distance of the order of the drop diameter.

A formula for the critical radius of the drop can be written in terms of the function $P_x(r)$, which describes the energy distribution of turbulent velocity pulsations over different scales of length r [6]:

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$$a_{\text{cr}}(x) = \left(\frac{\sigma}{k_{\text{f}}\,\rho}\right)^{3/5} \left(\frac{\int\limits_0^\infty rP_x(r)\,dr}{\int\limits_0^\infty P_x(r)\,dr}\right)^{2/5} \left(2\int\limits_0^{2a_{\text{cr}}(x)} P_x(r)\,dr\right)^{-3/5}, \tag{2}$$

$$\Delta u = \left[2\int\limits_0^{2a_{\text{cr}}} P_x(r)\,dr\right]^{1/2}, \quad L = \int\limits_0^\infty rP_x(r)\,dr \Big/ \int\limits_0^\infty P_x(r)\,dr\,.$$

Equation (2) allows us to calculate $a_{\text{cr}}(x)$ as a function of the longitudinal coordinate for a homogenizer path. Then the atomization condition has the form

$$r > a_{\text{cr}}(x)\,. \tag{3}$$

The function $P_x(r)$ can be defined from the closed equation given in [6].

Criterion of Cavitation Atomization of Drops in the Flow. The efficiency of homogenization in the presence of conditions for cavitation depends on the number of cavitation impacts and on the number of disperse inclusions that pass through the zone of action of these impacts. Cavitation in the turbulent flow can appear because of the turbulent pressure pulsations in the liquid when the negative phase of pulsation pressure is realized. The pressure pulsations $B_{p'p'}(x)$ can be determined in terms of the function that describes the turbulent-energy distribution over different scales of length $P_x(y)$ [7]:

$$B_{p'p'}(x) = 2\rho^2 \int\limits_0^\infty yP_x(y)^2\,dy\,. \tag{4}$$

With account for this formula the cavitation criterion in the liquid flow will have the form

$$B_{p'p'}(x) > \overline{p}^{\,2}(x)\,. \tag{5}$$

Using the Θ function of Heaviside we can supplement the equation for $f_x(r)$ with a source term which provides the appearance of drops at the points of the flow where condition (5) is satisfied. This source will simulate the influence of cavitation on the process of homogenization.

Derivation of the Equation for $f_x(r)$. We write the equation for the probability-distribution function of the radii of the dispersed phase $f_x(r)$ in the case where in the turbulent flow the drops of the disperse liquid are reduced in size as a result of turbulent atomization and cavitation. The process of coalescence will not yet be taken into account.

Let us subdivide the entire spectrum of the drops by size into a macrocomponent and a microcomponent. The fraction of the drops with radii less than r will be called the microcomponent of the disperse flow (Mi), while the fraction with radii larger than r will be called the macrocomponent (Ma)

$$\int\limits_0^r f_x(r)\,dr = \text{Mi}\,, \quad \int\limits_r^\infty f_x(r)\,dr = \text{Ma}\,. \tag{6}$$

Now we write the equality

$$u \frac{\partial}{\partial x} \int_r^\infty f_x (R)\, dR = - W (r, x) \,. \tag{7}$$

Here, the probability flow through the point r in the drop-size space is denoted by the symbol $W(r, x)$. From the form of equality (7) with allowance for the fact that we try to describe only the process of reduction of the drops in size it follows that the function $W(r, x) > 0$.

We assume that the structure of the probability flow $W(r, x)$ is related to the probability density $f_x (r)$ by the formula

$$W (r, x) = \int_r^\infty f_x (R) \frac{\hat{\omega}_i (r, R)}{\tau_i (R)}\, dR \,. \tag{8}$$

Here $\hat{\omega}_i(r, R)$, $i = 1, 2$, denotes, the probability of transfer of the macrocomponent Ma to the microcomponent Mi through the point r in the drop-size space. The functions $\tau_i(R)$, $i = 1, 2$, describe the characteristic time of atomization of the drop with radius R into the drops of size $r < R$.

The time of breaking of a drop because of the interaction with the turbulent pulsations of the main liquid $\tau_1(R)$ can be evaluated by assuming that it is equal to the time in which the liquid, related to the velocity pulsation $\Delta u(2R)$, traverses a distance equal to the diameter of the considered drop:

$$\tau_1 (R) = 2R / \langle \Delta u (2R)^2 \rangle^{1/2} = 2R \Bigg/ \left[\int_0^{2R} P_x (r)\, dr \right]^{1/2} \,. \tag{9}$$

The time of breaking of a bubble related to the cavitation process $\tau_2(R)$ is evaluated in another way. We evaluate the characteristic time of cavitation breaking of the drop by taking it to be equal to the time of collapse of a cavern. The results obtained from the Rayleigh theory contain the following expression [8]:

$$\tau_{\text{col}} (R) = 0.91 R_{\text{cav}} \sqrt{\rho / p_\infty} \,. \tag{10}$$

