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Dispersion of water into oil in a rotor–stator mixer. Part 1: Drop breakup in dilute systems

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ABSTRACT

Most previous studies of liquid–liquid dispersion in complex geometry are limited to turbulent flow at low continuous phase viscosity. In this study, a viscous continuous phase was employed over a range of flow conditions including both the laminar and turbulent regimes. Equilibrium drop size was measured for water dispersed into viscous food grade mineral oils in a batch Silverson L4R rotor–stator mixer. The influence of fluid viscosities and interfacial tension (by adding an oil-soluble surfactant) were examined. In order to isolate the effect of drop breakage from coalescence, Part 1 is limited to dilute conditions (water phase fraction, $\phi = 0.001$). In the laminar regime, drop breakup was more likely due to a simple shear breakage mechanism than one for extension. Following Grace (1982), a semi-empirical drop size correlation was developed. For turbulent flow, the validity of the sub-Kolmogorov inertial stress model for correlating equilibrium mean drop size was verified. Surfactants were found to mostly decrease drop size by lowering interfacial tension. Except for laminar systems near the critical micelle concentration, where Marangoni stresses appear to play some role, the effect of surfactants on the drop size could be correlated using the equilibrium or static interfacial tension. The influence of water phase fraction and coalescence is considered in Part 2 (Rueger and Calabrese, 2013) of this paper.

Keywords: Liquid–liquid dispersion; Drop breakup; Rotor–stator mixer; High-shear; Drop size

1. Introduction

Mixing is industrially important in a variety of contexts where emulsions and liquid–liquid dispersions are produced, including paints and coatings, cosmetics and beauty care, food and household products, pharmaceuticals and other applications (Paul et al., 2004). Whether the purpose of the process is to create a stable emulsion or temporarily form large interfacial area per volume, it is desirable to obtain scaling laws so that multiphase processes can be scaled up using the results of laboratory-scale experiments.

Early liquid–liquid dispersion studies tended to use simple impellers in a baffled tank with a low viscosity continuous phase in turbulent flow. However, this is not the situation in many industrial processes; often, the equipment is more complex, the continuous phase has a high viscosity, and flows are not restricted to the turbulent regime. There is a variety of emulsification devices which provide a range of agitation rates and have a variety of throughput capacities such as stirred tanks, static mixers, valve homogenizers, and rotor–stator mixers. Rotor–stator mixers produce more intense shear fields than conventional stirred vessels because the rotor rotates at higher speeds, and more importantly, because of the close clearance between the rotor and stator.

For all dispersion processes, there is generally a trade-off between the required power input and the resultant drop size distribution (DSD). For turbulent fluids, this relationship between power input and DSD has been found to be applicable for a variety of mixing devices and independent of the specifics of the mixer geometry (Davies, 1987). For processes which are allowed to proceed to equilibrium, the stress that breaks up a drop into its smallest size is not due to the average power input, but to the maximum local energy dissipation rate (Zhou and Kresta, 1998). Therefore, what is needed to break up a drop to a specified size is a certain intensity of the maximum local shear stress which requires a specified local energy dissipation rate. Therein lies the advantage of rotor–stator mixers (pictured in Fig. 1); most of the energy that is supplied to this

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Nomenclature

Ca	Capillary number
$Ca_c = \mu_c \dot{\gamma} d_{32}/\sigma$	Capillary number based on continuous phase viscosity and d_{32}
$Ca_c' = \mu_c \dot{\gamma} d_{max}/\sigma$	Capillary number based on continuous phase viscosity and d_{max}
$Ca_d = \mu_d ND/\sigma$	Capillary number based on dispersed phase viscosity and rotor diameter
d	drop diameter
d_{32}	Sauter mean diameter
d_i	nominal diameter of the ith size bin
d_{max}	maximum stable drop diameter
D	impeller or rotor diameter
$E(k)$	turbulent energy spectral density function
g	gravitational constant
k	wavenumber in the energy spectral density function
K	constant of proportionality between the characteristic shear rate and the nominal shear rate
L	macro length scale of turbulence
N	rotor rotation rate
n_i	number of drops in the ith bin
N_B	number of bins in Eq. (8)
$N_P = P/\rho_c N^3 D^5$	Power number
P	power
$Re = \rho_c ND^2/\mu_c$	Reynolds number
t	time in Eq. (6)
$\overline{v'(d)^2}$	turbulent mean-square velocity difference across drop surface
$We = \rho_c N^2 D^3/\sigma$	Weber number
$\dot{\gamma}$	shear rate
$\dot{\gamma}_c$	characteristic shear rate (in laminar flow experiments)
δ	clearance between rotor blade and inner stator wall (shear gap)
ε	energy dissipation rate
η	Kolmogorov micro length scale of turbulence
$\lambda = \mu_d/\mu_c$	viscosity ratio
μ_d	continuous phase viscosity
μ_d	dispersed phase viscosity
ν_c	continuous phase kinematic viscosity
ϕ	phase fraction
ρ_c	continuous phase density
σ	equilibrium interfacial tension
$\sigma(t)$	dynamic interfacial tension
σ_0	initial dynamic interfacial tension
σ_∞	long time (equilibrium) interfacial tension in the presence of surfactants
τ	dynamic interfacial tension time constant
τ_c	disruptive stress acting on drop surface
τ_s	cohesive stress due to interfacial tension resisting drop deformation

