Breakup model of oscillating drops in turbulent flow field

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Abstract

Following the drop oscillation breakup mechanism, a theoretical model for drop breakup probability is proposed based on the three-dimensional Maxwell velocity distribution. The model considers both the interfacial energy increase constraint and viscous energy increase constraint. The model shows that for low-viscous drops, the breakup probability is determined by the Weber number (We_L), and for intermediate or high viscous drops, the breakup probability is determined by the combined influence of the Weber number (We_L) and the Ohnesorge number (Oh). By combining the theoretical model of drop breakup time constructed in our previous work, the breakup frequency model is obtained based on the statistical description framework. The accuracy and generality of the model were then validated using the direct experimental data. Moreover, effects of the drop diameter, turbulent energy dissipation rate, and interfacial tension on the predicted drop breakup frequency were analyzed in detail.

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Abstract: Following the drop oscillation breakup mechanism, a theoretical model for drop breakup probability is proposed based on the three-dimensional Maxwell velocity distribution. The model considers both the interfacial energy increase constraint and viscous energy increase constraint. The model shows that for low-viscous drops, the breakup probability is determined by the Weber number (We_L), and for intermediate or high viscous drops, the breakup probability is determined by the combined influence of the Weber number (We_L) and the Ohnesorge number (Oh). By combining the theoretical model of drop breakup time constructed in our previous work, the breakup frequency model is obtained based on the statistical description framework. The accuracy and generality of the model were then validated using the direct experimental data. Moreover, effects of the drop diameter, turbulent energy dissipation rate, and interfacial tension on the predicted drop breakup frequency were analyzed in detail.

Keywords: Oscillation; breakup model; Maxwell velocity distribution; turbulent energy dissipation; interfacial tension

1 Introduction

Turbulent dispersions of immiscible liquids are attracting and focused topics in practical applications, such as solvent extraction, food processing, chemical reaction, and so forth. Behaviors of drop swarms such as drop breakup and coalescence are fundamentally important as the understanding of those basic phenomena can improve the quantitative analysis of two-phase dispersion characteristics, interphase mass transfer, and interface reaction rate. Drop breakup refers to the process in which a spherical drop is deformed and stretched by the external flow and finally breaks up into several fragments. A complete description of the drop breakup usually requires knowledge of the following aspects, that is, the breakup probability of a drop with a certain size, duration of the drop breakup process, and number and size distribution of the fragments. The first two aspects are typically described using the concept of drop breakup frequency. Over the years, masses of breakup models for liquid drops were proposed by researchers, however, the existing breakup models vary from each other in forms and predicted results, implying a lack of understanding of the drop breakup mechanism. To gain more insight into the drop breakup phenomenon, single drop breakup experiments were carried out by some research groups[1–7]. Results of the single drop breakup experiments verified that drop diameter, interfacial tension, and external energy input are key parameters determining the drop breakup behaviors. However, the direct experimental data concerning the influence of various parameters on drop breakup, especially breakup time and breakup frequency, are still very limited. Moreover, no unifying drop breakup mechanism was found from the reported studies on drop breakup. To obtain the direct experimental data of drop breakup behaviors and analyze the mechanism of drop breakup in turbulent dispersions, since 2016, our group has been engaged in the study of breakup behaviors of drop swarms under turbulent conditions.[8–12] Through systematic experimental studies in different apparatuses, we quantified the influences of operating parameters and physical properties of the two phases on drop breakup. The direct experimental data of drop breakup frequency, breakup time, daughter drop size distribution, and so forth were obtained. In our recent work[13], we found that the drop breakup behavior is directly related to the second-order oscillation of the drop, correspondingly, breakup models can be constructed based on the drop oscillation breakup mechanism.

2 Breakup kernels for liquid drops

To quantitatively describe the drop breakup behavior, models of drop breakup frequency and daughter drop size distribution (DDSD) are crucial. As for the DDSD, U-shaped[14,15], M-shaped[16,17], and inverted U-shaped distributions[18–20] are generally reported in the literature. For binary breakup of liquid drops, the inverted U-shaped distribution has been adopted by most researchers and also verified by experimental data[8,9,21,22]. There are many DDSD models related to the inverted U-shaped distribution, and these models have limited impact on the quantitative study of the drop breakup process despite their differences. As such, researchers mainly focus on the modeling of the drop breakup frequency. Although a vast number of drop breakup frequency models exist in the literature, the vast majority of them are constructed under a limited number of modeling frameworks. In particular,frameworks based on the eddy-particle collision and statistical description are generally adopted for modeling. According to the eddy-particle collision framework, drop breakup frequency is the product of the eddy-drop collision frequency and the breakup efficiency. Prince and Blanch[23] gives an expression in the integral form based on the turbulent eddy size, then Luo and Svendsen[15]introduced the impact of the volume fraction of fragments and proposed the modeling framework in the double integral form, as is expressed in Eq..

Where is the collision frequency between eddies and drops, is the breakup efficiency for a mother drop with size *d*. In the statistical description framework, drop breakup frequency is counted as the product of the inversed breakup time and breakup probability, which is expressed as Eq..

Where is the drop breakup time, is the breakup probability of a drop with a diameter of d , and are the number of breakup events and total number of drops respectively. This framework was firstly proposed by

Coulaloglou and Tavlarides[18] in 1977 and then wildly adopted by researchers.

