AIChE WILEY InterScience® DISCOVER SOMETHING GREAT

Modeling the Breakup of Fluid Particles in Turbulent Flows

Ronnie Andersson and Bengt Andersson
Dept. of Chemical and Biological Engineering, Chalmers University of Technology, SE-41296, Gothenburg, Sweden

DOI 10.1002/aic.10832
Published online March 24, 2006 in Wiley InterScience (www.interscience.wiley.com).

A model for breakup of fluid particles in turbulent flows is presented in this article. In the new model two criteria, one stress and one energy criteria, are required to be fulfilled for breakup to occur. A new model for interaction frequency between fluid particles and turbulent eddies is also introduced. Analysis of the model reveals that eddies close in size and up to three times larger than the fluid particle contribute most to the overall breakup rate. This explains the experimental findings by the authors and by other researchers presented recently in the literature that fluid particles often deform significantly before breakup occurs. A high-speed imaging technique, 4000 Hz, was used to measure the breakup rate directly without introducing assumptions regarding the daughter size distributions and the number of fragments formed upon breakup. Validation with these measurements shows that the new model gives excellent predictions. © 2006 American Institute of Chemical Engineers *AIChE J,* 52: 2031–2038, 2006
Keywords: breakup rate, deformation, bubble, drop, turbulence, multiphase

Introduction

Turbulent multiphase flows play a central role in chemical engineering. Understanding basic phenomena, such as breakup and coalescence of fluid particles, is the key to improving the modeling of the mass transfer and reaction rates in the chemical processes. Over the years several models have been proposed to predict the breakup rate and the daughter size distributions of fluid particles in turbulent flows. The fact that no unifying breakup model has been found implies that the phenomenon is not well understood. Only few studies exist in literature where the breakup of single bubbles and drops has been studied experimentally. Due to the lack of experimental data, the models proposed in the literature are based on several crude assumptions regarding the interaction between fluid particles and turbulent eddies. The lack of experimental data has resulted in widespread predictions of the total breakup rate, the resulting daughter size distribution, and the number of fragments formed upon breakup.[1] When experimental data are presented on fluid particle breakup, it is mainly transient measurements of the size distribution evolution that are reported.[2] Unfortunately, such data give no direct information on how breakup occurs. Improved methods are required to separate the breakup frequency from the daughter size distributions and number of fragments formed during breakup. The best method is direct experimental studies of single breakup events, which provide much more useful information. Unfortunately, such measurements are not trivial, since the phenomena occur on very small length scales at time scales comparable to the eddy turnover times, and time resolution in the order of 1-5 kHz is required.

Breakup Models—Literature Review

Breakup of bubbles and drops, hereafter referred to as fluid particles when no distinction between the two is required, has been the subject of investigation for several decades starting with the pioneering work by two researchers, Kolmogorov[3] and Hinze.[4] The early work mainly focused on determining correlations for the maximum stable fluid particle diameters, d_{max}, and the average fluid particle diameter, d_{32}, under various operating conditions. Although predictions of the average diameter are useful, it is in many cases more useful to know the

Correspondence concerning this article should be addressed to B. Andersson at bengt.andersson@chalmers.se.

size distribution and its evolution. For that purpose models of the breakup rates are required. Implementation of breakup and coalescence models in population balance equations allows such information to be calculated. A lot of work has been spent on deriving closures for the breakage and coalescence rates required in the population balance equations. In a recent work, the authors evaluated the potential of such an approach as a tool for virtual prototyping of liquid-liquid dispersions in a new type of chemical reactor.[2] It was concluded that reasonable predictions can be made but that more studies of the basic phenomena are required.

Several models are proposed in literature for the breakup rate of fluid particles. Most models for fluid particle breakup in turbulent flows are based on assumptions that there is no relative mean velocity difference between the continuous and dispersed phase. Under such conditions only the turbulent velocity fluctuations cause breakup. The breakup models proposed in literature can be divided into empirical and phenomenological models. The reader who wants an overview of the most frequently used breakup models can find more information in a recent review article.[1] The most promising of the breakup models are the phenomenological models that are derived on a theoretical basis and contain no adjustable model parameters.

The breakup frequency is usually modeled as the product of the collision frequency between turbulent eddies and fluid particles and a breakup efficiency. Definition of the collision frequency and the breakup efficiency requires several assumptions. The main drawback with these models is that several assumptions regarding the interaction between fluid particles and turbulent structure must be made in their derivation. Many models postulate, in analogy to the kinetic theory of gases, that the turbulent breakup is due to collisions between turbulent eddies and fluid particles, that is, the eddies bombard the fluid particle interface. These models require a definition of an effective swept volume, that is, the product between a cross section area and a characteristic velocity difference between the turbulent eddies and the fluid particles. In addition, it is required that the number density of eddies within a given size range is specified. To obtain such a relation it is assumed that the size range falls within the inertial subrange and that the energy is distributed according to the classical -5/3 power law. Furthermore, it is often assumed that the energy for a fixed wave number follows an energy distribution function. It is necessary to define the integration limits to close the breakup models. Particularly, the choice of the upper integration limit strongly affects the breakup frequency, that is, the choice of turbulent eddy sizes that affects breakup.[1] While the choice of the lower integration limit has low to moderate effect on the predicted breakup frequency, the upper limit can strongly affect the overall breakup frequency. The selection of the fluid particle diameter scale as the upper integration limit is done by several authors. It is reasonable to assume that large eddies, let's say an order of magnitude larger than the fluid particle, mainly transport the fluid particles and do not affect the breakup frequency. On the contrary, it is not reasonable to expect that eddies about twice as large as the fluid do not contribute to the breakup.

