



Extent and mechanism of coalescence in rotor-stator mixer food-emulsion emulsification



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ABSTRACT

Food-emulsions often have high volume fractions of dispersed phase and are thus expected to show coalescence during emulsification, however, food-emulsion coalescence is difficult to measure in homogenizer equipment. This study experimentally estimates the rates of fragmentation and coalescence in a high viscosity and high volume fraction model emulsion subjected to pilot-scale rotor-stator mixing in order to quantify the relative effect of coalescence and discuss the mechanism of coalescence during batch processing of high-fat emulsion foods. Rate constants of both processes are estimated using a previously suggested method relying on parameter fitting from the dynamic evolution of the total number of emulsion drops (Hounslow and Ni, 2004). The results show substantial coalescence taking place. Scaling of rates with respect to rotor tip speed suggests coalescence and fragmentation controlled by a turbulent viscous mechanism.

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1. Introduction

Emulsification can be described as a combination of drop fragmentation and coalescence (Håkansson et al., 2009; McClements, 2005; Walstra, 2005). Industrial processing of food emulsions (Santana et al., 2013) is designed to favor fragmentation, reduce coalescence and thus obtain small drops with narrow drop size distributions at minimal energy input and processing time. Studies on understanding emulsification therefore often focus on the effect of fragmentation. High intensity emulsification fragmentation is often classified in three broad mechanistic classes (Walstra, 2005): Turbulent inertial (TI) fragmentation brought about by interactions between drops and turbulent eddies smaller than the drop (Hinze, 1955), turbulent viscous (TV) fragmentation from shearing of drops by eddies larger than the drop (Hinze, 1955), and in case of laminar flow, a laminar viscous (LV) shear mechanism (Grace, 1982). For each mechanism, a basic scaling exists between resulting drop diameter (d) and emulsion characteristics, such as disperse and continuous phase viscosities (μ_D , μ_C) densities, (ρ_D , ρ_C), interfacial tension (σ) and the dissipation rate of turbulent kinetic energy (ϵ) (Hinze, 1955; Walstra, 2005; Zhang et al., 2012),

$$d \propto \rho_C^{-3/5} \epsilon^{-2/5} \sigma^{3/5} \quad (\text{TI}) \quad (1)$$

$$d \propto \mu_C^{-1/2} \rho_C^{-1/2} \epsilon^{-1/2} \sigma \quad (\text{TV}) \quad (2)$$

or in the laminar case, the velocity gradient (G) (Grace, 1982; Walstra, 2005)

$$d \propto c \left(\frac{\mu_D}{\mu_C} \right) \mu_C^{-1} G^{-1} \sigma \quad (\text{LV}). \quad (3)$$

where c in Eq. (3) is a concentration ratio dependent constant.

Many food emulsions (e.g. mayonnaises, cake batters, creamy sauces and dressings) have high volume fraction of disperse phase and consequently high emulsion viscosity, and are often processed with rotor-stator mixers where the mean effective dissipation rate of turbulent kinetic energy is assumed proportional to the cube of rotor frequency (\dot{N}) or tip speed (U) (Zhang et al., 2012)

$$\epsilon \propto \dot{N}^3 \propto U^3 \quad (4a)$$

and laminar shear rate is proportional to rotor tip speed

$$G \propto \dot{N} \propto U \quad (4b)$$

Combinations of Eqs. (1)–(3) with 4 suggest different scaling

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between measurables such as rotor frequency and drop diameters, and hence, comparisons between empirical and theoretical scaling have been used extensively to determine dominant mechanisms of fragmentation for emulsions in similar devices (Rueger and Calabrese, 2013a, 2013b; Tcholakova et al., 2011). Theoretically, d in Eqs. (1)–(3) should be interpreted as the maximum stable drop diameter (Hinze, 1955); however, it is often replaced by a mean drop diameter (e.g. volume or surface weighted average, d_{43} and d_{32} respectively) in applications, since these are arguably proportional to each other (Rueger and Calabrese, 2013a), at least when disperse phase viscosity is low (c.f. Becker et al., 2013).

This methodology for finding dominant regimes assumes that coalescence is sufficiently low not to influence the final drop size. However, experimental measurements with different methods show substantial coalescence during emulsification (Howarth, 1967; Lobo et al., 2002; Niknafs et al., 2011; Taisne et al., 1996), especially in high volume fraction systems (Mohan and Narsimhan, 1997; Niknafs et al., 2011). Emulsification of complex high volume fraction food emulsions such as mayonnaises, spreads and creamy sauces are therefore expected to be influenced by coalescence, but the extent and impact in industrial conditions is still largely unknown.