By comparing Eq. and Eq., it can be seen that the physical meanings of the breakup probability (or called breakup efficiency) in the two modeling frameworks are equivalent to each other. Therefore, the main difference between the two modeling frameworks comes from the processing of the time term, which is described by the breakup time in the statistical description framework, and by the collision frequency in the eddy-particle collision framework. In comparison, the physical meaning of the breakup time is more explicit, which describes the time scale of the drop breakup process. To model the drop breakup time, some researchers[5,18], starting from the eddy turnover time, assume that . According to our recent study[24], the above correlation will be valid only if the turbulent stress is much greater than the interfacial stress. However, such cases are less likely to happen in liquid-liquid dispersions, especially under steady-state operating conditions. To quantify the influence of each parameter on the drop breakup time, our group performed systematic experimental[13] and theoretical studies[24] and proposed a novel breakup time model based on the drop oscillation breakup mechanism:

Where c_1 and c_2 are constant parameters, $c_1 = 14.0$ and$c_2 = 8.0$. We_L has a form of the Weber number and characterizes the relative magnitude of turbulent stress and interfacial stress, We_L is expressed as Eq..

Where L is the maximum scale of the local turbulent structure. In practical calculations, L values by the minimum length-scale of the local runner[24].

For low-viscous drops, the influence of the drop viscosity on drop breakup time is insignificant, thus, Eq. can be simplified to Eq.:

Eq. and Eq. were validated by the systematic experimental data and can be applied to Eq. directly. In combination with accurate modeling of the drop breakup probability, an accurate description of drop breakup frequency can be achieved.

This study aims to construct an accurate model of the drop breakup frequency. Firstly, based on the three-dimensional Maxwell velocity distribution and the drop oscillation breakup mechanism, the theoretical model of the drop breakup probability is deduced. Further, the breakup frequency model is obtained by combining the breakup probability model with the breakup time models of Eq. and Eq.. Subsequently, the direct experimental data of drop breakup frequency in different apparatus are used to verify the accuracy and applicability of the constructed model in this study. After that, drop breakup frequencies versus key parameters are calculated, meanwhile, the impacting mechanisms of the operating parameters and properties of two-phase on drop breakup frequency are analyzed. The results of this study can be directly applied to the population balance model, which further serves for the accurate prediction of drop size distributions and dispersion characteristics in liquid-liquid dispersion equipment.

3 Model development

3.1 Theoretical model of drop breakup probability

The drop breakup probability characterizes the instability of a drop under the acting of the external force and can be statistically expressed as the ratio of the number of broken drops to the total number of mother drops, that is:

Various forms of drop breakup probability models exist in the literature, and the majority of these models are constructed based on Maxwell-Boltzmann velocity/energy distributions. However, due to different selections of breakup constraints, there are significant differences in the physical significance and predicted results of the models constructed by different researchers. According to our recent research[13], the drop breakup process is directly related to the oscillation behaviors of the drop surface. This implies that the breakup probability can be modeled based on the oscillation mechanism.

To build a breakup probability model based on surface oscillation, we adopt the following assumptions:

The turbulence is assumed to be locally isotropic.

This assumption is widely adopted in model constructions. Kolmogorov pointed out that at sufficiently high Reynolds numbers, the fine-scale structure of turbulent flows is statistically characteristic.[25] For devices such as stirred tanks and extraction columns, experimental results also indicate that the turbulent structure is locally isotropic.[26–28]Therefore, the theory of local isotropy of turbulence can be used in the construction of the breakup model.

Only binary breakup is considered.

Experimental studies showed that the binary breakup is dominant in steady-state liquid-liquid dispersions, where the diameter of most drops is close to the equilibrium size.[8,9,21,29] However, as the drop size deviates from the equilibrium size, the proportion of the binary breakup decreases accordingly. [2,3,9] According to our previous studies [8,21], the tensile breakup is the most common breakup pattern in turbulent dispersions. For multiple tensile breakups, usually, two larger daughter drops and several satellite drops are generated. Since the contribution of satellite drops is relatively small (according to the description of Andersson and Andersson [6,30], satellite drops account for about 8% volume of the mother drop), the effect of satellite drops can be approximately neglected and the binary breakup assumption can still be considered as a reasonable choice.

The drop will break up when the surface oscillation velocity exceeds a critical limit.

When a drop comes in contact with the turbulent eddy, energy is transferred to the drop surface in the form of turbulent pulsations, which cause the oscillation of the drop surface. According to the breakup process captured by the high-speed camera, drop breakup is mainly caused by the second-order oscillation, higher-order oscillations are almost non-contributory to the drop breakup. In the second-order oscillation, the drop moves along a central axis in a stretching-contraction cycle, and the relative velocity between the two poles of this axis is defined as the oscillation velocity, which is positive for the stretching motion and negative for the contraction motion, and the direction of the central axis is defined as the direction of the oscillation velocity. The oscillation velocity defined in this way is a vector, and its magnitude is defined as the oscillation speed. When the oscillation speed is too small to break the drop, the energy absorbed from the turbulent pulsation will be dissipated in several oscillation periods, and the drop eventually reverts to the original. On the contrary, when the oscillation speed is large, the drop does not have enough time to dissipate the energy through surface oscillations and eventually breaks up to form several fragments (only the binary breakup is considered in this study). We refer to the minimum oscillation speed at which the drop breaks up as the critical oscillation speed, . (For simplification, the velocity and the speed are both expressed by the same word ‘velocity’)

The definition of the second-order oscillation velocity shows that it is a three-dimensional random vector in the locally isotropic turbulence, and therefore conforms to the three-dimensional Maxwell velocity distribution. The three-dimensional Maxwell velocity distribution can be expressed as:

Herein is the root-mean-square velocity of the surface oscillation. is the probability density function. Integrating Eq. and we obtain the expression of the drop breakup probability:

Solve Eq. and we obtain Eq.:

As a result, the expression of the drop breakup probability based on the three-dimensional Maxwell velocity distribution is obtained. Herein is the upper incomplete gamma function. Next, we need to determine the expressions for the root-mean-square oscillation velocity and the critical oscillation velocity.

