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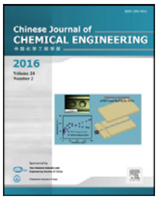

Analysis of drop deformation dynamics in turbulent flow

Stephanie Nachtigall *, Daniel Zedel, Matthias Kraume

Chair of Chemical and Process Engineering, Technische Universität Berlin, Berlin 10587, Germany

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ABSTRACT

Drop breakage and coalescence influence the particle formation in liquid–liquid dispersions. In order to reduce the influencing factors of the whole dispersion process, single drops where coalescence processes can be neglected were analyzed in this work. Drops passing the turbulent vicinity of a single stirrer blade were investigated by high-speed imaging. In order to gain a statistically relevant amount of drops passing the area of interest and corresponding breakage events, at least 1600 droplets were considered for each parameter set of this work. A specially developed fully automatic image analysis based on Matlab® was used for the evaluation of the resulting high amount of image data. This allowed the elimination of the time-consuming manual analysis and furthermore, allowed the objective evaluation of the drops' behavior. Different deformation parameters were considered in order to describe the drop deformation dynamics properly. Regarding the ratio of both main particle axes (θ_{axes}), which was therefore approximated through an ellipse, allowed the determination of very small deviations from the spherical shape. The perimeter of the particle (θ_{peri}) was used for the description of highly deformed shapes. In this work the results of a higher viscosity paraffin oil ($\eta_d = 127$ mPa·s) and a low viscosity solvent (petroleum, $\eta_d = 1.7$ mPa·s) are presented with and without the addition of SDS to the continuous water phase. All results show that the experimentally determined oscillation but also deformation times underlie a wide spreading. Drop deformations significantly increased not only with increasing droplet viscosity, but also with decreasing interfacial tension. Highly deformed particles of one droplet species were more likely to break than more or less spherical particles. As droplet fragmentation results from a variety of different macro-scale deformed particles, it is not assumed that a critical deformation value must be reached for the fragmentation process to occur. Especially for highly deformed particles thin particle filaments are assumed to induce the breakage process and, therefore, be responsible for the separation of drops.

1. Introduction

Turbulent dispersions of two immiscible liquids are part of various applications for example in chemical, food or pharmaceutical industries. Drop breakage and drop coalescence thereby determine the resulting drop size distribution (DSD). Consequently, both counteracting phenomena influence the interfacial area for energy or mass transfer between both phases and the resulting process efficiency. The great industrial importance goes along with comprehensive scientific research activities over the last decades. In dispersed systems drop breakage and coalescence not only occur simultaneously but also influence each other. Therefore, they must be investigated separately to gain a more detailed understanding of different influencing factors and the physical basics of each process. In order to reduce the high amount of influencing factors of the whole process it is advantageous to analyze single droplets. No interactions between droplets can occur and they do not influence the flow structure, which was shown for example by Galinat *et al.* [1].

In this work drop breakage in turbulent flow is analyzed. The main focus lies on the drop deformation dynamics, which was found to be not negligible (for example [2,3]). For example the breakage time still is discussed contradictorily [4] and, therefore, open for further improvements.

The basis and current state of the art of turbulent particle breakup will be described below. Furthermore, an overview of existing single fluid particle breakage investigations will be given, which shows the variety of different approaches existing in literature. Corresponding literature references, which focus on the description and experimental analysis of droplet breakup in laminar flow (for example [2,5,6]) also exist. As this work is dedicated to drop breakage in turbulent flow, this will however not be further described here in more detail.

1.1. Turbulent particle breakup

In the pioneer works of Kolmogorov [7] and Hinze [8] particle breakup due to turbulent velocity and corresponding pressure fluctuations is described based on Kolmogorov's theory of local isotropic turbulence from 1941 [9,10].

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* Corresponding author.
E-mail address: Stephanie.Nachtigall@tu-berlin.de (S. Nachtigall).

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It is widely accepted that breakage occurs when the external forces and stresses exerted on particles by the continuous phase exceed the internal forces and stresses which hold the particle together. Thereby, turbulent velocity fluctuations over the distance of the particle diameter are suggested to be responsible for breakup [8,11]. In general, larger turbulent structures were found to be responsible only for the transportation of the particles, while smaller eddies, not containing enough energy for breakage, only deform particles [12]. According to Hinze [8], the deforming stresses due to turbulent pressure fluctuations can be approximated by $\tau_{\mathrm{def}} \approx \rho_{\mathrm{c}}\overline{u'(d_{\mathrm{p}})^2}$. Applying Kolmogorov's theory of local isotropic turbulence [9,10] and assuming turbulent structures in the inertial subrange to be responsible for the breakup lead to:

$$\tau_{\mathrm{def}} = 2\rho_{\mathrm{c}}(\varepsilon d_{\mathrm{p}})^{2/3}. \tag{1}$$

The deforming stresses τ_{def} induce a flow inside the particle which leads to a stabilizing viscous stress τ_η. The flow velocity can, thereby, be approximated by $\sqrt{\tau_{\mathrm{def}}/\rho_{\mathrm{d}}}$. Additionally, the particle is stabilized due to the interfacial tension. Hinze [8] described the stabilizing stresses as follows:

$$\tau_\eta \approx \frac{\eta_{\mathrm{d}}}{d_{\mathrm{p}}}\sqrt{\frac{\tau_{\mathrm{def}}}{\rho_{\mathrm{d}}}}, \quad \tau_\gamma \approx \frac{\gamma}{d_{\mathrm{p}}}. \tag{2}$$

