Effect of Structure of Turbulence on Drop Breakage*

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Possibilities of using several known models of microstructure of turbulence to model drop break-up rate are presented. The classical theory of turbulence neglects fine-scale intermittency. Here, intermittency is taken into account and modelled using multifractal formalism when applied to the inertial sub-range of turbulence. Maximum stable drop size, predicted by the classical theory of turbulence that neglects its intermittent character, is unequivocally determined by the mean energy dissipation rate and does not depend on the scale of the system provided that the mean energy dissipation rate is kept the same in each system. The breakage kinetics based on multifractal models of turbulence suggests a slow drift of the quasi-stable drop size to the stable drop size that is determined by the most vigorous turbulent events. Drop size distributions predicted by multifractal models are compared with predictions of the model based on the classical Kolmogorov theory of turbulence and with experimental data. All multifractal models predict the changes of the drop size in time better than the model based on the classical theory and are recommended to model the breakage processes.

Liquid—liquid dispersed systems are of importance to the chemical industry; typical examples are: extraction, many processes observed in chemical reactors including emulsion and suspension polymerization, preparation of emulsions, *etc.* Drop size distribution and its "dynamics" of changes are important features of dispersed systems as they have pronounced effect on the mass-transfer rate and the course of chemical reactions. Proper process and device designing, scale-up, and process operation require prediction of the drop size distribution by solution of the population balance equation. To formulate the population balance equation under conditions when drop coalescence can be neglected, one needs to use models and resulting expressions describing breakage frequency of drops of volume υ, $g_\upsilon(\upsilon, \vec{x}, t)$, at position $\vec{x}$ at time t, probability density function for daughter drops $\beta(\upsilon, \upsilon')$ representing the probability of forming of drops of volume υ from breakage of drops of volume υ', and the number of drops created during drop break-up $\nu_\mathrm{b}(\upsilon)$. Forms of these expressions depend considerably on the model of structure of turbulence used. The aim of this work is to present possibilities of using several known models of microstructure of turbulence to model drop break-up by using the population balance equation in the form

$$\frac{\partial n(\upsilon, \vec{x}, t)}{\partial t} = -g_\upsilon(\upsilon, \vec{x}, t)\; n(\upsilon, \vec{x}, t) + \int_\upsilon^\infty \beta(\upsilon, \upsilon')\; \nu_\mathrm{b}(\upsilon')\; g_\upsilon(\upsilon', \vec{x}, t)\, n(\upsilon', \vec{x}, t)\; \mathrm{d}\upsilon' \qquad (1)$$

To achieve this aim, at first models of turbulence microstructure are presented, then they are used to predict the maximum stable drop size. In the next part expressions for drop break-up rate $g_\upsilon(\upsilon, \vec{x}, t)$ are formulated and used to predict the drop size distribution and its time evolution. Then the results are compared with experimental data to verify the models.

THEORETICAL

Models of Turbulence

The first interpretation of the drop break-up in the turbulent field was presented in fundamental works of *Kolmogorov* [1] and *Hinze* [2]. In these works, the structure of turbulent field was interpreted using Kolmogorov—Obukhov theory [3, 4]. The Kolmogorov

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hypothesis defines the inertial sub-range

$$k_\mathrm{o} \ll k \ll k_\mathrm{K} \quad k_\mathrm{K} = 1/\langle\eta\rangle \tag{2}$$

where the kinetic energy is neither directly received from the mean flow, nor dissipated into internal energy.

For objects, the size of which falls within the inertial sub-range of turbulence ($\langle\eta\rangle \ll d \ll L$) this theory leads to the expression for normal pressure stress acting upon particles of diameter d dispersed in turbulent fluid

$$\overline{p(d)} \cong C_\mathrm{p}\rho_\mathrm{C} \left\langle \left[u_r\left(d\right)\right]^2 \right\rangle \cong C_\mathrm{p}\rho_\mathrm{C} \left[\langle\varepsilon\rangle\ d\right]^{2/3} \tag{3}$$

Presented expression is widely used in predicting of drop size in liquid—liquid dispersions. However, starting from experimental studies of *Batchelor* and *Townsend* [5] and pioneer analysis of *Obukhov* [6] and *Kolmogorov* [7] it is known that Kolmogorov theory is incomplete, as it neglects the problem of fine-scale intermittency that results in a strong variability of energy dissipation rate in time and space. Because of this variability

$$\langle\varepsilon^p\rangle \neq \langle\varepsilon\rangle^p \tag{4}$$

which also means that

$$\left\langle u_r^2 \right\rangle \neq \left(\langle\varepsilon\rangle\ r\right)^{2/3} \tag{5}$$

and eqn (*3*) is not correct.