The oscillation of the drop surface is caused by the random pulsation of the turbulence. The average oscillation kinetic energy per unit drop volume can be expressed as follows:

The drop oscillation kinetic energy is equal to the turbulent kinetic energy transferred to the drop surface, which, according to the energy spectrum theory of turbulence, is expressed as:

Where is the energy spectrum and$k$ is the wavenumber of the turbulence. Kolmogorov's hypotheses give the universal form of the energy spectrum in the inertial subrange:

The minimum wavenumber in Eq. inversely proportional to the scale of the maximum turbulent structure capable of transferring energy to the drop surface, which can be calculated by Eq..

Where L is the maximum scale of the local turbulent structure and$d$ is the drop diameter. a takes the value of 1.5. Substituting Eq. and Eq. into Eq., the expression for the turbulent kinetic energy is then obtained, as shown in Eq..

Taking , then we obtained the expression for the root-mean-square velocity :

To determine the expression for the critical oscillation velocity , the criteria for drop breakup need to be clarified. According to the oscillation breakup mechanism raised above, external turbulent pulsations acting on the drop surface cause the surface oscillations, and the oscillation energy is continuously propagated and dissipated at the surface in the form of capillary waves. When the oscillation energy is small, the energy will be eventually dissipated after several oscillation periods, conversely, when the energy exceeds a certain value, the drop will break up. Therefore, drop breakup should satisfy the energy constraint, that is, the surface oscillation energy is greater than the critical value, . According to Andersson and Helmi[30], there is a maximum interfacial energy increase during the drop breakup (), which is larger than the interfacial energy increase before and after the drop breakup (). Most models in the literature only consider , thus causing an underestimation of the energy constraint.[30] In present work, we use the maximum interfacial energy increase during the breakup process as the energy constraint, that is:

Meanwhile, we have:

The parameter of c characterizes the relative magnitude of the energy potential of the breakup process, According to Andersson and Helmi[30], the maximum interfacial energy increase has to be approximately 1.5 times the interfacial energy increase before and after drop breakup. According to the experimental results in our previous studies[8,9], drops are more inclined to undergo the equal breakup due to the turbulent energy can be better utilized under such cases. For binary equal breakups, then $c$values as 0.39. For practical drop breakup processes, drops are not ideally equal breakup, thereby leading to a smaller value of $c$than 0.39. By comparing with the experimental data, we find that$c$ takes a value of 0.365 to better match the experimental data, which will be discussed in detail in the following section.

From Eq. and Eq., it is obtained that:

Combining Eq., Eq., and Eq., the expression for the drop breakup probability is obtained as Eq..

Where We_L is the same with Eq..

3.2 Theoretical model of drop breakup frequency

According to the statistical description framework of Eq., the drop breakup frequency can be expressed as the product of the reciprocal of the breakup time and the breakup probability. Substituting Eq. and Eq. into Eq., the theoretical model of drop breakup frequency for low-viscous drops is then obtained, as shown in Eq..

4 Results and discussion

4.1 Validation

Recently, our group measures the drop breakup frequency in a pulsed disc and doughnut column[9,21] and in a pump-mixer[8,10,13]. The experimental data can be directly used for the model validation. To compare the experimental data of different researchers conveniently and to show the results intuitively, the dimensionless abscissa and ordinate were adopted. The We_L is used as the abscissa and drop breakup probability is used as the ordinate. It should be noted that only the experimental data of the drop breakup frequency is provided in our previous work, therefore, it needs to be further processed in conjunction with the breakup time model to obtain the data of the breakup probability, that is . The above processing is still essentially a direct experimental data validation of drop breakup frequency rather than drop breakup probability. Another point to note is the value of$L$, which characterizes the maximum scale of the local turbulent structure. L depends on equipment parameters and can be valued by the minimum length-scale of the local runner[24]. For the pulsed disc and doughnut column used by Zhou et al.[9,21], L values as the distance between the adjacent disc and doughnut plate, $L = 30$ mm. For the stirred tank, L values as the height of the blade, that is $L =$ 10 mm (pump-impeller in Zhou et al.[8,10]) and $L = 8.8$ mm (Rushton Turbine impeller in Zhou et al.[13]).

In our previous study, the multiple breakup is artificially treated as the cascaded binary breakup. Although this treatment does not affect the application of the breakup model to the population balance model (as the cascaded binary breakup assumption is also applied in the construction of the daughter drop size distribution model). However, from a practical physical point of view, the cascaded binary breakup assumption leads to an overestimation of the breakup probability of small-sized drops. To avoid the impact of the cascaded binary breakup assumption on the model validation, experimental data of the initial drop breakup are adopted and are applied to compare with the predicted results of the breakup model proposed in this work, as shown in Figure 1.