Luo and Svendsen proposed a model for breakup of fluid particles.[5] This model has been used in several studies to calculate the breakup rate in bubbly and droplet flows.[2,6-8] The main reason this model has become popular is that it differs from many other models in that it contains no adjustable model parameters. Hence, this model can be implemented when the hydrodynamics conditions and the fluid properties are known. The specific breakup rate is given by:

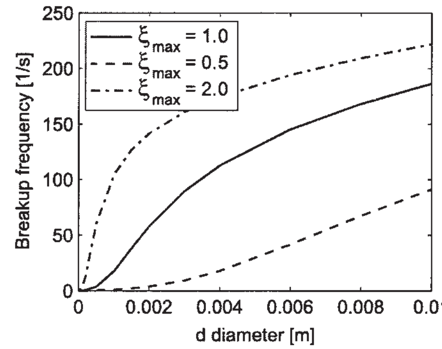

Figure 1. Predicted breakup frequency as function of the upper integration limit.

$$\Omega_{B,s}(d) = \frac{0.923}{2} \times \left(\frac{\varepsilon}{d^2}\right)^{2/3} \int_0^1 \int_{\xi_{\min}}^{\xi_{\max}} \frac{(1+\xi)^2}{\xi^{11/3}} \exp\left(-\frac{12 c_f \sigma}{\beta \rho \varepsilon^{2/3} d^{5/3} \xi^{11/3}}\right) d\xi df_{bv} \quad (1)$$

Although the model constants appearing in Eq. 1 are derived from theories of isotropic turbulence and no adjustable parameters are introduced, the model is very sensitive to the choice of the integration limits. Luo and Svendesen proposed that the lower integration limit is 11-31 times the Kolmogorov scale and the upper scale is equal to the diameter of the fluid particle. The dependence on the upper integration limit for the Luo and Svendsen model is shown in Figure 1. In this figure the breakup frequency is plotted as a function of the fluid particle diameter for three different upper integration limits, namely: half $\xi = 0.5$, the nominal $\xi = 1.0$, and the double $\xi = 2.0$ limit. Here $\xi = \lambda/d$ defines the ratio between the eddy size and the size of the fluid particle. Thus, the nominal value $\xi = 1.0$ means that only turbulent eddies less than or equal to the fluid particle size contribute to breakup. The calculations were done for gas bubbles, $\sigma = 0.072$ [N/m], and the energy dissipation rate was $\varepsilon = 10$ [m^2/s^3].

As seen in Figure 1, the relative difference between the predicted breakup frequencies is much larger for small fluid particles. For 2 mm particles, the ratio $\Omega_{B,s,\xi=2.0}/\Omega_{B,s,\xi=1.0} = 2.4$ and for 0.5 mm particles the ratio is $\Omega_{B,s,\xi=2.0}/\Omega_{B,s,\xi=1.0} = 17$. The strong dependence on the integration limits is not unique for the Luo and Svendsen model. It clearly shows the importance of defining the range of turbulent structures that interact with fluid particles correctly.

Hagesaether and colleagues proposed a refinement of the Luo and Svendsen model. One of the refinements was done by relaxing criteria of the upper integration limit.[9] Martinez and colleagues proposed a model that uses no restriction on the eddy scales. However, that model contains an empirical parameter.[10,11]

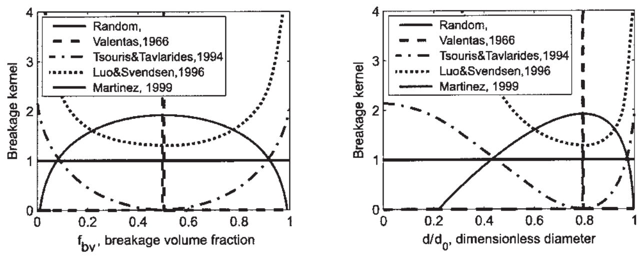

Figure 2. Breakage kernels proposed in literature.

(a) Kernels as function of the breakage volume fraction, (b) kernels as functions of the dimensionless diameter.

Besides prediction of the breakup frequency, the resulting daughter size distribution and the number of fragments formed upon breakup must be specified to describe the breakup process. Due to the limited and contradictory experimental data, the vast majority of all models are based on assumptions of binary breakage. Several of the breakup models are based on the surface energy increase arguments. The simplest model, assuming two equal size fragments, was suggested by Valentas and Amundson[12] in 1966. Since equal-sized breakup requires more energy compared to unequal sized breakup, these models typically predict highest probability for breakage into two fragments of unequal size. Tsouris and Tavlarides[13] comment that sufficient experimental data do not exist in literature to suggest adequate daughter drop probability density functions. They proposed a model for the daughter size distribution by assuming that the daughter size distribution is linearly related to the energy requirements for the formation of daughter drops. Hence, this model gives minimum probability for equal size drop breakup. Due to the lack of experimental data, they also assume that the average number of drops produced in the breakup process is two, that is, only binary breakage occurs. The model proposed by Luo and Svendsen[5] is based on the surface energy criteria. This model predicts a daughter size distribution that is U-shaped. Noteworthy is that this model predicts the highest probability for stripping of infinitesimally small fragments. This is due to the fact that it takes an infinitesimal surface energy increase to rip off infinitesimally small fragments from the mother particle.