In this work intermittency will be characterized using the method proposed by *Frisch* and *Parisi* [8]. Starting from the Navier—Stokes equation for $\rho =$ const and $\nu =$ const

$$\frac{\partial u_i}{\partial t} + u_j \frac{\partial u_i}{\partial x_j} = -\frac{1}{\rho}\frac{\partial p}{\partial x_i} + \nu \frac{\partial^2 u_i}{\partial x_j^2} \tag{6}$$

these authors formulated the principles of the multifractal model of turbulence. This model is based on the invariance of the Navier—Stokes equation after the following transformation

$$x_i' = \lambda\, x_i \tag{7}$$

$$u_i' = \lambda^{\frac{\alpha}{3}} u_i \tag{8}$$

$$t' = \lambda^{1-\frac{\alpha}{3}} t \tag{9}$$

provided that $\langle\eta\rangle < r$ ($r = \sqrt{(x_i)^2}$), $r' < L$, and $L \ggg \langle\eta\rangle$, which means that the inertial sub-range of turbulent scales is considered, where viscous effects are negligible. The exponent, α, and scale factor, λ, are arbitrary, provided that r and r' are in the inertial sub-range. The consequence of the above transformation is an analogous transformation for dynamic pressure

$$\left(\frac{p'}{\rho}\right) = \lambda^{2\alpha/3}\left(\frac{p}{\rho}\right) \tag{10}$$

that will be used in the next part of the paper. Eqn (*8*) enables us to show that the velocity increment over a distance r equals to

$$\begin{aligned} u_r = u_L \left(\frac{r}{L}\right)^{\alpha/3} &= \left[\langle\varepsilon\rangle\ L\right]^{1/3}\left(\frac{r}{L}\right)^{\alpha/3} = \\ &= \left[\langle\varepsilon\rangle\, r\right]^{1/3}\left(\frac{r}{L}\right)^{\frac{\alpha-1}{3}} \end{aligned} \tag{11}$$

Introducing now eqn (*11*) into eqn (*3*) gives normal stresses acting upon a particle of diameter d [9]

$$p\left(d,\alpha\right) = C_\mathrm{p}\rho_\mathrm{C}\left[\langle\varepsilon\rangle\ d\right]^{2/3}\left(\frac{d}{L}\right)^{\frac{2}{3}(\alpha-1)} \tag{12}$$

Turbulent events described by different exponent α appear in real flows with different probability. Application of the multifractal model of turbulence enables us to determine probability density function $P(\alpha)$ in the box of size r in the d_s-dimensional space

$$P(\alpha) \cong \rho(\alpha)\left(\frac{r}{L}\right)^{d_\mathrm{s}-f_\mathrm{d}(\alpha)} \quad \text{for } \frac{r}{L} \to 0 \tag{13}$$

where $\rho(\alpha)$ is an α-dependent factor, whereas the multifractal spectrum $f_\mathrm{d}(\alpha)$ is interpreted as a fractal dimension in a d_s-dimensional space. Using expressions (*11—13*) one can define several models of the turbulence microstructure.

For exponent $\alpha = \alpha_\mathrm{min} = \alpha_\mathrm{max} = 1$ one has $P(\alpha) = \delta(\alpha - 1)$, and the energy dissipation rate has uniform value $\langle\varepsilon\rangle$, which corresponds to the classical Kolmogorov theory [3].