It can be seen that the experimental data and the predicted results of the model almost converge on a single curve, implying that the two are in good agreement, which also verifies the accuracy and applicability of the breakup model constructed in this study.

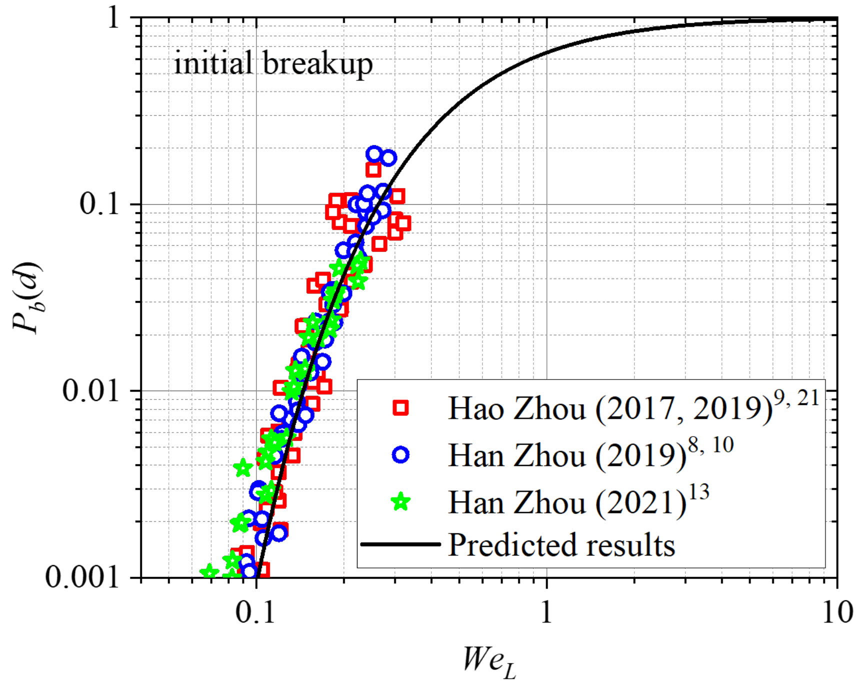

Figure 1 Comparison of the predicted results with the initial breakup experimental data (Hao Zhou (2017, 2019)[9,21], Han Zhou (2019)[8,10], Han Zhou (2021)[13])

To further verify the generalizability of the breakup model constructed in the present work, experimental data of drop breakup frequency which are obtained by single drop breakup experiments were chosen. Andersson and Andersson [6] measured the drop breakup frequency of the kerosene-water system and three valid data points were provided in the reported literature. This work was carried out in a static mixer, the maximum scale of the local turbulent structure L was taken as the local flow channel width, $L = 3$ mm according to the information provided by Bouaifi et al. [31] Ashar et al.[3] measured the drop breakup frequency of the rapeseed oil-water system in a rotor-stator mixer, and L was also taken as the local flow channel width (stator opening width), $L = 6$ mm. Maaß and Kraume[1] determined the drop breakup probability of the toluene-water and petroleum-water systems in a mimic stirred tank, the value of L is taken as the blade height,$L = 14.5$ mm. It should be noted that the value of the turbulent energy dissipation rate was not directly given in their original work, but only the apparent fluid flow speed in the channel was provided as 1.5 m/s. According to the work of Maaß et al.[32], the blade tip speed v_{tip} in a stirred tank corresponding to this speed is $v_{tip} = 1.5/0.7$ m/s $= 2.14$ m/s. Accordingly, the turbulent energy dissipation rate in the vicinity of the impeller can be calculated as [33,34]:

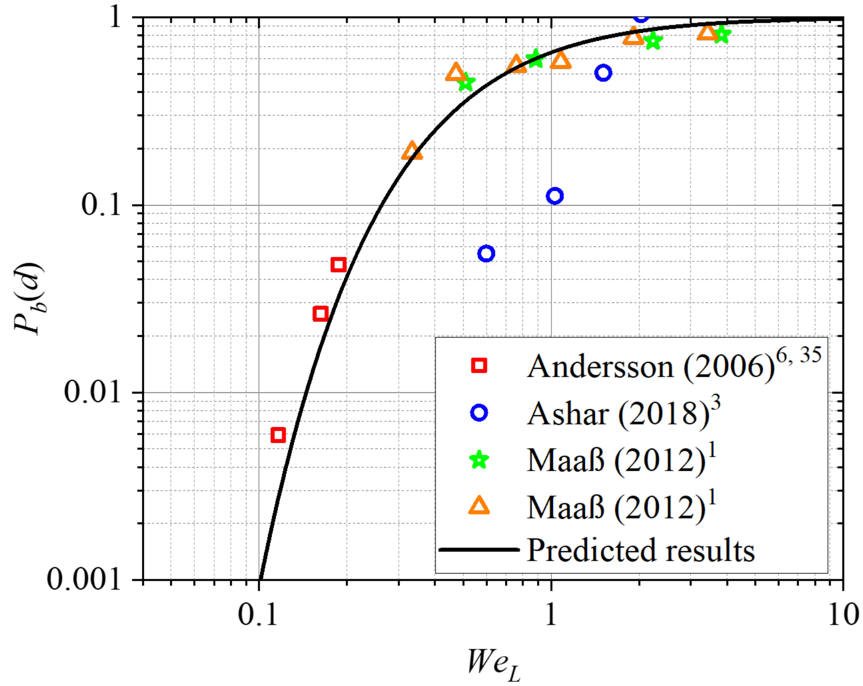

Figure 2 Comparison of the predicted results with experimental data in the literature[1,3,6,35]