Mechanistic understanding –for understanding and optimizing emulsification and equipment – thus requires measuring rates of the underlying processes of fragmentation and coalescence, rather than the combined result in terms of resulting drop size distributions. Several methods for estimating rates of fragmentation (e.g. Becker et al., 2014; Vankova et al., 2007) and coalescence (Howarth, 1967; Karbaschi et al., 2014; Lobo et al., 2002; Miller et al., 1963; Mohan and Narsimhan, 1997; Niknafs et al., 2011; Taisne et al., 1996) have been suggested; however, only the reflectivity technique (Howarth, 1967; Niknafs et al., 2011) and the moment evolution method (Hounslow and Ni, 2004) allow the determination of both processes using the same technique. Of these two, the latter has the advantage of allowing estimations from offline measurements in pilot- and production scale (c.f. Håkansson and Hounslow, 2013) and of allowing both processes to be quantified in one experiment.

Several theoretical models for rates of coalescence and fragmentation kernels have been offered; comprehensive reviews on both fragmentation (Liao and Lucas, 2009) and coalescence kernels (Liao and Lucas, 2010) are available elsewhere. Whereas the number of proposed fragmentation rate models is large (Liao and Lucas, 2009) and growing (e.g. Becker et al., 2014; Maındarkar et al., 2015; Raikar et al., 2010), coalescence rate expressions of early origin (e.g. Delichatsios and Probst, 1975; Saffman and Turner, 1956; von Smoluchowski, 1916) are still used extensively in literature.

The objective of this study is to apply the moment evolution rate extraction method to a pilot scale rotor-stator emulsification system with high dispersed phase volume fraction and emulsion viscosity comparable to a complex emulsion food such as a mayonnaise or a creamy sauce (Pons et al., 1994; Singla et al., 2013) in order to, first, estimate the influence of coalescence on the emulsification process and, secondly, by investigating the scaling of fragmentation and coalescence rates discuss implications on dominant mechanism of coalescence and fragmentation in emulsification of high disperse phase volume fraction food emulsions.

2. Theory and calculations

The moment evolution method (Hounslow and Ni, 2004; Håkansson and Hounslow, 2013) estimates the rates of fragmentation and coalescence by fitting the experimental evolution of moments of the drop size distribution to theoretical models. Assuming that the fragmentation rate (g) is of first order with

regards to drop volume (v)

$$g(v) = g_0 \cdot v, \quad (5a)$$

that the coalescence rate (β) can be approximated by a sum kernel

$$\beta(v_1, v_2) = \beta_0 \cdot (v_1 + v_2) \quad (5b)$$

with constants g_0 and β_0 , and that each breakup gives rise to m drops on average, the per unit volume number of emulsion drops (N) is described by (Hounslow and Ni, 2004)

$$\frac{dN}{dt} = -\beta_0 \varphi_D N + g_0(m-1)\varphi_D, \quad N(0) = N_0 \quad (6)$$

which is solved by

$$N(t) = N_0 \exp(-t\varphi_D\beta_0) + \frac{g_0(m-1)}{\beta_0} (1 - \exp(-t\varphi_D\beta_0)) \quad (7)$$

where φ_D is the volume fraction of disperse phase. Whereas the simultaneous determination of coalescence and fragmentation from size distributions is generally ill-posed (Ramkrishna, 2000, pp. 222), the specific form of Eq. (7), with a time-scale depending only on coalescence rate, makes it suitable for determination of both rates (Hounslow and Ni, 2004).

For a system without coalescence, the corresponding expressions are

$$\frac{dN}{dt} = g_0(m-1)\varphi_D, \quad N(0) = N_0 \quad (8)$$

and

$$N(t) = N_0 + g_0(m-1)\varphi_D t. \quad (9)$$

The number of drops at time t can be obtained by combining the volume fraction of disperse phase and the volumetric mean drop diameter (d_{43}):

$$N(t) = \frac{6\varphi_D}{\pi d_{43}^3(t)} \quad (10)$$

Thus, rates of coalescence (β_0) and fragmentation ($m \cdot g_0$) for a system can be obtained by fitting measured d_{43} over time to Eq. (7) or (9) and (10). The relative fit to models with fragmentation only (Eq. (9)) and fragmentation with coalescence (Eq. (7)) could be used to determine if substantial coalescence occurs.

It should be noted that the method does not allow for independent estimates of fragmentation rate, g_0 , and the number of fragments per breakup, m . Following previous applications of this method (Håkansson and Hounslow, 2013; Hounslow and Ni, 2004), it is assumed that an average of four fragments are formed per fragmentation event ($m = 4$). There is no scientific consensus on the true number of fragments formed per breakup during emulsification under different conditions, and the experimental technique did not allow for verification of the assumption, however, $m = 4$ is of order of magnitude similar to previous studies (c.f. Liao and Lucas, 2009 and references therein). Moreover, as long as the number of fragment does not depend on the rotor tip speed or continuous phase viscosity, it will not influence scaling behavior of rates. Consequently, scaling instead of absolute rates are used for drawing conclusions on dominating mechanisms in this study.

Due to the inhomogeneous distribution of velocity gradients and turbulence in the mixer (Mortensen et al., 2011; Utomo et al., 2008), inhomogeneous fragmentation and coalescence rates are expected. However, the moment evolution method is based on

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