The oldest model of intermittent turbulence is log-normal model [6, 7]. The log-normal distribution of the energy dissipation rate ε_r is equivalent to the parabolic distribution of $f_\mathrm{d}(\alpha)$

$$f_\mathrm{d}(\alpha) = D_\mathrm{o} - \frac{(\alpha-\alpha_\mathrm{o})^2}{4(\alpha_\mathrm{o} - D_\mathrm{o})} \tag{14}$$

where $d_\mathrm{s} = 1$, $D_\mathrm{o} = 1$, and $\alpha_\mathrm{o} = 1.117$ [10]. According to these authors eqn (*14*) agrees well with the results of measurements for $0.51 < \alpha < 1.78$. The next continuous α-distribution is empirical distribution proposed by *Bałdyga* and *Podgórska* [11]

$$f\left(\alpha\right) = a + b\alpha + c\alpha^2 + d\alpha^3 + e\alpha^4 + f\alpha^5 + g\alpha^6 + h\alpha^7 + i\alpha^8 \tag{15}$$

where $a = -3.51$, $b = 18.721$, $c = -55.918$, $d = 120.90$, $e = -162.54$, $f = 131.51$, $g = -62.572$, $h = 16.10$, $i = -1.7264$ for $d_\mathrm{s} = 1$ and $\alpha \geq 0.12$, where this minimum α value was measured by *Meneveau* and *Sreenivasan* [12].

A theoretical derivation of multifractal spectrum $f_\mathrm{d}(\alpha)$ was performed by *She* and *Leveque* [13]

$$f_\mathrm{d}\left(\alpha\right) = 1 + C_1'\left(\alpha - \frac{1}{3}\right) - C_2'\left(\alpha - \frac{1}{3}\right)\ln\left[\frac{1}{3}\left(\alpha - \frac{1}{3}\right)\right] \tag{16}$$

$$C_1' = \left[\frac{1 + \ln\left(\ln \frac{3}{2}\right)}{\ln \frac{3}{2}} - 1 \right] \quad C_2' = \frac{1}{\ln \frac{3}{2}}$$

In this model the minimum value of a multifractal exponent α is slightly larger than the value measured by *Meneveau* and *Sreenivasan* [12] and equals $\alpha_{\min} = 1/3$.

Stable Drop Size

Maximum stable drop size in a dilute noncoalescing dispersion can be calculated by a balance between dispersive turbulent stresses, which for inertial sub-range of turbulence are given by eqn (*12*), and stabilizing stresses. In the case of dispersed phase of low viscosity that is considered here the stabilizing forces are due to interfacial tension. The case of viscous drops is considered elsewhere [11].

According to the classical Kolmogorov theory [3] corresponding to $\alpha = 1$ and $P(\alpha) = \delta(\alpha - 1)$ the maximum stable drop size is given by the well-known equation

$$d^{\mathrm{o}}_{\max} = \frac{C_{\mathrm{x}} \sigma^{0.6}}{\langle \varepsilon \rangle^{0.4} \rho_{\mathrm{C}}^{0.6}} \qquad (17)$$

which in a dimensionless form is often formulated as $d^{\mathrm{o}}_{\max}/D \propto We^{-0.6}$.

The three multifractal models predict the correction factor A in the equation $d_{\max}/D \propto We^{-0.6A}$ where $A = [1 - 0.4(1-\alpha)]^{-1}$. In the case of log-normal energy dissipation rate there is no limitation for α. The log-normal distribution does not result in any stable drop size. In two other cases asymptotically stable size is determined as $d_{\max} \propto D We^{-0.926}$ and $d_{\max} \propto D We^{-0.818}$ for $\alpha_{\min} = 0.12$ and $\alpha_{\min} = 1/3$, respectively.

Drop Break-up Rate for Considered Models of Turbulence Microstructure

The break-up rate of drop of diameter d, $g_d(d, \vec{x}, t)$, is proportional to the frequency of turbulence motion on the scale of drop size d

$$g_d(d, \vec{x}, t) \propto \langle \varepsilon (\vec{x}, t) \rangle^{1/3} d^{-2/3} \qquad (18)$$

Eddies larger than d convey the drops and eddies smaller than d are not strong enough to disperse it. Of course, not all drop-eddy interactions lead to drop break-up. The multifractal models of turbulence predict directly which eddies are vigorous enough to cause drop break-up. *Bałdyga* and *Podgórska* [11] derived breakage frequency function by summing up for any position $\vec{x}$ at any time t the contributions to the break-up frequency from all vigorous enough eddies

$$g_d(d, \vec{x}, t) = \int_{\alpha_{\min}}^{\alpha_{\mathrm{x}}} g(\alpha, d, \vec{x}, t) \, P(\alpha, \vec{x}, t) \, \mathrm{d}\alpha \qquad (19)$$