mixer is dissipated near the mixing head, with relatively little energy, except that required for mild recirculation, being dissipated in the bulk flow (Yang, 2011). This means that most of the energy put into a rotor–stator mixer is spent in increasing the maximum local shear stress which generally yields smaller drop size.

The issue of how drops break up in rotor–stator devices is itself a key question in this study. The mechanism(s) of drop breakup is/are dependent on the flow regime (laminar, transitional, or turbulent), the specifics of the mixing apparatus, and the fluids in question. The flow regime for most previous studies has been turbulent due to the use of a low viscosity continuous phase. However, this study includes both the laminar regime and the turbulent regime with a viscous continuous phase. This introduces several challenges. Drop breakup in complex laminar flows is poorly understood due to the dependence of the deformation rate field on mixer geometry. Therefore, data must be interpreted by reference to drop breakup in simple or idealized flow geometry. In turbulent flow of viscous liquids, it is possible to produce drops that are smaller than the Kolmogorov microscale, η, invalidating the use of the well-known Weber Number correlation which only applies for $L \gg d \gg \eta$, where L is the turbulent macroscale.

In this work, water is dispersed into mineral oils of different viscosity grades. The increased viscosity of the continuous phase not only results in lower Reynolds number, but also causes the ratio of dispersed to continuous phase viscosity to be quite low. This becomes relevant when comparing the results of the laminar flow experiments to drop breakup studies for idealized flow fields.

In Part 1 of this study, we consider the effect of the continuous phase viscosity, the viscosity ratio, and the interfacial tension (by adding oil-soluble surfactant, Tergitol NP-4) on the mean drop size in dilute water-in-oil emulsions where coalescence phenomena can be ignored. Both laminar and turbulent regimes are explored. Specifically, the goal is to define dimensionless groups and develop mechanistic models that correlate the data, in order to develop general rules for scaling up drop size. While a rotor–stator mixer is used in this study, it does not mean that the results are necessarily restricted to these devices. The arguments developed herein can be applied to other process equipment in which drops break according to the same mechanism(s).

2. Theory

Dilute emulsions (dispersed phase fraction, $\phi = 0.001$) are the simplest to analyze because the structure of the flow field is essentially unchanged by the presence of the drop phase, and because of the absence of coalescence due to the rarity of drop–drop interactions. For more concentrated emulsions (discussed in Part 2) (Rueger and Calabrese, 2013), the equilibrium DSD is reached when there is a dynamic balance between the rates of breakage and coalescence (Coulaloglou and Tavlarides, 1977). The absence of coalescence in dilute systems allows for the isolated study of the effect of breakage so that an emulsion may be said to have reached "equilibrium" when all of the drops are below the maximum stable drop size, as determined by the maximum deformation rate in the flow field (Leng and Calabrese, 2004). Part 1 includes dilute data for both laminar and turbulent flow regimes with the results analyzed separately.

2.1. *Turbulent systems*

The analysis of drop breakup in turbulent flow began with Kolmogorov's theory of cascading turbulent eddies and small-scale isotropic turbulence (Kolmogorov, 1941a,b,c, 1949). Hinze (1955) applied this work to describe a critical Weber number based on drop diameter which determines whether or not a drop breaks in a given deformation field. When