



Emulsification of particle loaded drops in simple shear flow



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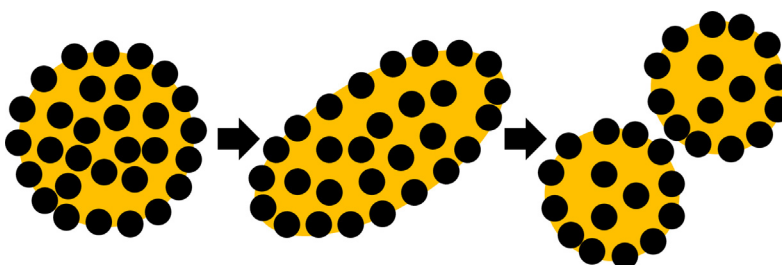
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HIGHLIGHTS

- Particle load of drops influences their breakup in simple shear flow.
- Influence depends on particle interaction with liquid/liquid interface.
- Particles situated in the liquid/liquid interface do not hinder drop breakup.
- Particles situated inside the disperse phase hinder drop breakup.
- Changes in drop breakup behavior can be related to the shear thinning flow properties of the disperse phase.

GRAPHICAL ABSTRACT



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ABSTRACT

We investigated the influence of a particle load of disperse phase drops on their emulsification. Newtonian liquids were chosen for this investigation and a surfactant was present in all cases. A Couette-like shear cell was used enabling us to apply simple shear flow. The drop size distribution and drop morphology of the resulting oil-in-water emulsion were characterized. The influence of the wetting characteristics of the particles and the influence of the volume fraction of solids were investigated by comparing the breakup of silicone oil drops containing two types of particles, which showed different wetting behavior with that of pure silicone oils. One type of particles was situated in the liquid-liquid interface, the other type remained mainly wetted in the oil drops. With increasing volume fraction both suspension types showed shear rate dependent rheological behavior due to particle-particle interactions. Emulsification results were interpreted by means of apparent critical capillary numbers and correlated with the measured drop-to-matrix viscosity ratio. To calculate the viscosity ratio, the viscosities of the disperse phases were measured for each applied shear stress. Depending on the wetting characteristics, the particle load of the disperse phase showed different influence on the emulsification result. Particles situated in the liquid-liquid interface did not affect the apparent critical capillary number and therefore the drop breakup in the investigated volume fraction range. This result was explained by the reduction of particle loading during drop deformation and breakup due to the shearing-off of highly filled drops. Therefore the matrix fluid of the disperse phase governs the overall emulsification result. In contrast, particles located inside the disperse phase hindered the drop breakup. In comparison to Newtonian liquids with comparable bulk viscosities, the apparent critical capillary number was higher. It increased with increasing particle volume fraction. Additionally the apparent critical capillary number showed a dependence on the applied stress in case of the particle containing drops. This behavior is assumed to be due to an inhomogeneous viscosity inside the particle-loaded drop. We suggest using an effective drop viscosity to calculate the viscosity ratio in case of this type of system.

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Abbreviations: PEG, polyethylene glycol; SDS, sodium dodecyl sulfate; AK10-1000, silicone oils with different viscosity; PS, polystyrene.

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Nomenclature

Symbols

Ca	[-]	capillary number
Ca_{crit}	[-]	critical capillary number
Ca'_{crit}	[-]	apparent critical capillary number
x	[μm]	drop diameter
$x_{90,3}$	[μm]	characteristic drop diameter, corresponding to 90% of the cumulative undersize distribution
M	[Nm]	torque
M_{rotor}	[Nm]	torque at the rotor surface
n	[rpm]	rotational speed
r	[m]	cone radius of the rotor
w	[%]	mass fraction
d	[mm]	capillary tube diameter

Greek symbols

β	[$^\circ$]	half of the rotor-cone angle ($^\circ$)
η	[Pa s]	dynamic viscosity
η_d	[Pa s]	dynamic viscosity of the disperse phase
η_c	[Pa s]	dynamic viscosity of the continuous phase
λ	[-]	viscosity ratio
ρ	[kg/m ³]	density
σ	[N/m]	interfacial tension
τ	[Pa]	shear stress
Φ	[%]	volume fraction
ω	[rad/s]	angular velocity