An improvement of the Luo and Svendsen model was proposed by Hagesaether et al.,[14] who introduced a concept of energy density that reduced the probability that infinitesimal fragments were formed. Martinez[10] proposed a model where the probability of splitting of a fragment of a certain size is proportional to the difference between the disruptive stresses over a length scale corresponding to the length scales of the fragments and the confinement stresses of the mother particle. Contrary to the model proposed by Luo and Svendsen, this models gives an inverted U-shape. Hence, Martinez's assumptions result in a model that predicts highest probability for equal-sized breakup. Wang et al.[15] developed a model that accounts both for the surface energy increase and capillary pressure in the resulting fragment. Due to the capillary pressure criterion, this model also reduces the probability for splitting of infinitesimal fragments but still predicts the highest probability for unequal sized breakup. The daughter size distributions predicted by these models are shown in Figure 2. As seen in the figure, there is no general trend. The Tsouris model predicts zero probability for equal-sized breakup and maximum probability for unequal sized breakup, Luo's model predicts nonzero probability for equal-sized breakup and predicts a very high probability for stripping off infinitesimally small fragments, while the Martinez model gives highest probability for equal-sized breakup.

Despite the huge amount of work spent on developing models for drop and bubble breakage in turbulent flows, little experimental data exist that can be used to validate these models. Further improvement of the breakup rate model and the daughter size distributions clearly requires experimental data. In addition, due to the lack of experimental data, no improvements have been made in model development for the average number of fragments formed upon breakage. Some studies have shown that the binary breakup is not necessarily the most likely event. In a recent article by the authors, it was shown experimentally that the number of fragments formed upon breakup differ between bubbles and drops. That was also shown to be the case for the daughter size distribution.[16]

Model Development

The model presented here is based on measurements by us and others[16,17] showing that the fluid particles are highly deformed before breakup occurs. This deformation takes a few milliseconds, indicating that it is mainly larger long lived eddies that are responsible for the breakup. Furthermore, it was shown that small fragments are never ripped off a large fluid particle; instead, an internal flow redistribution mechanism was identified to form unequal sized fragments. These observations on the breakup process, along with models for the number densities of turbulent eddies, and eddy time scales, allow development of a physical breakup rate model. The main idea behind the new model is that the stress exerted by the turbulent eddies must be high enough to deform the fluid particles. This constitutes the stress criterion. This stress must also be maintained over some time for the particle to be stretched sufficiently to break. This constitutes the energy criterion. Both these criteria must be fulfilled for the particle to break.

The turbulence in the inertial sub-range is considered as very structured eddies and the stress acting on the fluid particles is due to local velocity differences. The velocity within a turbulent eddy is depicted in Figure 3a. The turbulence is created continuously, with the fluid particles randomly located in the turbulent field. The particles could be located in areas with small turbulent eddies (Figure 3b), in the outer part of a large turbulent eddy (Figure 3c), or within a large turbulent eddy (Figure 3d). In the last case, fluid particles with lower density than the continuous phase will move towards the low pressure region in the center of the eddy, heavier particles will move outward through the velocity gradient in the outer part, and neutral buoyancy particles will stay where they are and possibly break when the turbulent eddy after some time has decreased in size. Pairing of eddies is very frequent and trapping of fluid particles between two counter rotating eddies is a very effective way of transferring energy from the turbulent eddies (Figure 3e). Furthermore, the time for fluid particles to interact with an eddy is bounded by the lifetime of the eddy.

In the inertial sub-range the energy spectrum is given by $E(\kappa) = c_1\varepsilon^{2/3}\kappa^{-5/3}$ and the mean velocity fluctuations are given by $\bar{u}_\lambda = c_2(\varepsilon\lambda)^{1/3}$. By using these relations, Luo and

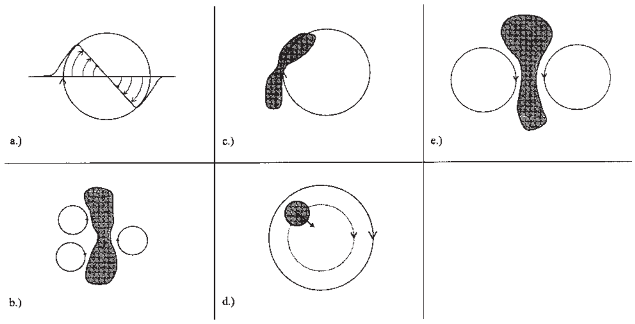

Figure 3. Interaction between turbulent structures and fluid particles.

(a) Velocity profile in a turbulent eddy; (b) deformation due to interaction with several small eddies; (c) deformation in the outer edge of the eddy; (d) transport with large eddy, possible movement towards eddy center for gas bubble but not for liquid drops; (e) deformation between paired eddies.