Combining the above parameters with the physical data provided in the literature, we nondimensionalized the experimental data and plotted them in Figure 2. It can be seen that the predicted results of the model are in good agreement with the experimental data of Andersson and Maaß, which further validates the accuracy and generalizability of the theoretical model. However, Figure 2 shows that the predicted results of the model are in relatively poor agreement with the experimental data of Ashar et al., especially at the lower We_L , which we analyze as a result of the higher drop viscosity of the experimental system in Ashar et al. Moreover, the high spatial inhomogeneity of the turbulent energy dissipation rate in their experimental device can also cause deviations between the model predictions and experimental results.

4.2 Comparison of drop breakup frequency models

As is discussed in Section 2, most breakup frequency models in the literature are based on limited modeling frameworks. Particularly,frameworks based on the eddy-particle collision and based on the statistical description are mostly adopted by researchers. Breakup frequency models proposed by Coulaloglou and Tavlarides[18] (CT model, as shown in Eq.) and proposed by Luo and Svendsen[15] (Luo model, as shown in Eq.) are the most representative and well-known models under the above two frameworks, respectively.

CT model:

Luo model:

In such a scenario, the two models are also analyzed in the present work. To intuitively compare the experimental data of drop breakup frequency obtained by different researchers, the method recommended by Lehr et al. [36] to dimensionless the drop size and drop breakup frequency is introduced, that is:

Where L^* and T^* have length and time dimensions respectively:

In the original model of Coulaloglou and Tavlarides[18], the drop breakup frequency depends on the dispersed phase density, that is, the density ρ_φ in Eq. and Eq. takes the value of the dispersed phase density ρ_δ . Firstly, we compared the predicted results of the CT model with the experimental data, as shown in Figure 3a. It can be seen that by changing the specific values of the adjustable parameters, the original CT model can conform to the experimental data under different experimental facilities, but the accurate prediction of the breakup frequency under different facilities cannot be achieved by a fixed combination of the parameters. From the original literature of Coulaloglou and Tavlarides, it can be seen that the density term comes from the estimation of the turbulent stress. According to the current cognition of most researchers, the magnitude of the turbulent stress should be related to the continuous phase density, which means that the dispersed phase density in the CT model should be replaced by the continuous phase density, and thus, the density ρ_φ in Eq. and Eq. is taken as the continuous phase density ρ_ς in the following analysis. And then, we compare the predicted results of the density-corrected CT model with the experimental data, and the result is shown in Figure 3b. It can be seen that by adjusting the adjustable parameters combination, the modified CT model can better match the experimental data of different researchers relative to the original model. Also, we plot the predicted results of the Luo model in Figure 3b, and it shows that the Luo model overestimates the actual magnitude of the breakup frequency.

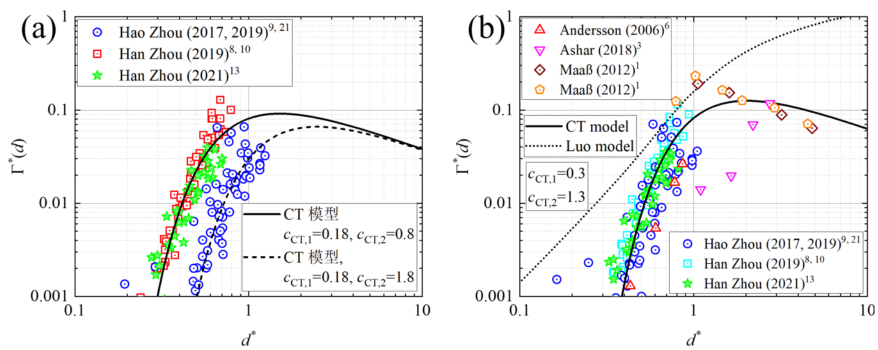

Figure 3 Comparison of the predicted results with experimental data. (a) based on the dispersed phase density; (b) based on the continuous phase density

For the CT model, the adjustable parameters can be optimized to fit the experimental data of different researchers, while the predictive capability of the CT model is still not comparable to the breakup frequency model constructed in this study. Moreover, it should be noted that the breakup time in the CT model is assumed to be linearly related to the eddy turnover time, . As is discussed in our previous study[13,24], this is only applicable in the case of We_L [?] 1, which is difficult to be satisfied for most liquid-liquid dispersion systems under normal operating conditions.

4.3 Sensitivity analysis of model parameters

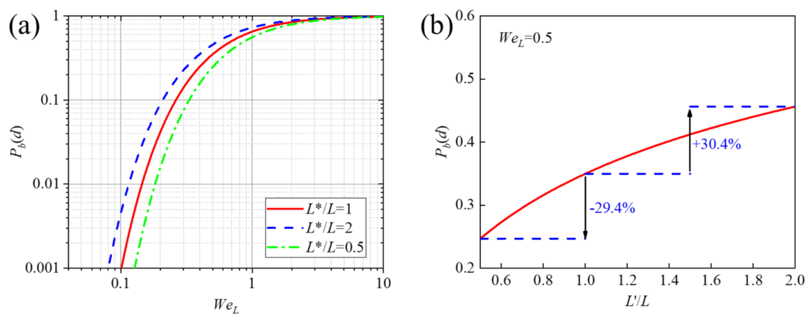

Figure 4 Sensitivity analysis of model parameters. (a) prediction results of the breakup probability versus We_L ; (b) prediction results of the breakup probability versus