Assuming that the radius of the cavern is approximately equal to that of the considered drop of the disperse liquid and that $p_\infty = \bar{p} (x)$, we obtain

$$\tau_2 (R) = \tau_{\text{col}} = 0.9 R \sqrt{\rho / \bar{p} (x)} \,. \tag{11}$$

Let us consider the functions $\hat{\omega}_i(r, R)$, $i = 1, 2$, in expression (8). They describe the probability of transfer of the macrocomponent to the microcomponent through the point r in the drop-sizes space as a result of the turbulent atomization (with $i = 1$) and cavitation explosion of the drop (with $i = 2$). The function $\hat{\omega}_i(r, R)$ is represented in the form

$$\hat{\omega}_i (r, R) = \int_0^r \omega_i (\tilde{r}, R)\, d\tilde{r} \,. \tag{12}$$

Here, the functions $\hat{\omega}_i(\tilde{r}, R)$ describe the contribution of the occurrence probability of event $R \to \tilde{r}$ to the total probability of transition from R to the microcomponent [0–r]. Equality (12) implies that the probability of atomization of the drop with radius R into the drops with radius $\tilde{r} < r$ consists of a sum of probabilities of all the transitions $R \to \tilde{r}$, where $\tilde{r} \in$ [0–r].

For the function $\hat{\omega}_1(\tilde{r}, R)$ we obtain the condition

$$\hat{\omega}_1 (R, R) = \Theta \,[R - a_{\text{cr}} (x)] \,, \tag{13}$$

which implies that the probability of turbulent atomization of the drop with radius R into smaller drops on satisfaction of the condition of turbulent atomization (3) is a reliable event. At the same time, if the drops are of $R < a_{\text{cr}}(x)$ in size, no turbulent atomization occurs at this point at all.

We substitute expression (12) with $i = 1$ into formula (8) and rewrite equality (7) in the form

$$u \frac{\partial}{\partial x} \int_r^\infty f_x(R)\, dR = -\int_r^\infty \frac{f_x(R)}{\tau_1(R)} \int_0^r \omega_1(\tilde{r}, R)\, d\tilde{r} dR\,. \tag{14}$$

Differentiation of the left- and right-hand sides of equality (14) with respect to the variable r gives

$$u \frac{\partial f_x(r)}{\partial x} = -\frac{f_x(r)}{\tau_1(r)} \Theta\,[r - a_{\text{cr}}(x)] + \int_r^\infty \frac{f_x(R)}{\tau_1(R)} \omega_1(r, R)\, dR\,. \tag{15}$$

For the cavitation mechanism of atomization there is no limitation on the value of the drop radius, i.e., on satisfaction of criterion (5) a drop of any size can be atomized because of the collapse of the cavitation cavern with an intensity determined by the characteristic time $\tau_2(R)$. Therefore, for the cavitation mechanism of atomization the contribution to the right-hand side of the equation for $f_x(r)$ will have the form

$$u \frac{\partial f_x(r)}{\partial x} = -\frac{f_x(r)}{\tau_2(r)} \int_0^r \omega_2(\tilde{r}, r)\, d\tilde{r} + \int_r^\infty \frac{f_x(R)}{\tau_2(R)} \omega_2(r, R)\, dR\,. \tag{16}$$

Combining the right-hand sides of Eqs. (15) and (16), we obtain

$$u \frac{\partial f_x(r)}{\partial x} = -\frac{f_x(r)}{\tau_1(r)} \Theta\,[r - a_{\text{cr}}(x)] + \int_r^\infty \frac{f_x(R)}{\tau_1(R)} \omega_1(r, R)\, dR - \frac{f_x(r)}{\tau_2(r)} \int_0^r \omega_2(\tilde{r}, r)\, d\tilde{r} + \int_r^\infty \frac{f_x(R)}{\tau_2(R)} \omega_2(r, R)\, dR\,. \tag{17}$$

Let us pass to the problem concerning the structure of the functions $\omega_i(r, R)$. Taking into consideration the qualitative experimental indications that in regimes with small distinctions of the Weber number from its critical value the drop is subdivided mainly into two large fragments [9], we accept the hypothesis that the function $\omega_1(r, R)$ describes the turbulent atomization of drops mainly into two equal portions. Other possibilities are not eliminated but they decrease in conformity with the normal law of probability distribution as the asymmetry of subdivision of the drop increases. The maximum contribution to the rate of growth of the function $f_x(r)$ in the space of the drop radii r occurs as a result of the atomization of the drops with radius R mainly into two equal drops, i.e., with $R = \sqrt[3]{2}r$; therefore,

$$\omega_1(r, R) = \frac{1}{N_1} \exp\left\{-\left(r - R/\sqrt[3]{2}\right)^2 / 2\sigma_{1r}^2\right\}, \tag{18}$$

where $\sigma_{1\text{r}}$ is the variance of the probability distribution that is selected so that at the boundary of the domain of definition by the variable r ($r = R, r = 0$) the function $\omega_1(r, R)$ is small. With allowance for the "rule of three sigmas" we obtain that $\sigma_{1\text{r}} = 0.06R$.