Svendsen showed that the number density of turbulent eddies in the interval λ to $\lambda + d\lambda$ is given by[5]:

$$\dot{n}_\lambda = c_3(1 - \alpha_d)/\lambda^4. \tag{2}$$

The notation dot $\dot{n}_\lambda$ is used to indicate that a variable is for a certain eddy size λ. In this case, n is eddies per volume with dimension $1/m^3$ and $\dot{n}_\lambda$ has the dimension $1/m^4$. The breakup rate is then given by an integration of an interaction frequency $\dot{\omega}(d_0, \lambda)$ and a probability $P(d_0, \lambda)$ that the particle will break up:

$$\Omega_B(d_0) = \int_{\lambda_{\min}}^{\lambda_{\max}} \dot{\omega}(d_0, \lambda) P(d_0, \lambda) d\lambda \tag{3}$$

We proposed that the interaction frequency, between turbulent eddies and a fluid particle, is proportional to the volume of the fluid particle, and that an eddy if energetic enough can cause one breakup during its turnover time. Assuming that the bubbles are randomly located in the turbulent flow, we can calculate the interaction between a bubble of size d_0 and a turbulent eddy of size λ from the product of number density and the volume of the particle $\dot{n}_\lambda \cdot (nd_0^3/6)$. The particle then interacts with this eddy during the eddy turnover time $\tau(\lambda)$. For constant epsilon over the eddy scales, the time scale of eddies of different sizes is given by $\tau(\lambda) = \lambda^{2/3}/\varepsilon^{1/3}$, assuming that an eddy of size λ has the energy $\frac{1}{2}u_\lambda^2 = \frac{1}{2}c_2^2(\varepsilon\lambda)^{2/3} \approx \varepsilon^{2/3}\lambda^{2/3}$, which dissipates at the rate ε. The interaction frequency is then for a given eddy size written as:

$$\dot{\omega}(d_0, \lambda) = \frac{c_3(1 - \alpha_d)}{\lambda^4} \frac{n_{d_0}\pi d_0^3}{6} \frac{\varepsilon^{1/3}}{\lambda^{2/3}} = \frac{c_3\pi(1 - \alpha_d)d_0^3\varepsilon^{1/3}n_{d_0}}{6\lambda^{14/3}} \tag{4}$$

Hence, regardless of how the fluid particle moves through the continuous phase, a definition of the interaction frequency that scales with the size of the fluid particles is obtained. Thus, the assumption of a swept volume, which requires models for the relative velocity and a cross-sectional area, is not introduced.

It is common to use the specific breakup rate and the specific interaction frequency. These are defined as the breakup rate and interaction frequency normalized with the number density of fluid particles in a size class and the dispersed phase holdup and has the unit 1/s. Hence, the specific interaction frequency is given by:

$$\dot{\omega}_s(d_0, \lambda) = \frac{\dot{\omega}(d_0, \lambda)}{n_{d_0}(1 - \alpha_d)} \tag{5}$$

and the specific breakup rate is given by:

$$\Omega_{B,s}(d_0) = \int_{\lambda_{\min}}^{\lambda_{\max}} \dot{\omega}_s(d_0, \lambda) P(d_0, \lambda) d\lambda \tag{6}$$

Traditionally, it is assumed that only eddies equal to or smaller than the fluid particle contribute to breakup. We do not apply this strict criterion since our observations show that both bubble and drop deformations prior to breakup involve large scale elongations. In our model, the breakup rate is modeled as the product of the interaction frequency and the breakup probability. While the interaction frequency strongly depends on the choice of the upper and lower integration limits, the breakup rate should not. This is possible when physical criteria are defined for the breakup probability. In our model, two criteria are used: the energy criterion and the stress criterion.

The average eddy energy is given by:

$$\bar{e}(\lambda) = \rho_c \frac{\pi\lambda^3}{6} \frac{\bar{u}_\lambda^2}{2}. \tag{7}$$

In contrast, when an eddy that is larger than the fluid particle interacts, it is reasonable that energy can be transferred only partially. For large eddies, $\lambda > d_0$, the energy can be estimated from the energy contained in the toroid volume formed by the cross section of the particle and the circumference of the turbulent eddy:

$$\bar{e}_{\text{limited}}(\lambda, d_0) = \frac{\rho_c}{2}\bar{u}_\lambda^2 \pi\lambda \frac{\pi}{4} d_0^2 = \frac{\rho_c \pi^2}{4} d_0^2 \varepsilon^{2/3} \lambda^{5/3}. \tag{8}$$

The available energy is determined according to the size ratio between the turbulent eddy and the fluid particle:

$$\bar{e}_{available}(\lambda, d_0) = \min[\bar{e}(\lambda), \bar{e}_{\text{limited}}(\lambda, d_0)] \tag{9}$$

It is assumed that for each λ there is a distribution of fluctuating velocity and the normalized energy distribution is given by[9]:

$$\varphi(\chi) = \exp(-\chi), \tag{10}$$

where $\chi = e(\lambda)/\bar{e}(\lambda) = u_\lambda^2/\bar{u}_\lambda^2$ defines the ratio of the eddy energy to the average eddy energy and $\int_0^\infty \varphi(\chi)d\chi = 1$. Hence, the distribution around the mean energy is independent of the eddy size.

Our observations show that the fluid particles always go through a highly deformed state before breakup occurs. Thus, the energy associated with the deformed complex may be seen

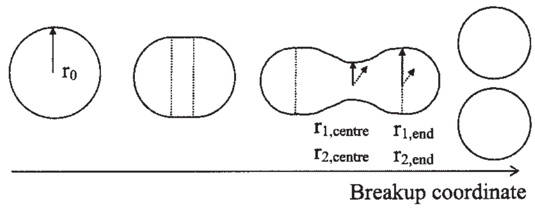

Figure 4. Deformation and breakup of a fluid particle.

as an activation energy. The increase in interfacial energy is given by $\sigma\pi d_0^2\gamma$. It was shown experimentally that $\gamma \approx 0.3$. We postulate that breakup occurs when the eddy energy exceeds the increase in interfacial energy, for the deformed complex, and when the disruptive turbulent stress exceeds the cohesive interfacial stress. Thus, two criteria are used to predict the probability for breakup when a turbulent structure and fluid particle interact.