The weakest eddies that are vigorous enough to cause drop break-up are labelled by the scaling exponent α_{x} resulting from the comparison of dispersive and stabilizing stresses

$$\alpha_{\mathrm{x}} = \frac{2.5 \ln\left(L(\vec{x}, t) \langle \varepsilon (\vec{x}, t) \rangle^{0.4} \rho_{\mathrm{C}}^{0.6} \sigma^{-0.6} C_{\mathrm{x}}^{-1} \right)}{\ln(L(\vec{x}, t)/d)} - 1.5.$$

Regarding drops smaller than the integral scale of turbulence, L, but not necessarily so small that $d/L \to 0$, one gets the expression for the drop break-up frequency in the form

$$g_d(d, \vec{x}, t) = C_{\mathrm{g}} \sqrt{\ln\left(\frac{L(\vec{x}, t)}{d}\right)} \langle \varepsilon (\vec{x}, t) \rangle^{1/3} d^{-2/3} \cdot \int_{\alpha_{\min}}^{\alpha_{\mathrm{x}}} \left(\frac{d}{L(\vec{x}, t)}\right)^{\frac{\alpha + 2 - 3 f(\alpha)}{3}} \mathrm{d}\alpha \qquad (20)$$

with $f(\alpha)$ given either by eqn (*14*), or (*15*) or (*16*).

RESULTS AND DISCUSSION

The maximum stable drop size predicted by the classical theory of turbulence, so neglecting its intermittent character, is unequivocally determined by the mean energy dissipation rate $\langle \varepsilon \rangle$ and does not depend on the scale of the system at constant $\langle \varepsilon \rangle$. The three multifractal models of turbulence suggest a slow drift of the quasi-stable drop size from the value described by $\alpha \cong 1$, corresponding to the most probable turbulent events, to the really stable drop size determined by the most vigorous turbulent events that are characterized by the smallest α value. It should be pointed out that using $f(\alpha)$ expressed in the form given by eqn (*15*) with $\alpha_{\min} = 0.12$ results in asymptotically stable drop size proportional to $We^{-0.93}$. Such an exponent on the Weber number was observed by *Konno* and *Saito* [14] after long agitation time.

The stirred tank turbulence is strongly inhomogeneous and various regions of the tank differ in properties of turbulence including energy dissipation rate, $\langle \varepsilon \rangle$, and integral scale of turbulence, L, so also drop break-up rates in the impeller zone should be much larger than in the bulk. To take this into account a one-dimensional, single circulation-loop plug flow model was used in this investigation. Along the loop there are defined zones differing in properties of turbulence. Two zones are assumed in this paper with local values of the average energy dissipation rate $\langle \varepsilon \rangle$ calculated using correlation by *Okamoto et al.* [15]. Integral scale of turbulence L in the impeller zone is assumed to be $L = 0.1\,D$ and in the bulk zone 3 times larger. The flow rate within the circulation loop is assumed to be equal to the pumping capacity of the impeller. The model enables calculation of the drop size distribution at any position in the circulation loop. Using the drop break-up frequency $g_d(d, \vec{x}, t)$, eqn (*20*), and probability density function for daughter drops, $\beta(v, v')$, one can predict evolution of the drop size distribution in time by solving the population balance equation (*1*). A U-shaped distribution of $\beta(v, v')$ proposed by *Tsouris* and *Tavlarides* [16] was used in modelling. Number of daughter drops was assumed to be 2. To solve the population balance equation (*1*) it was discretized in the drop volume domain $\left(v = \pi d^3/6\right)$ using the method of *Kumar* and *Ramkrishna* [17]. This method considers drop population in discrete size ranges concentrated at representative volumes. The size range contained between two volumes v_i and v_{i+1} is called the i-th section and is represented by the grid point w_i, such that $v_i < w_i < v_{i+1}$ and $v_i = (w_{i-1} + w_i)/2$. The grid applied in this work is of a geometric type with $w_{i+1} = 1.05w_i$. Events leading to the formation of drop sizes other than the representative size are incorporated into the set of discrete equations in such a way that two properties corresponding to two moments of interest are exactly preserved. In our case a new drop is assigned to the adjoining representative volumes in such a way that drop numbers and mass is preserved. The resulting set of ordinary differential equations was solved using the fifth-order Fehlberg method with error control and adjustment of the time-step of integration. Comparison of the drop size distributions obtained using the *Kumar* and *Ramkrishna* [17] method agrees very well with the results obtained using discretization technique based on uniform grid yielding extremely accurate solution. The advantage of the Kumar and Ramkrishna discretization method used is that it requires smaller number of size ranges. Resulting discrete drop size distributions are presented both as distribution curves for volume fraction, f_v *vs.* d, and cumulative distributions, F_v *vs.* d. Fig. 1 shows a comparison of the evolution of drop size distribution predicted by multifractal model with $f(\alpha)$ given by eqn (*15*) with experimental data reported by *Konno et al.* [18]. The authors studied break-up of drops in the system where dispersed phase was represented by the mixture of *o*-xylene and carbon tetrachloride and as a continuous phase the distilled water with a small amount of Na_3PO_4 was used. Physical properties of this system are as follows: $\rho_C = 998$ kg m^{-3}, $\rho_D = 1040$ kg m^{-3}, $\sigma = 0.034$ N m^{-1}. Dispersed phase volume fraction used in experiments was equal to $\phi = 0.002$. Parameters of the break-up model $C_g = 0.0035$ and $C_x = 0.23$ were determined by *Bałdyga* and *Podgórska* [11]. Experimentally determined drop size distribution for $t = 3$ min was used as an initial size distribution for calculations. As seen, the experimental and calculated drop size distributions agree well at short (10 min, 30 min) and long agitation times (300 min). It can be shown that the break-up takes place mainly in the impeller zone, where the energy dissipation rate is much larger than in the bulk. However, the drop size distributions in both zones are almost identical and the difference between them is too small to be shown in the figure. More examples of model predictions and experimental data one can find in the previously mentioned work [11], where the same pair of C_g and C_x parameters was used for different systems. Here we are rather interested in comparison of the multifractal model based on multifractal spectrum $f(\alpha)$ given by eqn (*15*) with two other multifractal models and the model based on the classical Kolmogorov theory of turbulence.

