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Development of an improved breakage kernel for high dispersed viscosity phase emulsification

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ABSTRACT

Modelling of droplet breakage in emulsification is traditionally governed by a range of empirically adjustable parameters in the appropriate breakage rate and daughter size distribution models. The development of a purely phenomenological modelling approach is desirable to obtain more universally applicable breakage models and reduce the need for extensive experiential-based parameter identification. A modification adapting the phenomenological breakage modelling framework proposed by Luo and Svendsen (1996) to high-viscosity dispersed phases is proposed in this work. The performance of the new model was confirmed by comparison to experimental data obtained from the emulsification of silicone oils with four different viscosities. The new model was compared to two recent traditional breakage rate models and found to provide improved results, without the need for empirically adjusted parameters.

1. Introduction

1.1. Motivations

Using Population Balance Equations (PBE) for the modelling of emulsification systems has evolved considerably over the last decade due to the availability of increasingly sophisticated drop size analysis techniques, such as in-situ video probes with automated image treatment algorithms (Becker et al., 2011; Khalil et al., 2010) or Fixed Beam Reflectance Measurement probes (FBRM) (Boxall et al., 2012) as well as ever increasing computational capabilities. These improvements have allowed the development of better phenomenological models linking the complex droplet-scale effects and interactions influencing the breakage and coagulation mechanisms to the observable evolution of the Droplet Size Distributions (DSD). The breakup of viscous droplets in a turbulent regime is a particularly challenging area of research, as a number of different effects, which cannot be easily quantified, need to be considered. Such effects concern most notably (a) the drop–eddy interactions, the effect of which depends strongly on the relative size and which are by their very nature governed by a probabilistic distribution of available eddy and droplet sizes, (b) the elongation, deformation, and relaxation undergone by a droplet inside a complex flow field before breakage (as for example investigated in single-drop experiments by Maaß and Kraume, 2012), (c) dynamic surfactant effects after a sudden surface area increase due to breakage, and (d) the influence of dispersed phase concentration on turbulence structures and interactions of adjacent droplets (Tcholakova et al., 2011). Turbulent breakage in highly concentrated ($\varphi > 30\%$) emulsions are a particularly challenging subject because the dispersion ceases to behave like a single-phase Newtonian fluid and the estimation of turbulent eddie dissipation in the diminishing volume of continuous phase becomes increasingly difficult.

Experimental data clearly shows that the dispersed phase viscosity has a significant effect on the mean and maximum drop sizes, as well as the shape of the DSD. Andersson and Andersson (2006) show in single-drop experiments that the highly viscous droplets tend to deform more dramatically before breakage, leading to a more uneven daughter size distribution as well as longer breakup times. The focus of this study is to address this point. Dilute emulsions of silicone oils in water in the presence of excess of fast-acting emulsifier (Tween 20) are studied in order to minimize the effects of (c) and (d) as well as coagulation. This allows for the effects of (a) and (b) to be studied in isolation, in order to develop an accurate breakage model taking drop–eddy interactions and viscous deformations into account. Even though dispersed phase concentration are kept at a moderate level, some corrections will be made to account for its effect on the overall breakage process; these corrections are briefly outlined in

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Section 1.3. The objective of this paper is to develop a break-up kernel that takes into account the dispersed phase viscosity and concentration in stirred systems with low dispersed phase concentrations ($\varphi < 10\%$).

This paper is structured as follows: a brief introduction to the recent developments concerning the breakage rate and daughter size distribution kernels, is given in the remainder of this section. The framework proposed by Luo and Svendsen (1996) as well as the most recent improvements with respect to this model's applicability to liquid–liquid systems is discussed in detail in Section 2. A new modification to this framework, taking viscous deformations in a breaking droplet into account, proposed in this work is proposed and discussed in Section 3. This is followed by an evaluation of the new model and comparison to experimental data in Section 4, and a conclusion is given in Section 5.

1.2. The population balance equation

The population balance equation for a homogeneous system with a continuous number density distribution (n) along one internal coordinate (x), undergoing only breakage is given in Eq. (1); see Ramkrishna (2000) for details.

$$\frac{dn(x,t)}{dt} = \int b(x,v)S(x)n(x,t)dv - S(x)n(x,t) \tag{1}$$

The breakage frequency kernel and daughter size distribution are denoted by $S(x)$ and $b(x,v)$ respectively. The main models proposed for these terms are given in the following section.