The breakup criterion based on the interfacial energy increase for the deformed complex is given by:

$$e_{\text{interfacial energy}}(d_0) \geq \sigma\pi d_0^2\gamma. \tag{11}$$

In dimensionless form, this criterion is written as $\chi_{\text{interfacial energy}} \geq e_{\text{interfacial energy}}(d_0)/\bar{e}_{available}(\lambda, d_0)$. This criterion states that there must be enough energy available for deformation and breakup. In addition, it is required that the disruptive stress equals or exceeds the interfacial stress.[18] In a similar way, the stress criterion is given by:

$$\tau_t \geq \tau_i \tag{12}$$

Here τ_t denotes the disruptive stress, that is, the dynamic pressure due to the turbulent eddies. τ_i is the stabilizing interfacial stress due to interfacial tension. Equation 12 equals:

$$\rho_c \frac{u_\lambda^2}{2} \geq \frac{2\sigma}{d} \tag{13}$$

where d is the characteristic diameter of the deformed particle. The stress exerted by the turbulent eddy must balance the stress difference over the deformed fluid particle. As shown in Figure 4, the radius is a function of time and position. When the dumbbell form is obtained, one of the curvatures points outwards instead of inwards. This means that the maximum stress required to deform the particle occurs at a certain point before the deformed particle eventually breaks up.

Equation 13 can be written $u_\lambda^2 \geq 4\sigma/\rho_c d$ and in dimensionless form the stress criterion is given by $\chi_{\text{disruptive stress}} \geq (4\sigma/\rho_c d)/\bar{u}_\lambda^2$.

According to the hypothesis, breakup occurs when the two criteria are fulfilled, that is, the eddy energy exceeds the required energy for deformation and the disruptive stress exceeds the stabilizing interfacial stress. The distribution function of eddy energy allows the fraction of eddies that fulfill both criteria to be calculated. The minimum dimensionless eddy energy required for breaking a fluid particle is given by $\chi_{\min} = \max[\chi_{\text{interfacial energy}}, \chi_{\text{disruptive stress}}]$.

In this model d_0 was used as an estimate of d in Eq. 13, and the probability that a turbulent eddy of size λ will break a fluid particle of size d_0 is then calculated from:

$$P(d_0, \lambda) = \int_{\chi_{\min}}^{\infty} \varphi(\chi)d\chi \tag{14}$$

The breakup probability predicted by Eq. 14 increases with eddy size. For small eddies the energy criterion is the strongest criterion, whereas the stress criterion is the most restricting for large turbulent eddies.

Using this breakup model, the choice of the upper and lower integration limits in Eq. 6 is not crucial. This is due to the fact that the very small eddies, although many in number, do not fulfill the stress and energy criteria. In contrast, very large eddies fulfill both criteria, particularly the energy criterion, but due to the low number density and the long lifetimes of these eddies, they contribute little to the overall breakup rate. The integration limits may be taken as $\lambda_{\min} = d_0/10$ and $\lambda_{\max} = 10d$. The specific breakup rate is thus given by:

$$\Omega_{B,s}(d_0) = \int_{d_0/10}^{10d_0} \dot{\omega}_s(d_0, \lambda)\, P(d_0, \lambda)d\lambda \tag{15}$$

As shown in the Results and Discussion section, the breakup rate is only significantly affected by eddies close in size to the fluid particles. Hence, increasing the integration limits only affects the computational time, not the predicted breakup rate.

Experimental Measurements of the Breakup Rate

Traditionally the breakup rates are calculated by solving the inverse problem, that is, determine the breakup rate from the evolution of the size distribution by solving the populations balance equations. This method benefits from the fact that it is enough to obtain still images of the dispersion at different positions along a flow coordinate. However, the method requires assumption of the resulting daughter size distribution and the number of fragments formed upon breakup. Hence, the breakup rates are not determined explicitly and errors are easily introduced into the calculations since the breakup rate cannot be completely separated from the daughter size distribution and the number fragmentation.

The alternative method to overcome these drawbacks is to make direct measurements of the breakup rate. All successful breakups are counted within a control volume together with the total number of fluid particles within each size category. This is, however, not a trivial task since it requires that the breakup dynamics is resolved so that each particle can be identified. In the present work, such measurements were performed by using a digital high-speed imaging technique.

The measurements were performed by injecting drops in a turbulent flow and analyzing the outcome in a given measurement volume. To determine the breakup rate, it is not enough to identify only the drops breaking up but also the drops that do not break up. Knowledge of the fraction of drops that did break up and the residence time allowed breakup rate to be calculated correctly. By dividing the drops into discrete size classes, the

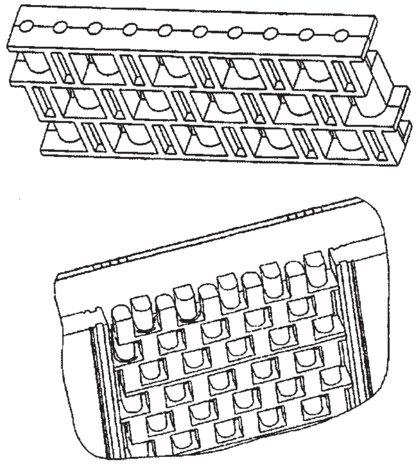

Figure 5. Reactor used in the experimental study.

