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On the effect of dispersed phase viscosity and mean residence time on the droplet size distribution for high-shear mixers.

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HIGHLIGHTS

- Emulsification of Silicon oils with viscosities varying 3 orders of magnitude.
- Three droplet break-up mechanism regions found.
- Processing parameter effect on the size of each of the two types of daughter drops.
- Bimodal Drop Size Distribution analysed with two generalized gamma functions.

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ABSTRACT

Properties of emulsified product such as stability, rheology and interfacial area dependent on their microstructure, specially their mean droplet size and droplet size distribution. Mechanistic models in literature focus on predicting the maximum droplet diameter or Sauter mean diameter but not in their size distribution. The effect of viscosity (9.58–295 mPa s), mean residence time and stirring speed (50–150 s^{-1}) have been investigated using an in-line laboratory scale rotor-stator and dilute (negligible coalescence) coarse emulsions with seven Silicon Oils of different viscosity.

Low viscous oils produced monomodal distributions whereas the ones for intermediate and high viscous oils were bimodal. The mode or modes of the distributions were used for the modelling of the large and small daughter droplet sizes. The droplet size modelling had a mean absolute error (MAE) of 8%. To model the distributions by volume two Generalized Gamma functions were used and fitted using the least absolute error. The distributions were reasonably well-described while predicting the Sauter mean diameter of both mono and bimodal distributions with a MAE of 13.8%.

1. Introduction

High-shear mixers are able to create small droplets with large interfacial areas due to their localised energy dissipation rates, high rotor speeds and the narrow spacing between the rotor and the stator. These mixers are widely used to produce cosmetics, foods, paints, pharmaceuticals and chemical (Zhang et al., 2012; Atiemo-Obeng and Calabrese, 2003), but despite their wide applicability there is almost no fundamental understanding on these devices (Atiemo-Obeng and Calabrese, 2003). The two main types of high-shear mixers used are the radial discharge batch and the in-line rotor-stators. In-line rotor-stators allow for continuous processing and offer versatility to change from one product formulation to another using the same equipment by valve switching.

The droplet size distribution (DSD) of an emulsion affects its stability (Ma et al., 2005), rheology (Derkach, 2009) and absorption in drug delivery systems (Ma et al., 2010). For example, narrow DSDs are less susceptible to coalescence and Ostwald ripening; therefore personal care products with broad DSD are stabilized by large amounts of surfactants which may cause irritation, skin drying and allergic reactions (Nazir et al., 2013). The rheology of emulsions depends on the droplet-droplet interactions and droplet deformability among other parameters, which are a function of viscosity (both phases), volume fraction, mean droplet size and their DSD (Derkach, 2009); this is important in products such as paint (Watson and Mackley, 2002).

In this study we deal with dilute systems; for these systems drop coalescence is considered negligible and drop breakage can be isolated for its study. Mechanistic models assume that

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Nomenclature

$\dot{Q}$	volumetric flow rate [$m^3\ s^{-1}$]
$\bar{d}_{30}$	volume arithmetic mean [µm]
$\bar{d}_{32}$	Sauter mean diameter [µm]
$\bar{t}_{res}$	mean residence time [s]
A_i	*i*th fitting constant [–]
$C_{L,j}$	fitting constant for large daughter droplet correlation for variable *j* [–]
$C_{s,j}$	fitting constant for small daughter droplet for variable *j* [–]
CI	confidence interval [–]
D	diameter of the impeller [m]
d_i	diameter of the *i*th droplet [µm]
d_{max}	maximum droplet diameter [µm]
E	energy density [J kg^{-1}]
$f_v(d_i)$	frequency by volume of the droplets of the *i*th diameter [–]
MAE	mean absolute error [%]
Mo	mode [µm]
Mo_L	mode of the large daughter droplets [µm]
Mo_s	mode of the small daughter droplets [µm]
N	stirring speed [s^{-1}]
n	number of passes [–]
n_{ri}	refractive index [–]
P	power draw [W]
$P_n(d_i)$	probability by number of droplets of *i*th size [%]
$P_v(d_i)$	probability by volume of droplets of *i*th size [%]
$P_{v,L}(d_i)$	probability of large daughter droplets of *i*th size [%]
$P_{v,s}(d_i)$	probability of small daughter droplets of *i*th size [%]
$P_{v,T}(d_i)$	total probability of droplets of *i*th size [%]
pn	pump number [–]
R^2	coefficient of determination [–]
s	specific gravity [–]
V	swept volume [m^3]