1. Introduction

Emulsion based products are widely-used in food, chemical, and pharmaceutical industries. In many applications, emulsions do not consist of two pure Newtonian liquids, but an additional solid phase is incorporated in the disperse liquid phase, for example in the production of inorganic/organic particles via miniemulsion polymerization [1–3] or the incorporation of solids in polymer blends [4]. As the final drop size distribution determines important emulsion properties like stability, rheology, or particle morphology, it is necessary to analyze the drop breakup mechanism during the emulsification step and to understand the influence of additional compounds in such multicomponent mixtures. Many experimental and numerical results regarding the breakup of a Newtonian drop in a Newtonian matrix are reported in literature [5–11]. The deformation and breakup of a single Newtonian drop immersed in a Newtonian matrix fluid under quasi-static flow conditions are governed by the intensity and duration of the deforming viscous stresses and the counteracting interfacial and viscous stresses. The behavior can be characterized by the capillary number Ca and the viscosity ratio λ . Ca describes the balance between flow induced shear stress and counteracting interfacial stress:

$$Ca = \frac{\tau x}{2\sigma} \quad (1)$$

where x is the drop diameter, τ the shear stress and σ the interfacial tension. λ is defined as the ratio between the viscosities of the disperse phase η_d and continuous phase η_c :

$$\lambda = \frac{\eta_d}{\eta_c} \quad (2)$$

For drop breakup to occur, Ca has to exceed a critical value, the critical capillary number Ca_{crit} . Depending on the difference between the applied Ca and Ca_{crit} , different breakup mechanisms are found [12,13]. For $Ca \geq Ca_{crit}$ binary breakup occurs and the drop breaks into two main parts and several smaller satellite drops. For $Ca \gg Ca_{crit}$ the drop stretches into a long filament and breaks

because of Rayleigh instabilities. This breakup mechanism is often referred to as capillary breakup. Additionally, the influence of λ on the drop breakup depends on the flow regime. In simple shear flow, for $\lambda > 4$ no drop breakup is possible as the drop starts to rotate. In contrast, in elongational flow, even at significantly higher viscosity ratios, drops can be broken up [14]. Therefore, besides the rheological properties of the emulsion, mainly the shearing conditions influence drop breakup and the final product properties [12,15,16]. Regarding industrial technological processes not only Newtonian disperse phases are of interest but also non-Newtonian and most often complex multi-component emulsion systems containing surface-active molecules and solid material. To tailor-make emulsion-based microstructures, the interplay of these different components during processing has to be understood. For systems containing surfactants, it is important to mention that besides the above described drop breakup mechanisms, tip streaming can occur in simple shear flow. Tip streaming is related to the presence of interfacial tension gradients, leading to the expulsion of a stream of tiny drops at the drop tips [17].

Few studies deal with the deformation and breakup behavior of particle loaded drops. Single drop experiments by Smith and Van de Ven [18] showed in a qualitative way that particles inside the disperse phase influence drop deformation and breakup behavior. Though, a pronounced effect was only detected for high volume fractions of particles. Furthermore, interactions of applied surfactant with the liquid-liquid- as well as the liquid-solid interface, and the occurrence of three-phase contacts are pointed out to change the drop behavior under simple shear. Other authors report an increase in drop size when particle loaded drops are emulsified using ultrasonication or high pressure homogenization [19,20]. Numerical simulation of the influence of nanoparticles on drop deformation showed a decreased deformation for increased volume fraction of particles [21]. Besides an increasing viscosity, particle containing phases generally show a visco-elastic or shear thinning behavior with increasing particle load [22,23]. Various publications consider the drop deformation and breakup of non-Newtonian disperse phases without particle load. Numerical and experimental studies showed that for visco-elastic drops the mode of deformation and breakup not only depends on the viscosity ratio but also on the elasticity of the drops [24–27]. Regarding shear thinning drops, one has to distinguish whether the comparison with Newtonian drops is done at the same zero-shear viscosity or the same viscosity at the applied shear stress. If the zero-shear viscosity is considered, the deformation of shear thinning drops is higher than the deformation of the corresponding Newtonian liquid [28]. If instead the viscosity at the applied shear stress is considered, shear thinning drops are less deformed and breakup occurs at a higher capillary number in comparison to Newtonian liquids [29–32]. In case of shear thinning disperse phases, the critical capillary number is dependent on the degree of shear thinning behavior [33] and the applied shear stress [31]. As reason for the divergent behavior, the inhomogeneous viscosity profile inside a shear thinning drop under shear is reported. Because of the shear thinning properties of the drop, the viscosity profile inside the drop shows a strong increase from the liquid-liquid interface towards the center of the drop [28,33]. De Bruijn showed that the breakup behavior of shear thinning liquids can be described by the theory developed for Newtonian liquids provided that the inhomogeneous drop viscosity profile is taken into account by calculating the viscosity ratio with an adjusted effective disperse phase viscosity [27].

In this work, an experimental investigation on the emulsification of particle loaded drops suspended in a Newtonian matrix is presented. To investigate the influence of the wetting behavior of the particles, two types of particles with different 3-phase contact angles were used. The drop breakup behavior is not investigated via single drop experiments but by the analysis of the resulting drop

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