The resulting three stresses, which determine turbulent particle breakup are characterized by two dimensionless numbers. Hinze [8] chose one which he allocated to the group of the Weber numbers and one of the viscosity group, which is also commonly known as the Ohnesorge number:

$$We = \tau_{\mathrm{def}} d_{\mathrm{p}}/\gamma, \quad Oh = \eta_{\mathrm{d}}/\sqrt{d_{\mathrm{p}}\rho_{\mathrm{d}}\,\gamma}. \tag{3}$$

While the Ohnesorge number describes the stabilizing effect of the viscosity in comparison to the stabilizing effect of the interfacial tension, the Weber number describes the deforming stress due to a dynamic pressure in comparison to the stabilizing effect of the interfacial tension. The Weber number must exceed a critical value for fluid particle breakage to occur, which determines the maximum stable particle size. As the whole process can be described by these dimensionless numbers, the corresponding critical Weber number must depend on the Ohnesorge number [8]:

$$We_{\mathrm{crit}} \propto 1 + f(Oh). \tag{4}$$

In the past a lot of results of different mixing applications were not explainable only on basis of the described theory. Consequently, extensions and modifications of the concept can be found in literature. Some authors assumed velocity gradients in the mean flow to be mainly responsible for breakage and their neglecting not being valid. Depending on the process parameters, Kumar *et al.* [13,14] suggested that besides the turbulent fluctuations other breakage mechanisms occur in a stirred tank. In accordance to Wichterle [15] they expected the shear layer on the blade to be mainly responsible for drop breakage. Additionally, the elongation flow behind the stirrer was suggested to influence drop breakage, which was also found for example by [16,17]. Ali *et al.* [18] and Chang *et al.* [19] described effects of the mean velocity depending on the Reynolds number and the dispersed phase properties.

When turbulent structures were supposed to be mainly responsible for the breakage process, some authors assumed eddies which are smaller than the droplets being responsible for breakage. This was for example concluded by Nambiar *et al.* [20] from their assumption of breakages into small droplets, whose surface energies correspond to the energy of smaller eddies. Additionally, different authors (for example [21,22]) suggested particle oscillations to be mainly influenced by smaller eddies. Instead, the observation of macro-scale deformations led other authors [23] to the conclusion that much larger turbulent structures are responsible for fluid particle breakage. As Andersson and Andersson [3,24] observed drop oscillations, as well as macro-scale deformations, they consequently stated that all kinds of eddy sizes can be involved in the breakage process.

Sprow [25] already postulated in 1967 that depending on the droplet size processes in the inertial or in the viscous subrange can become responsible for drop breakage. Therefore, the Kolmogorov length scale $\lambda = (\nu^3/\varepsilon)^{1/4}$ [9,10] which is a measure of the smallest occurring eddies in the systems, is considered. Droplets much smaller than this scale are expected to be influenced by processes in the viscous subrange (see for example [26]), where viscous (for example. [26,27]) or inertial forces (for example [28]) are assumed to be responsible for breakage.

Risso [2] criticized the Weber concept and stated that the breakup of particles in turbulent flow cannot be described by a simple force balance between deforming and stabilizing effects. Instead he expected particle dynamics to mainly influence the behavior prior to breakage. Fluid particles undergo deformation and relaxation processes. They are determined by the stabilizing interfacial tension and dispersed phase viscosity, as well as deforming forces of one or more successive eddies. While some authors described fluid particles through a linear damped oscillator [2,21,22,29], Kumar *et al.* [30] considered drop oscillations in a stirred tank through a two-stage model. Droplets thereby change position between the stirrer vicinity where deformations and breakages occur and outside this area where relaxation processes dominate. Consequently, fluid particle breakup can result from more successive eddies through resonant effects. In order to describe macro-scale deformations prior to breakage Andersson and Andersson [3] defined two criteria which must be satisfied for the particle breakup to occur. Besides a simple stress criterion that must be fulfilled for the achievement of the critical particle deformation, an energy criterion was formulated in order to describe the duration of the deformation process.

It has to be mentioned here that the deformation dynamics is at least partly considered through the stabilizing effect of the dispersed phase viscosity which mainly influences the deformation time (for example [31]). Different approaches exist for the prediction of maximum stable drop diameters considering the dispersed phase viscosity within the framework of the critical Weber concept. Besides more or less simple summations of both stabilizing forces (for example [32–34]), the droplets were described by a Kelvin–Voigt model [35–37], which can also be regarded as an overdamped linear oscillator [38].

1.2. Experimental investigations of single fluid particle breakup

Despite comprehensive experimental research activities, a generally accepted and valid description of turbulent particle breakup and corresponding mathematical models are still missing. Instead, different and sometimes even contradictory results were concluded from experimental investigations. For example Sleicher [39] and Swarz and Kessler [40] investigated drop breakages in a turbulent pipe flow. Even though they used the same dispersed phase they came to inconsistent results. Sleicher [39] observed drop breakages mainly near the pipe wall, where gradients of the mean flow dominate. Instead, Swarz and Kessler [40] found breakage to occur in the turbulent core of the flow, which accordingly led to the description of different breakage mechanisms.

One reason might be the restricted transferability of the results from one set-up to another [2]. Not only the flow properties, but also the experimental conditions can differ significantly in two different set-ups. The widespread application of tap water, instead of deionized water (for example [21,22]), might be one problem here. Also, missing specifications of the experimental parameters, like the investigated initial particle size (for example [41]), restrict the comparability. Furthermore, breakage characteristics were defined in different ways. The critical Weber number and the corresponding maximum stable diameter are given as examples here. They were derived from the events where no