(a) Flow-directing insert; (b) mixing elements in the flow insert.

breakup rate for drops of different sizes were calculated. This method allows the breakup rates to be calculated correctly at the cost of very high processing times. Unfortunately, such measurements generate very large amounts of data. In addition, since it is not enough to determine the size of each particle on the images, which itself is a difficult task to do for fluid particles, the particles located at different positions in a sequence of images must be identified correctly. Due to the high processing times, the breakup rates are reported for a narrow range of drop sizes. This is not a severe limitation since the model proposed in this article does not contain any adjustable parameters and the data are used to validate the model, not to develop a model from experimentally measured breakup rates.

The measurements were performed in a new multipurpose reactor developed by Alfa Laval, shown in Figure 5. The Reynolds number based on the hydraulic diameter is in the range of 4000 to 10,000 in the experiments. This appears low but the reactor is designed to give high turbulence at low Reynolds numbers. A better measure of turbulence is the Taylor scale Reynolds number Re_λ.[19] Re_λ is in the range 51 to 77 and a Re_λ in this range indicates that there is an inertial subrange, but the overlap between the production scales and the dissipation scale is large. A Re_λ of 77 corresponds to an Re of 66,000 in a straight pipe with the same hydraulic diameter. The energy dissipation rate, determined from CFD simulations, was close to 10 m^2/s^3, and typical sizes of drops that broke up were about 1 mm in diameter. This agrees well with the classical stress analysis proposed by Hinze, which predicts that 1 mm dodecane drops will not sustain the disruptive turbulent stresses under these conditions.[4]

Results and Discussion

A comparison between the measured and predicted breakup rates for dodecane drops in a turbulent flow is shown in Figure 6. The measured breakup rates are determined from observations of approximately 300 drops; and the specific breakup rates, Eq. 6, are calculated for three drop sizes, for the same hydrodynamic conditions and fluid properties as in the experimental study. Approximately 100,000 images, 100 GB data, were recorded and analyzed.

In addition, the predictions were compared to the ones from the breakup model proposed by Luo and Svendsen.[5] This model has been used successfully by the authors and other researchers.[2,8] The main reason this model was chosen was that it is derived from first principles and contains no adjustable model parameters. It is, therefore, possible to use this model without extensive experiments and parameter fitting. The authors have used that model previously to describe drop size distributions in this reactor and concluded that the Luo and Svendsen model gives good predictions of the Sauter mean diameter evolution.[2] In that study, simulations based on a CFD-population balance modeling approach, it was also concluded that width of the predicted size distributions was too large. In view of results from the new model presented in this article, this observation can be explained.

As shown in Figure 6, the new model gives better predictions of the breakup rates than the model proposed by Luo and Svendsen. However, partly this is due to the fact that the model proposed by Luo and Svendsen gives the conditional breakup rate, that is, it gives breakup rate into fragments of different sizes. This means that the daughter size distribution can be calculated directly from the breakup rate model. In a recent article, the authors showed that the Luo and Svendsen model, which predicts highest probability for unequal-sized breakup of all fluid particles, does not agree with experimental measurement. In fact, there are several differences between bubble and drop breakup. One of these differences is that bubbles often result in unequal-sized fragments but drops results in equal-sized fragments.[16]

The Luo and Svendsen model predicts highest probability for breakup into unequal-sized fragments. Our studies on drop breakup show, on the contrary, that equal-sized breakup has highest probability.[16] Thus, although too high a breakup rate of drops is predicted with the Luo and Svendsen model, it is possible to predict the evolution of the Sauter mean diameters. However, the resulting size distribution will not be predicted correctly. This was also noted by the authors but could not be explained at that time.[2] Fewer breakup events are required to reduce the diameter when fragments of equal sizes are formed. Considering that the Lou and Svendsen model predicts highest probability for unequal-sized breakup, the breakup rates predicted with their model and the model proposed here are consistent in the sense that they predict a similar rate for reducing the Sauter mean diameter.

Many published models are sensitive to the size of turbulent

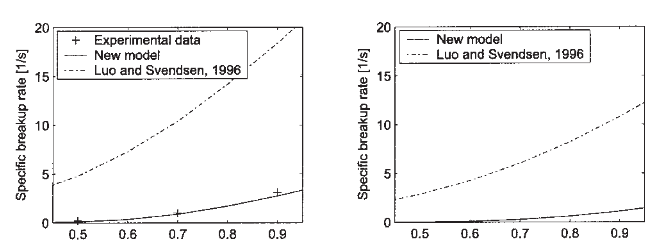

Figure 6. Specific breakup rate.

(a) Comparison of predicted and measured specific breakup rates, dodecane drops, $\varepsilon = 8.5$ [m^2/s^3], $\sigma = 0.053$ [N/m]; (b) comparison of the model by Luo and Svendsen and the new model, air bubbles, $\varepsilon = 8.5$ [m^2/s^3], $\sigma = 0.072$ [N/m].

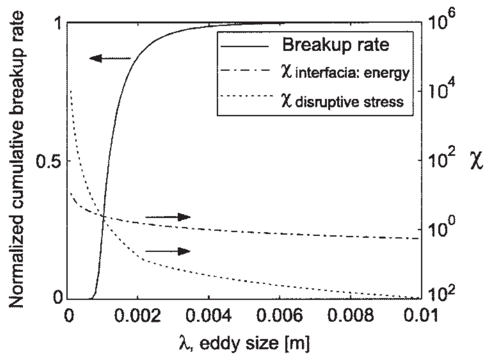

Figure 7. Contribution of turbulent eddies to the total breakup rate of a 1 mm dodecane drop, $\varepsilon = 8.5$ [m^2/s^3].