In the constructed breakup model, L characterizes the maximum scale of the local turbulence structure and is directly related to the parameters of the apparatus. For practical liquid-liquid dispersion equipment, L values as the minimum scale of the local runner. For example, in a pulsed disc and doughnut column extraction column, the value is taken as the distance between plates. For mixing equipment, since drop breakup occurs mainly in the region of the blade runner,L is taken as the blade height. For static mixers, L is counted as the runner width of the minimum mixing unit, and so forth. Based on the above criteria, analysis in Section 4.1 shows that the predicted results of the model are in good agreement with the experimental data, which indicates the accuracy and applicability of the criteria valuing L . However, due to the diversity and complexity of the practical apparatus, the minimum scale of the local runner may deviate from the maximum scale of the local turbulence structure, thereby influencing the predictive accuracy of the proposed breakup model. In such a scenario, the sensitivity of the value of L to the predicted breakup probability is analyzed in this section, as shown in Figure 4. Herein, denotes the value of the maximum scale of the local turbulence structure and L denotes the actual value used for the model calculation. Figure 4a shows the prediction results of the breakup probability versus We_L , it indicates that the larger the value of , the larger the predicted result of the breakup probability, which is because the larger the value of means the higher the turbulent stress and thus the drop is more prone to break up. Besides, Figure 4b shows that the deviation of the predicted results of the breakup probability model is approximately within $\pm30\%$ when the value of fluctuates between 0.5 and 2.0.

4.4 Influence of the drop diameter on drop breakup frequency

Figure 5 shows the calculated drop breakup frequency versus drop diameter. It indicates that the drop breakup frequency firstly increases with the increase of the drop diameter and then reaches it's maximum, further increase the drop diameter will decrease the predicted drop breakup frequency. The definition of the breakup frequency (see Eq. in Section 2) indicates that it is the product of the reciprocal of the breakup time and the breakup probability. As both the drop breakup probability and breakup time increase with the increase of drop diameter when the drop diameter is relatively small, with the increase of the drop diameter, the breakup probability increases more quickly than the breakup time, and the overall performance is that drop breakup frequency increases with the increase of drop diameter. However, when the drop diameter exceeds a certain value, the effect of drop diameter on the breakup time is more significant than that of the breakup probability, and then the performance is that the breakup frequency gradually decreases with the increase of drop diameter. Moreover, Figure 5 shows that as the turbulent energy dissipation rate increases or the interfacial tension decreases, the peak value of the breakup frequency gradually increases and moves toward smaller drop diameters, indicating that higher turbulent kinetic energy dissipation rate and lower interfacial tension are more favorable for the drop to break up, which is consistent with the experimental conclusions in our previous studies[8,9].

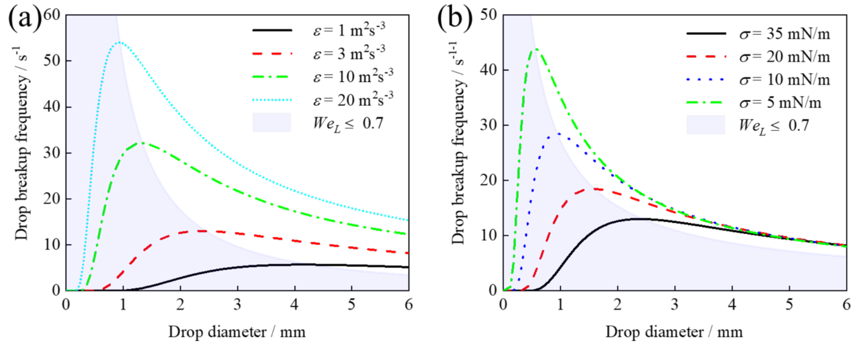

Figure 5 Influence of the drop diameter on drop breakup frequency. (a) with different turbulent energy dissipation rates, $\sigma = 35$ mN/m; (b) with different interfacial tensions, $\epsilon = 3$ m^{2}s^{-3}

The shading regions in Figure 5 are where the drop breakup frequency can be accurately predicted by the proposed model in this study. It is noted that the breakup model constructed in the present work is based on the binary breakup assumption and the breakup time model adopted in this study is also valid for the breakup process with a small number of fragments generated (generally, no more than 4 fragments). According to our previous study[24], the predicted results of the breakup time are in good agreement with the experimental data when We_L is lower than 0.7 (corresponding to a breakup probability of approximately 0.5). Therefore, we can approximate $We_L = 0.7$ as the criterion, when $We_L < 0.7$, the model can accurately predict the breakup frequency (corresponding to the shaded regions), and when $We_L > 0.7$, the predictive accuracy of the breakup frequency prediction will be limited, and the effect of multiple breakups needs to be quantified. Noteworthy, according to our previous experimental results[8–10,12,21], drop breakups are dominated by the low-fragment breakups (especially, the proportion of the binary breakup is more than 50%, and the breakups with the fragments number lower than 4 take the proportion of more than 90%) under steady-state operating conditions. Thus, the breakup model constructed in this study can be accurately applied to predict the drop breakup frequency in the practical apparatus.