1.3. Energy dissipation rate correction for dispersed phase concentration

The mean energy dissipation rate is generally calculated assuming a single-phase fluid with the properties of the continuous phase. This is reasonable for low dispersed phase concentrations (i.e. $\varphi < 1\%$), however, the effect of the dispersed phase becomes more pronounced for more concentrated emulsions and must therefore be taken into account. This can be done by modifying the energy dissipation rate to obtain the energy dissipation effectively seen by the emulsion droplets, ε_{eff}. A simple correlation to account for the damping effect of high dispersed phase hold-up was be obtained by Coulaloglou and Tavlarides (1977):

$$\varepsilon_{eff} = \frac{\varepsilon}{(1+\varphi)^3} \tag{2}$$

A more complex correction factor, based on expressions for the emulsion viscosity and density was derived by Alopaeus et al. (2002):

$$\varepsilon_{eff} = \varepsilon\left(\frac{(1-\varphi)(\varphi(\rho_d/\rho_c)+(1-\varphi))}{1+1.5\varphi(\mu_d/(\mu_d+\mu_c))}\right) \tag{3}$$

These correction factors can either be explicitly included in the breakage models or used modify ε before executing the calculations. Unless mentioned otherwise, the correction presented in Eq. (2) is used in conjunction with all of the breakage rate kernels presented in the subsequent sections (the subscript "*eff*" is omitted for clarity of the equations) of this work, in order to account for the effect of dispersed phase concentration.

1.4. Daughter size distributions

Binary breakage is assumed implicitly in the daughter size distribution, $b(V_0,V_1)$ for the purpose of the numerical simulations presented in this work; an appropriate factor must be inserted into Eq. (1) when multiple daughter fragments are being considered (Ramkrishna, 2000). A number of different daughter size distribution functions have been proposed in the literature, the major part of which assume binary breakage; see Liao and Lucas (2009) for a detailed review. This assumption requires the number distribution function, $b(V_0,V_1)$, to be symmetrical about equal sized breakage in addition to the requirement of mass/volume conservation. While distributions for multiple fragments have been used by some authors (Kotoulas and Kiparissides, 2006; Raikar et al., 2010); precise information about the nature of multiple breakage is difficult to predict and therefore it is most common to model the formation of multiple daughter droplets as a series of subsequent binary breakage events. The most commonly used daughter distributions are based on statistical models, such as uniform (Narsimhan et al., 1979), Gaussian (Coulaloglou and Tavlarides, 1977) or beta distributions (Konno et al., 1983). However, attempts have been made to develop phenomenological models; most notably by Martinez-Bazan et al. (1999b, 2010), as well as the U- or M-shaped distributions based on the Luo and Svendsen (1996) framework, which will be discussed in detail in Section 3.

1.5. Breakage rate kernels

A large range of breakage rate models for turbulent liquid–liquid dispersions can be found in the literature; more comprehensive reviews than the brief overview of the state of the art presented here can be found in Becker et al. (2011), Maaß et al. (2012), Liao and Lucas (2009), and Maaß and Kraume (2012). The mechanistic formulation of the breakage rate function in terms of breakage time (t_b) and fraction of breaking drops, presented by Coulaloglou and Tavlarides (1977) still forms the basis of most breakage models in use today. It is based on expressions for the breakage time (t_b) and the ratio of droplets breaking (N_b) to total droplets (N_0). This model depends on the mean energy dissipation rate (ε), the surface tension (σ), and the dispersed phase density (ρ_d); it does however not take the dispersed phase viscosity (or the viscosity ratio between the two phases) into account. It depends on two empirically determined tuning parameters (C_1 and C_2).

$$S(d_i) = \left(\frac{1}{t_b}\right)\left(\frac{N_b}{N_0}\right) = C_1\frac{\varepsilon^{1/3}}{d_i^{2/3}}\exp\left[\frac{C_2\sigma}{\rho_d\varepsilon^{2/3}d_i^{5/3}}\right] \tag{4}$$

Most commonly used breakage models assume that breakage takes place in the inertial sub-range (Kolmogorov, 1941) and are therefore based on the turbulent energy transferred to a droplet being larger than a critical value. This energy is typically expressed in terms of one or more of the following: (1) turbulent kinetic energy available to the droplet (Coulaloglou and Tavlarides, 1977; Baldyga and Bourne, 1999; Bałdyga et al., 2001), (2) turbulent kinetic energy (Luo and Svendsen, 1996; Martínez-Bazán et al., 1999b; Tsouris and Tavlarides, 1994), (3) turbulent fluctuations around the droplet (Alopaeus et al., 2002), or (4) inertial forces of the bombarding eddy (Lehr et al., 2002). All of these models take their starting point from physical considerations; however, other attempts have been made to exploit the self-similar behaviour of the breakage rate function to derive breakage rate models (Narsimhan et al., 1984; Sathyagal et al., 1996).

The original model by Coulaloglou and Tavlarides (1977) Eq. (4), makes the assumption that viscous deformations of the breaking droplet are negligible and surface energy alone determines the