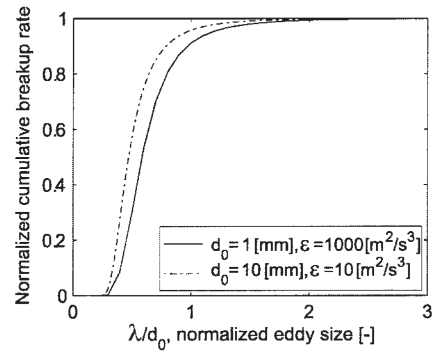

Figure 9. Cumulative breakup rate for fluid particles far from the equilibrium size.

eddies that is selected to interact with the fluid particles. The new model proposed here is not sensitive to the choice of these limits. The normalized breakup rate, calculated using the following integration limits: $\lambda_{min} = d_0/10$ and $\lambda_{max} = 10d_0$, for a 1 mm drop is shown in Figure 7. As shown in this figure, eddies smaller than the drop diameters hardly contribute to the overall breakup rate, since they do not fulfill the two physical breakup criteria. In contrast, turbulent eddies with size between 1 and 2mm contribute most to the breakup. Hence, the integration limits could be reduced to $\lambda_{min} = d_0$ to $\lambda_{max} = 2d_0$ and still account for 80% of the breakup. The reason why very large eddies do not contribute much to the overall breakup is simply because they are very few, as given by the number density of turbulent eddies in the interval λ to $\lambda + d\lambda$ $\dot{n}_\lambda = c_3(1 - \alpha_d)/\lambda^4$.

Often the upper limit for eddies responsible for breakup is taken as the size of the fluid particle.[5,15,20] Experimental observations have shown that fluid particles stretch significantly before breakup occurs.[16,17] Eddies much smaller than the fluid particle cannot generate such large scale deformations. According to the predictions with our new model, it is the turbulent eddies whose size is approximately equal to and up to three times larger than the fluid particles that cause breakup. These results make it much more understandable how bubbles and drops can undergo a highly deformable state before breakup occurs. For fluid particles that are close to the equilibrium size, this behavior is similar for small and large drops. As shown in Figure 8, both 1 mm and 10 mm fluid particles are affected most by turbulent eddies that are close in size up to 2-3 times their diameter.

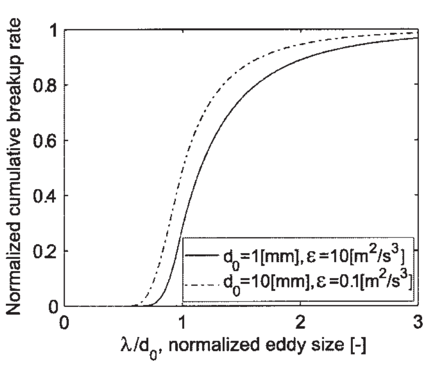

Figure 8. Cumulative breakup rate for fluid particles close to the equilibrium size.

For fluid particles that are much larger than the equilibrium size, the proposed model predicts that eddies smaller than the fluid particle also contribute significantly to the total breakup rate. This is reasonable since these eddies then contain enough energy and exert high enough stress to fulfill both criteria in the breakup model. As a consequence, fluid particles that are far from the equilibrium size may not be deformed to the same extent, that is, a factor 2-3 times the initial diameter. This, however, remains to be investigated experimentally. Simulations for 1 and 10 mm fluid particles at turbulent energy dissipation rates 100 times the ones in Figure 8 are shown in Figure 9. Clearly, for fluid particles much larger than the equilibrium size, the model predicts that turbulent eddies smaller than the fluid particle size contribute to more than 90% of the total breakup rate. In an STR the energy dissipation rate may differ two orders of magnitude between the impeller and bulk region. Under such conditions, it is reasonable that breakup may occur without the fluid particles being deformed significantly prior to breakup.

Conclusions

Traditionally most models proposed in the literature assumed that only eddies equal to or smaller than the size of the fluid particle cause breakup. In a recent article, the authors reported that both bubbles and drops are subject to large scale deformations prior to breakup. This means that turbulent structures close in size to the fluid particles should be the ones that cause breakup. Hence, the strict criterion on the eddy size that contributes to breakup is not reasonable to use. Instead, the possible bandwidth of eddies that cause breakup should be relaxed. Furthermore, most of the models proposed in the literature require assumptions of a cross sectional area and relative velocity to define the collisions frequency. These assumptions introduce additional uncertainties in the breakup model and should, therefore, be avoided, if possible.

A new model that is insensitive to the upper and lower interaction limits was successfully devised. In contrast to other models proposed in the literature, this model predicts that turbulent structures close in size to the fluid particle contribute most to the breakup. The new model was formulated in terms of interaction frequencies, which do not require any assumptions regarding the collision frequencies. Validation with experimental measurements of the breakup rate shows that this model gives better results than other models proposed in the literature. The new model predicts that the eddy sizes that significantly contribute to the overall breakup rates are given by a relatively small range of eddies. It was also found that fluid particles close in size to the thermodynamic equilibrium are more affected by eddies somewhat larger than the particle. This agrees with the experimental observations in this study and by other authors, which show that fluid particles often are subject to large deformation before breakup occurs.

The properties of the new model presented in this work can be summarized as:

- The concept of collision frequencies is not used; instead, interaction frequencies are used.
- Two physical criteria, based on energy and stress restrictions, were successfully implemented.
- In contrast to other models in literature, the new model is not sensitive to the upper and lower integration limits.
- Predictions from the new model give very good agreement with experimental measurements of the breakup rate.
- Comparison with other models in literature shows that the new model gives better predictions.
- Eddies whose size is approximately equal to and up to three times as large as the fluid particles are responsible for breakup.

