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Continuous, recycle and batch emulsification kinetics using a high-shear mixer

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HIGHLIGHTS

- Results for three emulsification arrangements were linked using a Poisson chain model.
- The model allowed for comparison in-between arrangements using in-line rotor-stators.
- A simplification of the Poisson chain model for long times was proposed.
- The simplified model explained kinetics and volume dependency for batch systems.
- Modes were used as measure of central tendency due to the noise that bimodality produced.

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ABSTRACT

In-line rotor-stator mixers are widely used to emulsify immiscible liquids, but there is little understanding on the size reduction kinetics and on their performance if used in a continuous or in a recycle arrangement. In this study, a systematic series of experiments were performed to study the droplet size reduction of a 10 cSt silicon oil coarse emulsion using different flow rates and number of passes for a continuous and a recycle configuration using a L5M-A Silverson Laboratory mixer.

The droplets size distributions obtained were bimodal. It was found that the mode of the larger daughter droplets is a better parameter to follow the emulsification kinetics than the Sauter mean diameter; the volume fraction of the small daughter droplets was estimated to be ≈6% using two generalized gamma functions. For the continuous arrangement it was found that impeller speed and mean residence time inside the rotor-stator correlated with an average error of 3.2%. It was also found that a Poisson chain can link the results of both operation arrangements. This stochastic model allowed following the disappearance rate of the coarse droplets and the evolution of the daughter droplets' mode with an average error of 2.3%. The model developed allowed the comparison of both arrangements using an in-line rotor-stator.

A simplification of the Poisson chain model for long times was used to analyse batch emulsification kinetics. The droplet size reduction rate and batch volume could be explained in terms of the in-line results.

1. Introduction

Liquid-liquid mixtures are present in a wide variety of products such as pharmaceuticals, cosmetics and foods; and in unit operations such as extraction and emulsion polymerisation. The performance of these products and processes relies on the Droplets Size Distribution (DSD) of the emulsion as it determines the available area for mass transfer, stability and rheological behaviour. In many industries stirred vessels have been substituted by high-shear mixers due to their versatility and the high shear fields produced by a high tip speed and specially by the narrow gap in-between the rotor and the stator. High-shear mixers are versatile as they can be used in batch and continuous operation modes. Despite the popularity of high-shear rotor-stator mixers, we found no evidence in literature of a comparative model for these arrangements. Experimental studies studies using different arrangements and rotor-stator mixers include the one of Bourne and Studer (1992) who made experiments using a recycle and a continuous arrangement but were unable to stablish link both operation mode results. More recently Håkansson et al. (2016) emulsified mayonnaise

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Nomenclature

Latin symbols

$\dot{Q}$	volumetric flow rate [$m^3\,s^{-1}$]
$\overline{d}$	droplet mean diameter [µm]
$\bar{t}$	mean time [s]
a	surface to unit volume [m^{-1}]
C	fit constant [–]
D	diameter of the rotor [m]
d	droplet diameter [µm]
E	energy [J]
F	cumulative frequency [%]
f	density frequency [%]
K	power constant [–]
k	proportionality constant [–]
M	torque [N m]
Mo	mode by volume [µm]
N	rotational speed [s^{-1}]
n	number of passes [–]
P	power [W]
pn	pump number [–]
R^2	coefficient of determination [–]
T	temperature [K]
t	time [s]
V	volume [m^3]

Greek symbols

ϵ	energy dissipation rate [$m^2\,s^{-3}$]
τ	shape parameter in GGf [–]
λ	scale factor [–]
μ	dynamic viscosity [Pa s]
ν	kinematic viscosity [$m^2\,s^{-1}$]
ϕ	volume fraction of droplets [–]
ρ	density [$kg\,m^{-3}$]
σ	interfacial tension [$N\,m^{-1}$]
κ	shape parameter in GGf [–]
φ	volume fraction of the dispersed phase [–]

Dimensionless numbers

N_Q	Pumping number $\dot{Q}N^{-1}D^{-3}$
Po	Power number $PN^{-3}D^{-5}\rho^{-1}$
Re	Reynolds number $ND^2\rho\mu^{-1}$
We	Weber number $\rho N^2D^3\sigma^{-1}$

Subindeces

f	measured with viscid fluid
32	volume to surface ratio
adj	adjusted
$cont$	continuous arrangement
0	measured with inviscid fluid
i	counter (1, 2, ..., i)
KH	Kolmolgoroff-Hinze
Lam	laminar
m	measured
n	counter for number of passes
Q	flow
res	residence
S	Silverson rotor-stator
T	tank
$Turb$	turbulent
U	free flow
v	volume
Z	zero flow

Abbreviations

DSD	droplet size distribution
GGf	generalized gamma function
SLES	sodium laureth sulfate

using batch and continuous arrangements and used an average number of rotor-stator passes for comparison, nevertheless when the drop sizes are plotted as function of the average number of passes for both arrangements, these did not fall into a master curve. The focus of this work is to compare the multi-pass continuous operation mode and the batch recycle arrangement using an in-line rotor-stator.

The mechanistic models for emulsification in the turbulent regime are based on the work developed by Kolmolgoroff and Hinze (1955), who proved the existence of two emulsification regimes depending if the droplets are larger or smaller than the smallest possible eddies; namely the inertia and viscous regime. The order of magnitude of these or the Kolmolgoroff's scale λ_{KH} for isotropic turbulence can be determined by

$$\lambda_{KH} = \left(\frac{\nu^3}{\epsilon}\right)^{\frac{1}{4}} \tag{1}$$

where ν is the kinematic viscosity of the continuous phase and ϵ the energy dissipation rate which can be approximated as $\epsilon \sim N^3D^2$; where N is the stirring speed and D is the impeller's diameter. In their review on rotor-stator mixing devices (Atiemo-Obeng and Calabrese, 2004) claim that droplets obtained in these homogenizers are above the Kolmolgoroff length-scale, and thus they work in the turbulent inertia regime. In the turbulent inertia regime, where the diameter of the droplets is larger than λ_{KH}, Hinze (1955) developed a theory where the maximum stable droplets d_{KH} is determined by the balance between the fluctuations of the hydrodynamic pressure of the continuous phase and the Laplace pressure inside the droplets. Shinnar and Church (1960) assumed that $\overline{d}_{32} \propto d_{KH}$, for an inviscid dispersed phase and for fully turbulent flow they determined that

$$\frac{\overline{d}_{32}}{D} \propto We^{-3/5} \tag{2}$$

where $\overline{d}_{32}$ is the Sauter mean diameter and We is the dimensionless Weber number ($We = \rho N^2D^3\sigma^{-1}$). Most of the correlations found in literature for emulsification technology use $\overline{d}_{32}$ as the expression of central tendency to be determined. This is due to the importance that the interfacial surface to unit volume a has, which can be obtained from $a = 6\varphi/\overline{d}_{32}$, where φ is the volume fraction of the dispersed phase. $\overline{d}_{32}$ can be calculated from

$$\overline{d}_{32} = \frac{\sum_i f_v(d_i)}{\sum_i \frac{f_v(d_i)}{d_i}} \tag{3}$$

where $f_v(d_i)$ and d_i are the frequency by volume and the diameter of the ith droplet.

Energy dissipation theory has been verified by authors such as Chen et al. (1967) and has been used as starting point to extend dissipation theory to account for volume fraction (Leng and Calabrese, 2004), viscous dispersed phases (Davies, 1985; Calabrese et al., 1986) and different impellers (McManamey, 1979; Leng and Calabrese, 2004). Nevertheless, EL-Hamouz et al.