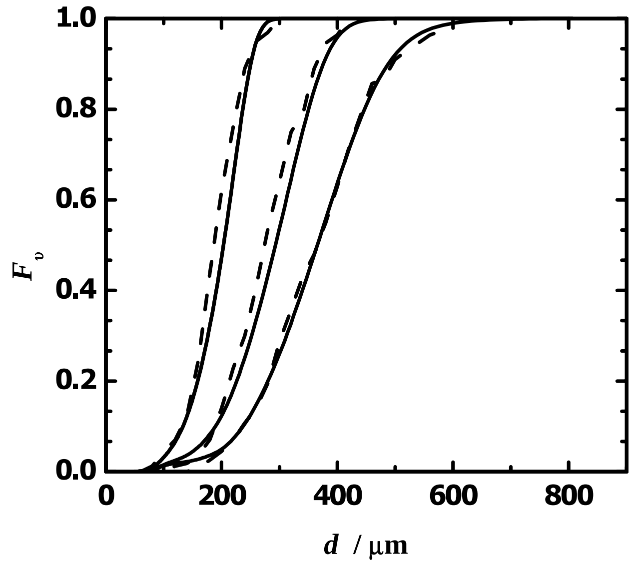

Fig. 1. Comparison between calculated (solid lines) and experimental [18] (dashed lines) transient drop size distributions (curves from right to left show distributions after $t = 10$ min, 30 min, 300 min). $T = 0.3$ m, $T/D = 2$, $N = 93$ min^{-1}.

Fig. 2 shows a comparison of drop size distribution predictions by the three multifractal models. It can be seen that all of them predict similar distributions, but differences between predictions increase with agitation time, when rare but most vigorous turbulent events affect the process. In the multifractal model corresponding to the log-normal distribution of energy dissipation rate, there is no limitation for multifractal exponent. This model predicts the fastest drop break-up. This is connected with the fact that $f(\alpha)$ agrees well with measurements only for $\alpha > 0.51$, for smaller α-values corresponding to the most violent bursts of turbulence the discrepancy between parabolic multifractal spectrum $f(\alpha)$ and experimental data of *Meneveau* and *Sreenivasan* [12] increases as α decreases. Application of the multifractal spectrum of *She* and *Leveque* [13] gives the slowest break-up, which results from the fact that the most vigorous eddies are determined by $\alpha_{\min} = 1/3$.