The new model requires an additional model for distribution of daughter sizes and number of fragments before it can be implemented in CDF-population balance modeling to validate if it can predict the evolution of both the Sauter mean diameter and the size distribution. At the moment, there is no available model that can predict the distributions. The data presented in a recent article by the authors shows that the daughter size distribution differs between bubble and drops and a successful model should, therefore, be able to account for this difference. It was also shown that while bubble breakup mainly results in two fragments, binary breakup has low probability for drop breakup.

Notation

d = drop diameter, m
e = eddy energy, kg m^2 s^{-2}
$\bar{e}$ = mean eddy energy, kg m^2 s^{-2}
E = energy spectrum, m^3 s^{-1}
$\dot{n}_\lambda$ = number density of eddies, m^{-4}
P = probability function, -
R = radius, m
$\bar{u}_\lambda$ = mean velocity fluctuations, m s^{-1}

Greek letters

α_d = volume fraction of the dispersed phase, -
ε = turbulent energy dissipation rate per unit mass, m^2 s^{-3}
κ = wave number, m^{-1}
λ = eddy size, m
μ = dynamic viscosity, kg m^{-1} s^{-1}
ν = kinematic viscosity, m^2 s^{-1}
ρ = density, kg m^{-3}
σ = interfacial tension, kg s^{-2}
τ = stress, kg m^{-1} s^{-2}
φ = energy distribution function for eddies, -
χ = ratio of eddy energy to mean eddy energy, -
ω = interaction frequency, m^{-3} s^{-1}
Ω = breakup rate, m^{-3} s^{-1}

Superscripts and subscripts

B = breakup
c = continuous phase
d = dispersed phase
i = interfacial stress
s = specific breakup rate
t = turbulent stress

Literature Cited

1. Lasheras JC, Eastwood C, Martinez-Bazan C, Montanes JL. A review of statistical models for the break-up of an immiscible fluid immersed into a fully developed turbulent flow. *Int J Multiphase Flow.* 2002;28: 247-278.
2. Andersson R, Andersson B, Chopard F, Noren T. Development of a multi-scale simulation method for design of novel multiphase reactors. *Chem Eng Sci.* 2004;59:4911-4917.
3. Kolmogorov AN. On the breakage of drops in a turbulent flow. *Dokl Akad Nauk SSSR.* 1949;66:825-828.
4. Hinze JO. Fundamentals of the hydrodynamic mechanism of splitting in dispersion processes. *AIChE J.* 1955;1:289-295.
5. Luo H, Svendsen HF. Theoretical model for drop and bubble breakup in turbulent dispersions. *AIChE J.* 1996;42:1225-1233.
6. Chen P, Dudukovic MP, Sanyal J. Three-dimensional simulation of bubble column flows with bubble coalescence and breakup. *AIChE J.* 2005;51:696-712.
7. Chen P, Sanyal J, Dudukovic MP. Numerical simulation of bubble columns flows: effect of different breakup and coalescence closures. *Chem Eng Sci.* 2005;60:1085-1101.
8. Venneker BCH, Derksen JJ, Van den Akker HEA. Population balance modeling of aerated stirred vessels based on CFD. *AIChE J.* 2002;48: 673-685.
9. Hagesaether L, Jakobsen HA, Svendsen HF. A model for turbulent binary breakup of dispersed fluid particles. *Chem Eng Sci.* 2002;57: 3251-3267.
10. Martinez C, Montanes JL, Lasheras JC. On the breakup of an air bubble injected into fully developed turbulent flow. Part 1. Breakup frequency. *J Fluid Mechanics.* 1999;401:157-182.
11. Martinez C, Montanes JL, Lasheras JC. On the breakup of an air bubble injected into a fully developed turbulent flow. Part 2. Size pdf of the resulting daughter bubbles. *J Fluid Mechanics.* 1999;401:183-207.
12. Valentas KJ, Bilous O, Amundson NR. Analysis of breakage in dispersed phase systems. *Industrial Eng Chem Fundamentals.* 1966; 5:271-279.
13. Tsouris C, Tavlarides LL. Breakage and coalescence models for drops in turbulent dispersions. *AIChE J.* 1994;40:395-406.
14. Hagesaether L, Jakobsen HA, Svendsen HF. Theoretical analysis of fluid particle collisions in turbulent flow. *Chem Eng Sci.* 1999;54: 4749-4755.
15. Wang T, Wang J, Jin Y. A novel theoretical breakup kernel function for bubbles/droplets in a turbulent flow. *Chem Eng Sci.* 2003;58:4629-4637.
16. Andersson R, Andersson B. On the breakup of fluid particles in turbulent flows: *AIChE J.* 2006; in press.
17. Eastwood CD, Armi L, Lasheras JC. The breakup of immiscible fluids in turbulent flows. *J Fluid Mechanics.* 2004;502:309-333.
18. Levic VG. *Physicochemical Hydrodynamics*. New York: Prentice Hall; 1962.
19. Pope S. *Turbulent flows*. Cambridge: Cambridge University Press; 2000.
20. Lehr F, Mewes D. A transport equation for the interfacial area density applied to bubble columns. *Chem Eng Sci.* 2001;56:1159-1166.

Manuscript received Oct. 9, 2005, and revision received Feb. 11, 2006.