In the case of cavitation atomization of the drop, the function $\omega_2(r, R)$ describes the method of reduction of the dispersed phase in size under the action of cavitation impacts, i.e., because of the collapse of the cavitation cavern and the explosion of the drop of radius R. We accept the hypothesis that with such an event the initial drop is broken into n drops of smaller size. As in the case of reduction in size due to turbulent atomization, we will assume that the asymmetric subdivision of a drop into n drops of smaller radius is also possible but the probability of these events decreases with increase in the asymmetry of atomization of the drop in conformity with the normal law of probability distribution. With consideration of the aforesaid, the form of the function $\omega^2(r, R)$ will take the form

$$\omega_2(r, R) = \frac{1}{N_2} \exp\left\{-\left(r - R/\sqrt[3]{n}\right)^2 / 2\sigma_{2r}^2\right\} . \tag{19}$$

The value of the variance σ_{2r} must be selected to be rather small so that at the boundary of the domain defined by r ($r = 0$, $r = R$) the function $\omega_2(r, R)$ is close to zero. Using the "rule of three sigmas," we obtain $\sigma_{2r} \le 0.06R$.

The normalization factor N_i can be calculated from the condition

$$\int_0^R \omega_i(r, R)\, dr = 1 . \tag{20}$$

Integration with respect to r in Eqs. (18) and (19) leads to the formula

$$N_i = N_i(R, \sigma_{ir}) = \sigma_{ir}\sqrt{\frac{\pi}{2}}\left\{\Phi\left(\frac{R}{\sigma_{ir}}\,\frac{\sqrt[3]{n_i} - 1}{n_i^{5/6}}\right) + \Phi\left(\frac{R}{\sigma_{ir}}\,\frac{1}{n_i^{5/6}}\right)\right\} , \tag{21}$$

where

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp(-t^2)\, dt , \quad n_1 = 2 , \quad n_2 = 8 .$$

Now we write the equation for the function $f_x(r)$ that takes into account only the coalescence of the dispersed-phase drop. As a starting point we use the Smoluchowski–Müller equation for the function $\varphi(v, x)$ describing the volumetric distribution of the particles [10]

$$u\,\frac{\partial\varphi(v, x)}{\partial x} = \frac{1}{2}\int_0^v \beta(v - v_1, v_1)\,\varphi(v - v_1, x)\,\varphi(v_1, x)\, dv_1 - \varphi(v, x)\int_0^v \beta(v, v_1)\,\varphi(v_1, x)\, dv_1 , \tag{22}$$

where the function $\beta(v_1, v_2)$ is the frequency of pair collisions of the particles of volumes v_1 and v_2, respectively. The function $\varphi(v, x)$ is defined in such a way that the integral with respect to the variable v of this function gives the total number of particles N in the disperse system. The function $\Psi(v, x)$ normalized to unity will be defined by the formula

$$\Psi(v, x) = \varphi(v, x)/N(x) . \tag{23}$$

Using Eq. (23), we obtain the equality

$$u\,\frac{\partial\Psi(v, x)}{\partial x} = \frac{u}{N(x)}\,\frac{\partial\varphi(v, x)}{\partial x} - u\,\frac{\varphi(v, x)}{N^2(x)}\,\frac{\partial N(x)}{\partial x} . \tag{24}$$

By expressing the function $\varphi(v, x)$ in terms of $\Psi(v, x)$ using formula (23) and with account for Eq. (22), we write the equation for the function $\Psi(v, x)$:

$$\begin{aligned} u\,\frac{\partial\Psi(v, x)}{\partial x} = N(x)\left\{\frac{1}{2}\int_0^v \beta(v - v_1, v_1)\,\Psi(v - v_1, x)\,\Psi(v_1, x)\, dv_1 \,-\right.& \\ \left. -\,\Psi(v, x)\int_0^v \beta(v, v_1)\,\varphi(v_1, x)\, dv_1\right\} - u\,\frac{\partial\Psi(v, x)}{N(x)}\,\frac{\partial N(x)}{\partial x} .& \end{aligned} \tag{25}$$

To pass from the volume of the drops to the radii, we use the equalities

$$\Psi(v, x)\,dv = f_x(r)\,dr\,, \quad f_x(r) = \frac{dv}{dr}\Psi(v, x)\,, \quad v = \frac{4}{3}\pi r^3\,.$$