In the case of classical Kolmogorov theory of turbulence the break-up probability is given by the probability density function $P(\alpha) = \delta(\alpha - 1)$ that would

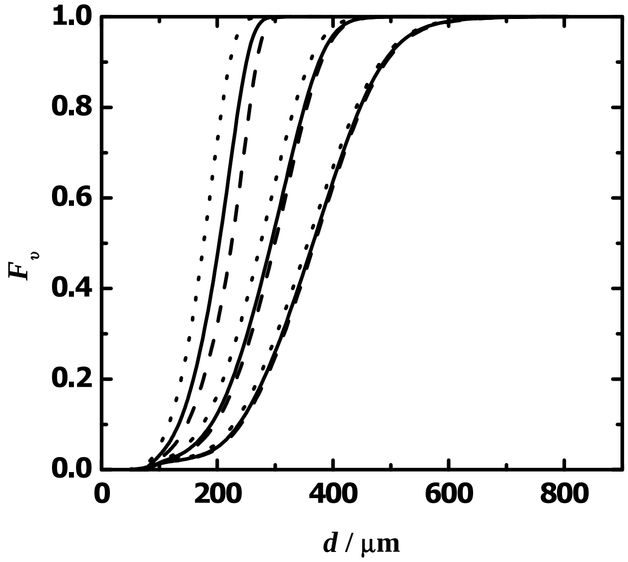

Fig. 2. Comparison of drop size distributions (curves from right to left show distributions after $t = 10$ min, 30 min, 300 min) for $T = 0.3$ m, $T/D = 2$, $N = 93\ \text{min}^{-1}$ predicted by different $f(\alpha)$ multifractal models: solid line – eqn (*15*), dashed line – model of *She* and *Leveque* [13], and dotted line – parabolic model.

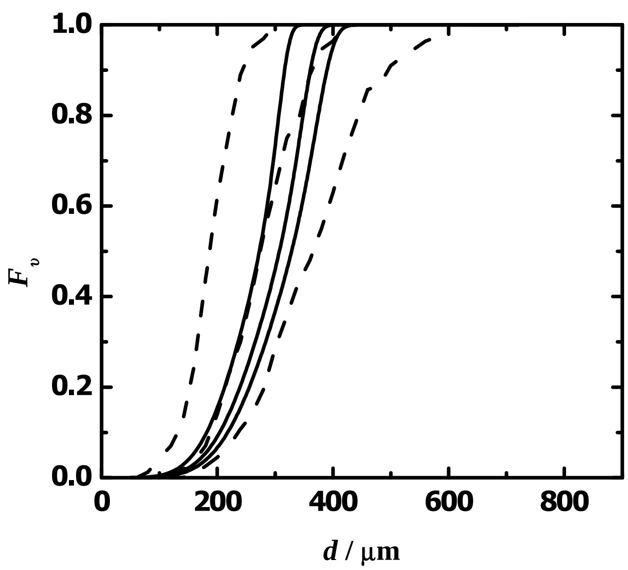

Fig. 3. Comparison between the experimental [18] (dashed lines), transient drop size distributions (curves from right to left show distributions after $t = 10$ min, 30 min, 300 min) and distributions calculated by the model of *Coulaloglou* and *Tavlarides* [19] (solid lines). $T = 0.3$ m, $T/D = 2$, $N = 93\ \text{min}^{-1}$.

be equal to 1, so additional assumption is needed. In what follows, the popular model of *Coulaloglou* and *Tavlarides* [19] is used. The authors assumed that the fraction of breaking drops is proportional to the fraction of turbulent eddies colliding with the drop that have energy greater than the droplet surface energy

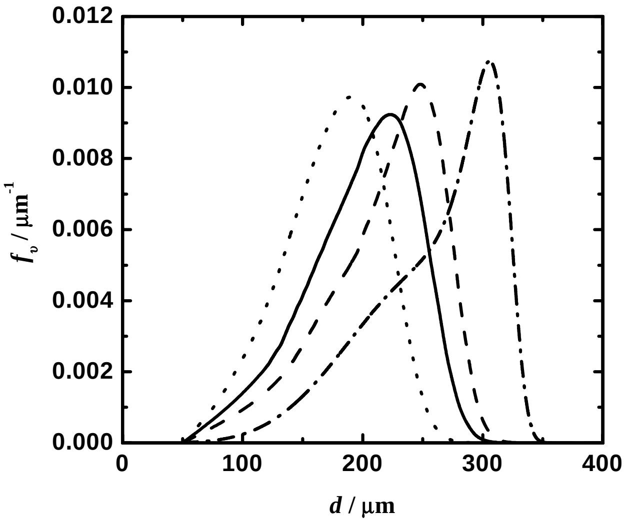

Fig. 4. Comparison of the drop size distributions after 5 h of agitation predicted by the model based on the classical theory of turbulence and multifractal models. Solid line – [11], dashed line – [13], dotted line – parabolic model, and dotted-dashed line – [19].