4.5 Influence of the turbulent energy dissipation rate on drop breakup frequency

The turbulent energy dissipation rate is one of the most important parameters affecting the drop breakup frequency. The larger the value of the turbulent energy dissipation rate, the more favorable the energy transfer from the eddies to the drop surface. Figure 6 showed that the drop breakup frequency gradually increases with the increase of the turbulent energy dissipation rate, which is consistent with the above analysis. Moreover, Figure 6a indicates that the breakup frequency versus the turbulent energy dissipation rate varies greatly for different drop diameters. The intersections of the gray straight line with each line in Figure 6a characterize values of drop breakup frequency at $We_L = 0.7$ for each case, and region below the intersection (or the shading region in Figure 6b) represents the applicable scope of the breakup model. Meanwhile, for the upper side of the intersection, the effect of the multiple breakups needs to be considered.

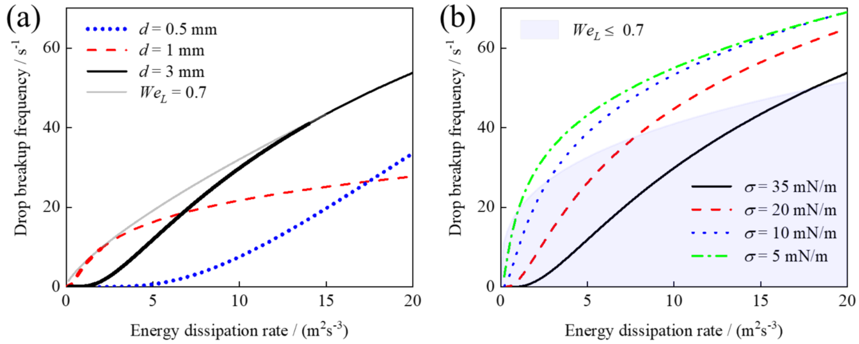

Figure 6 Influence of the turbulent energy dissipation rate on drop breakup frequency. (a) with different drop sizes, $\sigma = 35$ mN/m; (b) with different interfacial tensions, $d = 1$ mm

4.6 Influence of the interfacial tension on drop breakup frequency

Figure 7 shows the drop breakup frequency versus the interfacial tension. It can be seen that drop breakup frequency increases with increasing the interfacial tension at very small interfacial tensions, implying that the effect of the interfacial tension on drop breakup time is more significant compared with the effect on drop breakup probability. As the interfacial tension exceeds a critical value, Figure 7 shows that the breakup frequency decreases with the increase of the interfacial tension, implying that the effect of interfacial tension on drop breakup probability is more significant at this moment. The lower side of the intersection of the gray line with each line in Figure 7a (or the shading region in Figure 7b) also represents the accurate prediction ranges of the breakup model

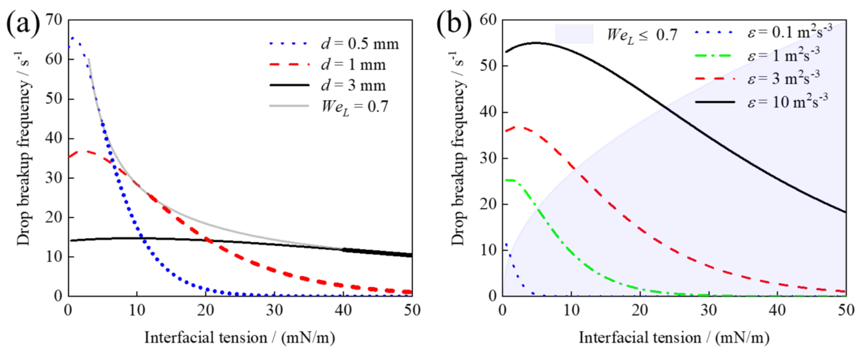

Figure 7 Influence of the interfacial tension on drop breakup frequency. (a) with different drop sizes, $\epsilon = 3$ m^{2}s^{-3}; (b) with different turbulent energy dissipation rates, $d = 1$ mm

4.7 Extension of drop breakup frequency model by considering the effect of drop viscosity

For systems with high viscosity, the effect of drop viscosity on drop breakup behaviors needs to be considered.[10,37] The breakup probability and breakup frequency model constructed in Section 3 is only applicable to describe the breakup behavior of low-viscous drops, thus it is necessary to extend the model by considering the effect of drop viscosity. According to the drop oscillation breakup mechanism, the influence of the drop viscosity is mainly reflected in two aspects:

1) The viscous hamper increases with the increase of drop viscosity, which is manifested by the decrease of

the oscillation amplitude with increasing the viscosity and thus leads to the increase of drop breakup time. The breakup time considering the effect of drop viscosity can be modeled by Eq. in Section 2.

2) The occurrence of the drop breakup should satisfy the energy constraint, in addition to overcoming the increase in the maximum interfacial energy during the breakup process, it is necessary to consider the effect of the increase in the drop viscous energy.

For a droplet with a size of d , the viscous energy it contains can be expressed as:

Where p_{vis} is the viscous energy per unit drop volume and can be calculated by Eq.:

Therefore, the increase in viscous energy during the drop breakup process is expressed as:

Based on the above analysis, the breakup constraint is expressed as Eq.:

Then it is obtained that:

Thus, we obtain the extension of the drop breakup probability model by considering the effect of drop viscosity, as shown in Eq..