As a result, we obtain the following equation for $f_x(r)$:

$$u\frac{\partial f_x(r)}{\partial x} = N(x)\left\{\frac{1}{2}\int_0^r \frac{r^2}{(r^3 - r_1^3)^{2/3}}\beta(r^3 - r_1^3, r_1^3)\, f_x(r^3 - r_1^3)^{1/3} f_x(r_1)\,dr_1 - \right.$$

$$\left. - f_x(r)\int_0^\infty \beta(r^3, r_1^3)\, f_x(r_1)\,dr_1\right\} - u\frac{f_x(r)}{N(x)}\frac{\partial N(x)}{\partial x}\,. \tag{26}$$

The function $N(x)$ can be defined in terms of $f_x(r)$ as

$$N(x) = V(x)/V_{\mathrm{m}}(x) = V(x)\bigg/\left(\frac{4}{3}\pi\int_0^\infty r^3 f_x(r)\,dr\right). \tag{27}$$

In Eq. (26), the function $\beta(r^3, r_1^3)$ has the meaning of the frequency of pair collisions of the drops in the disperse liquid. A qualitative analogy between turbulent and Brownian diffusions allows us to write the expression for the function $\beta(r^3, r_1^3)$ in the following form [11]:

$$\beta(r^3, r_1^3) = 4\pi\,[D_{\mathrm{t}}(r, x) + D_{\mathrm{t}}(r_1, x)]\,(r + r_1)\,, \tag{28}$$

here $D_{\mathrm{t}}(r, x)$ is the differential coefficient of turbulent diffusion that can be expressed in terms of the function $P_x(r)$ [12]

$$D_{\mathrm{t}}(r, x) = \delta\int_r^\infty (\tilde{r}P_x(\tilde{r}))^{1/2}\,d\tilde{r}\,, \tag{29}$$

where δ is the coefficient accounting for a possible increase in the diffusion of the drops in the case where the density of the drop is lower than that of the surrounding liquid.

The above-described processes of atomization and coalescence of the dispersed phase in the turbulent liquid flow occur simultaneously. Combining the right-hand sides of the equation for this function with allowance for the phenomena of coalescence, turbulent atomization, and reduction of the dispersed phase in size because of hydrodynamic cavitation, we write the resulting equation

$$u\frac{\partial f_x(r)}{\partial x} = N(x)\left\{\frac{1}{2}\int_0^r \frac{r^2}{(r^3 - r_1^3)^{2/3}}\beta(r^3 - r_1^3, r_1^3)\, f_x(r^3 - r_1^3)^{1/3} f_x(r_1)\,dr_1 - \right.$$

$$\left. - f_x(r)\int_0^\infty \beta(r^3, r_1^3)\, f_x(r_1)\,dr_1\right\} - u\frac{f_x(r)}{N(x)}\frac{\partial N(x)}{\partial x} - \frac{f_x(r)}{\tau_1(r)}\Theta[r - a_{\mathrm{cr}}(x)] +$$

$$+\int_r^\infty \frac{f_x(r)}{\tau_1(R)}\omega_1(r, R)\,dR - \frac{f_x(r)}{\tau_2(r)}\int_0^r \omega_2(\tilde{r}, r)\,d\tilde{r} + \int_r^\infty \frac{f_x(R)}{\tau_2(R)}\omega_2(r, R)\,dR\,. \tag{30}$$

The function $P_x(r)$ that describes the energy distribution over the scales of length r is calculated from the equation [6]

$$u\,\frac{\partial P_x(r)}{\partial x}=\frac{\partial}{\partial r}\left\{\left[\left(2\nu+2\gamma\int\limits_0^r\sqrt{rP_x(r)}\,dr\right)\left(\frac{\partial}{\partial r}+\frac{4}{r}\right)P_x(r)\right]\right\}+\frac{4}{3}\,\varepsilon\,\frac{r}{L^2}\exp\left(-\frac{r^2}{L^2}\right), \tag{31}$$

here ε is the rate of pumping of the turbulent energy due to the averaged-velocity field and $\gamma = \text{const} = 0.24$.

The equation for the function $f_x(r)$ will be solved later on.

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NOTATION

p, density of the dispersing liquid; L, turbulence macroscale; x, longitudinal coordinate of the homogenizer path; y, transverse coordinate of the homogenizer path; $\bar{p}(x)$, mean pressure at the point x of the homogenizer path; μ, mean velocity over the flow; $\Theta(x)$, Heaviside step function; R_{cav}, radius of the cavern; p_∞, pressure in the dispersing liquid; $V(x)$, total volume of the disperse liquid; $V_{\text{m}}(x)$, mean volume of the drop; v, kinematic viscosity. Subscripts: cr, critical; t, turbulent; col, collapse; cav, cavern; m, mean; f, force.

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