and that the fraction of eddies with kinetic energy greater than droplet surface energy is equal to the number fraction of eddies that have velocities greater than a corresponding fluctuating velocity. Fig. 3 shows evolution of drop size distribution predicted by the model of Coulaloglou and Tavlarides in comparison with the experimental data. It can be seen that the difference between the drop size distributions for short and long agitation times is smaller than that one predicted by multifractal models or observed experimentally. The differential volume fractions after 5-hour agitation predicted by all models discussed in this work are shown in Fig. 4. All multifractal models predict smaller droplets than the model based on the Kolmogorov theory; some differences between predictions of multifractal models can be used in the future for verification of models.

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SYMBOLS

A	correction factor in the exponent on Weber number	
C'_1, C'_2	constants in She and Leveque model of turbulence	
C_g, C_x	constants in the breakage model	
C_p	proportionality constant of order unity	
d_{max}	maximum stable drop diameter	m
d^o_{max}	maximum stable drop diameter when neglecting intermittency	m
d	drop diameter	m

d_s space dimension
D_o constant in eqn (*14*)
D impeller diameter m
$f_d(\alpha)$ multifractal spectrum
$f(\alpha)$ multifractal spectrum $f_d(\alpha)$ for $d_s = 1$
f_v differential volume fraction m^{-1}
F_v cumulative volume fraction
$g(\alpha, d, \vec{x}, t)$ characteristic frequency of eddies of scale d labelled by the multifractal exponent α s^{-1}
$g_d(d, \vec{x}, t)$ break-up rate of drops of diameter d at position $\vec{x}$ at time t s^{-1}
$g_v(v, \vec{x}, t)$ break-up rate of drops of volume v at position $\vec{x}$ at time t s^{-1}
k_K Kolmogorov wavenumber m^{-1}
k_o wavenumber for energy-containing eddies m^{-1}
k wavenumber m^{-1}
L integral scale of turbulence m
$n(v, \vec{x}, t)$ number density of drops of volume v at position $\vec{x}$ at time t m^{-6}
N impeller rotational speed s^{-1}
p pressure Pa
$p(d, \alpha)$ pressure stress acting upon drop of diameter d Pa
$\overline{p(d)}$ pressure stress acting upon drop of diameter d according to the Kolmogorov theory Pa
$P(\alpha)$ probability density function for α
r distance m
t time s
T tank diameter m
u, u_i velocity, velocity component $m\ s^{-1}$
u_L the r.m.s. velocity fluctuation $m\ s^{-1}$
u_r velocity difference over distance r $m\ s^{-1}$
v, v' drop volume m^3
We Weber number for stirred tank $\left(= N^2 D^3 \rho_C / \sigma\right)$
$\vec{x}$ position vector

Greek Letters

α_{max} supremum of multifractal exponent α
α_{min} infimum of multifractal exponent α
α_o constant in eqn (*14*)
α multifractal exponent
$\beta(v, v')$ probability density function for daughter drops m^{-3}
$\delta(x)$ Dirac delta function
ε_r local rate of energy dissipation averaged over a domain of size r $m^2\ s^{-3}$
ε turbulent energy dissipation rate per unit mass $m^2\ s^{-3}$
$\langle\varepsilon\rangle$ ensemble average of ε $m^2\ s^{-3}$
$\langle\eta\rangle$ Kolmogorov microscale m
λ scaling factor
ν kinematic viscosity $m^2\ s^{-1}$
$\nu_b(v)$ number of drops formed per breakage of drop of volume v
ρ density $kg\ m^{-3}$
$\rho(\alpha)$ factor in eqn (*13*)
σ interfacial tension $N\ m^{-1}$
ϕ dispersed phase volume fraction

Subscripts

C continuous phase
D dispersed phase

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