Where Oh is the Ohnesorge number, which characterizes the relative magnitude of the viscous and interfacial stresses:

Combined with the breakup time model of Eq. in Section 2, the extended breakup frequency model can be expressed as:

In our previous work[10], we investigated the effect of dispersed phase viscosity on drop breakup frequency and obtained the direct experimental data, which is applied to verify the accuracy of Eq.. Figure 8 shows the comparison of the predicted results with the initial breakup experimental data. Compared with the breakup model of Eq. which is constructed for low-viscous drops, Eq. is in better agreement with the experimental data. Moreover, Figure 8 shows that the drop breakup probability gradually decreases as Oh increases.

Based on the analysis in the above sections, we can conclude that for low-viscous drops, drop breakup probability is mainly determined by the Weber number (We_L), and for intermediate to high viscous drops, the breakup probability is determined by the combined influence of the Weber number (We_L) and the Ohnesorge number (Oh).

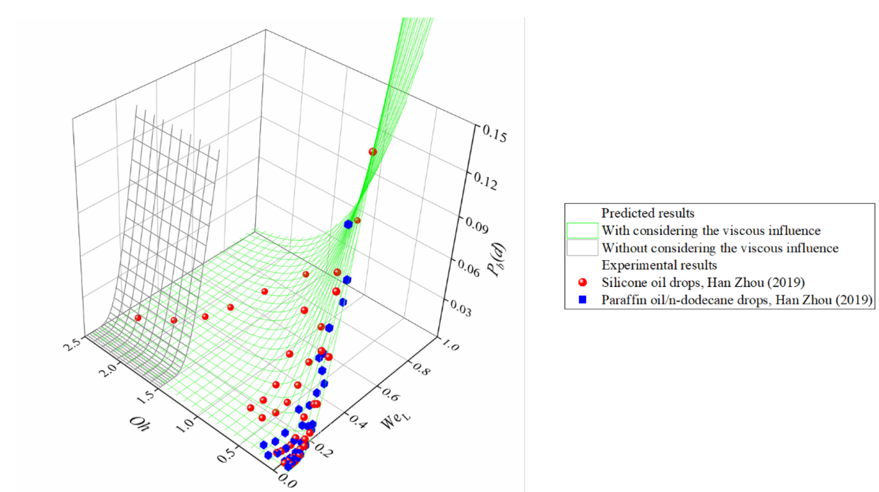

Figure 8 Comparison of the predicted results with the initial breakup experimental data, $c_{vis} = 3.0$

5 Conclusion

The construction of the accurate drop breakup model is crucial for the application of the population balance model and the study of liquid-liquid dispersion characteristics. Based on the drop oscillation breakup mechanism, in the present work, we proposed a theoretical model of drop breakup probability by adopting the three-dimensional Maxwell velocity distribution. The model considers the breakup constraints of both the maximum increase of the interfacial energy and the increase of drop viscous energy during the breakup process. The results show that for low-viscous drops, drop breakup probability is only related to the Weber number (We_L), which characterizes the relative magnitude of the external turbulent stress and the interfacial stress. When the drop viscosity exceeds a certain limit, the effect of drop viscosity needs to be considered and the Ohnesorge number (Oh), which characterizes the relative magnitude of the viscous stress of the drop and the interfacial stress, is introduced.

By combining the theoretical model of drop breakup time constructed in our previous work, the breakup frequency model is obtained based on the statistical description framework. The accuracy and generality of the model were then validated using the direct experimental data. Moreover, we systematically analyzed the effects of the drop diameter, turbulent energy dissipation rate, and interfacial tension on the predicted drop breakup frequency. Results showed that the breakup frequency increases monotonically with increasing turbulent energy dissipation rate. Differently, with the increase of the drop diameter or the interfacial tension, the breakup frequency firstly increases and then decreases. The reason for the above results is related to the coupling effect of the parameters on the breakup time and breakup probability. The results of this study are applicable to predict drop breakup frequency for drop breakups with low fragments generated. Combined with the results in our previous studies, the proportion of the breakups with the number of fragments lower than 4 accounts for more than 90% under steady-state operating conditions. Therefore, the model constructed in the present work can directly serve for the quantitative characterization of drop breakup behaviors under steady-state operating conditions.

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Notation

c_f coefficient of surface area,

d drop diameter, m

D the diameter of the impeller, m

$E(k)$ the energy spectrum of turbulence, m^3/s^2

the energy constraint, J

k the wavenumber of the turbulence, 1/m

L the maximum scale of the local turbulent structure, m

N the rotating speed of the impeller, 1/s

$N(d)$ the number of mother drops

Oh the Ohnesorge number

p_{os} the average oscillation kinetic energy per unit drop volume, J/m^3

p_t the turbulent stress, Pa

p_{vis} the viscous energy per unit drop volume, J/m^3

P_b (d) drop breakup probability

t_b drop breakup time, s

the critical oscillation velocity, m/s

the root-mean-square velocity of the surface oscillation, m/s

We_L the Weber number

Greek letter

drop breakup frequency, 1/s

the interfacial energy increase before and after drop breakup, J

the maximum interfacial energy increase during the drop breakup, J

the increase in viscous energy during the drop breakup process, J

the number of the broken drops

turbulent energy dissipation rate, m^2/s^3

eddy size, m

μ_δ dispersed phase viscosity, Pa s

continuous phase density, kg/m^3

dispersed phase density, kg/m^3

σ interfacial tension, N/m

dispersed phase volume fraction

collision frequency between eddies and drops, 1/(m^{4}s)

the size ratio between an eddy and a droplet

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