Greek symbols

α	parameter in the Fréchet probability density function [–]
β	parameter in the Fréchet probability density function [–]
η	Kolmogorov length scale [m]
κ	broadness parameter in the Generalized Gamma distribution [–]
λ	scale parameter in the Generalized Gamma distribution [–]
μ_c	viscosity of the continuous phase [Pa s]
μ_d	viscosity of the dispersed phase [Pa s]
$\bar{\varepsilon}$	mean energy dissipation rate per unit mass of fluid [W kg^{-1}]
ϕ_L	volume fraction of the large daughter droplets [–]
ϕ_s	volume fraction of the small daughter droplets [–]
ρ_c	density of the continuous phase [kg m^{-1}]
ρ_d	density of the dispersed phase [kg m^{-1}]
σ	interfacial tension [N m]
σ_d	standard deviation of the normal distribution [µm]
$\sigma_{\log(d)}$	standard deviation of the log-normal distribution [µm]
τ	skewness parameter in the Generalized Gamma distribution [–]
ε	energy dissipation rate per unit mass of fluid [W kg^{-1}]
ε_m	maximum energy dissipation rate per unit mass of fluid [W kg^{-1}]

Dimensionless numbers

Po	Power number $PN^{-3}D^{-5}\rho^{-1}$

Abbreviations

DSD	droplet size distribution
GGd	Generalized Gamma distribution
SLES	Sodium Laureth Sulphate

equilibrium for these systems is reached when all of the drops are smaller than a maximum stable drop size d_{max} (Leng and Calabrese, 2003). A linear relationship betwen d_{max} and the Sauter mean diameter $\bar{d}_{32}$ was proposed by Shinnar (1961) and has been used by many authors

$$\bar{d}_{32} = A_1 d_{max} \tag{1}$$

The Sauter mean diameter is one of the most important measures of central tendency used in emulsification technology because it is inversely proportional to the interfacial area of a given distribution. The previous relationship makes $\bar{d}_{32}$ and d_{max} in all the models presented in Sections 2.1 and 2.2 interchangeable. The equations below show how $\bar{d}_{32}$ is calculated if the number frequency $f_n(d_i)$ or the volume frequency $f_v(d_i)$ are given.

$$\bar{d}_{32} = \frac{\sum_{i=1} f_n(d_i) d_i^3}{\sum_{i=1} f_n(d_i) d_i^2} = \frac{\sum_{i=1} f_v(d_i)}{\sum_{i=1} \frac{f_v(d_i)}{d_i}} \tag{2}$$

where d_i is the *i*th droplet diameter.

As many emulsions may have the same d_{max} and/or $\bar{d}_{32}$ but different DSD, it is highly desirable to obtain a model which describes the whole distribution, specially when the DSD are bimodal.

2. Theoretical background

2.1. Mechanistic models

It is widely accepted that in turbulent flow droplets can break by two types of stresses depending on the droplets size in relation with the size of the smallest possible eddies. According to Kolmogorov (1949) the length scale of the smallest eddies η for isotropic turbulence is given by

$$\eta = \left(\frac{\mu_c}{\rho_c}\right)^{\frac{3}{4}} \varepsilon^{-\frac{1}{4}} \tag{3}$$

where μ_c and ρ_c are the viscosity and density of the continuous phase and ε is the local energy dissipation rate which value depends on the location of the tank, thereby it is more convenient to use the average energy dissipation rate $\bar{\varepsilon}$ or the maximum energy dissipation rate ε_m, both being proportional for geometrically similar systems (Leng and Calabrese, 2003)

$$\varepsilon_m \propto \bar{\varepsilon} \propto \frac{P}{\rho_c V} \propto \frac{Po \rho_c N^3 D^5}{\rho_c D^3} \propto PoN^3D^2 \tag{4}$$

where *P* is the power consumption, *V* is the volume of the vessel and *Po* is the dimensionless power number $\left(Po = P/\rho_c N^3 D^5\right)$. For a geometrically similar mixers and constant $Po : \varepsilon \sim N^3D^2$ where *N* is the