



Fluid dynamics and mass transfer at single droplets in liquid/liquid systems



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ABSTRACT

The design of contact apparatuses in which one liquid phase is dispersed into another is a challenging task with regard to fluid dynamics and mass transfer. To disentangle the complexity in these multiphase polydispersed systems with all their coupled mutual interactions, one first step towards a reliable prediction is to reduce the system to a single droplet problem, where a single droplet is moving in a quiescent ambient phase. Commonly, one is interested in the mass transfer coefficient (or Sherwood number) in case of the transfer of a soluble component, and the drop velocity of fall/rise. In order to save time and money, one objective is to predict the behaviour of a given system as reliable as possible to minimise the own experimental (and numerical) effort. However, although the issue is “only” to understand the single droplet, merely in a few basic cases analytical equations are available and applicable, and results based on CFD methods have to be judged with care. Additionally, in most cases experimental or numerical data for a given system may not be available at all, hence the practitioner has to select the best suitable empirical or semi-empirical approaches for the corresponding system which itself is a challenging task. This paper aims to help to make the required decisions and to enable the reader to characterise and confine a given extraction system regarding fluid dynamics and mass transfer. Thereto, a chart is presented to initiate a selection process starting from simpler cases to cases with increasing complexity, especially addressing the behaviour of systems dominated by Marangoni instabilities.

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1. Introduction

Dispersed systems consist of solid or fluid particles which are immersed in a surrounding ambient phase. They play a major role in many diverse industrial applications, such as chemical, pharmaceutical, food, petrol and polymers, waste water treatment, hydro-metallurgy, nuclear industry and many more. In most cases, engineers face the challenging task to separate the phases, extract the desired transfer component(s), with or without use of chemical reactions to obtain the required concentration or quality of a certain product.

Cyclones or centrifuges are used to separate particles from fluid streams. In mixer/settler plants, the contact area between the phases is increased in the mixer to accelerate mass transfer, in the settler stage the phases are allowed to separate by coalescence, agglomeration or flotation. Polymerisation or crystallisation processes are other examples for multiphase systems. In liquid/liquid extraction processes, liquid phases are brought in contact to exchange a transfer component according to the distribution

coefficient; in spray drying, a liquid component dries and solidifies in a hot gas stream. As different as the applications and apparatuses are, they all have in common that swarms of bubbles, droplets, or particles are dispersed in a continuous ambient phase, either a gas or a liquid. In the following, we will only focus on single droplets dispersed in liquids. Bubbles and rigid particles are only considered peripherally here. Note also that chemical reactions and heat transfer are not considered in this paper. The challenge for an engineer is to control, optimise and in the best case to predict the process in terms of mass and momentum transfer, anticipating the manifold and complex interactions in multiphase systems. In this regard, it is particularly useful to reduce the complexity of swarm systems to single droplets, since droplets are the smallest mass transfer unit, e.g. in a liquid/liquid extraction column.

All transport processes occur at or across the interface of each droplet in a collective with a characteristic size distribution, hence fluid dynamics and mass transfer are inseparably linked with interfacial properties and the associated interfacial phenomena, such as deformation, oscillation, Marangoni instabilities, surfactant ad- or desorption, etc. An intensive look into the literature reveals that the description of droplets in liquids seems to be more difficult than bubbles in liquids or rigid particles in gas, since the viscosity

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Nomenclature

Latin letters

A	cross section, m^2
c	concentration, kg/m^3
c^*	dimensionless mean concentration, –
C_D	drag coefficient, –
d_p	drop diameter, m
D	molecular diffusivity, m^2/s
D^*	ratio of diffusivities $D_{A,d}/D_{A,c}$, –
E	extraction efficiency, –
F	force, N
g	gravitational acceleration, m/s^2
k_B	Boltzmann constant, $1.38 \cdot 10^{-23}$, J/K
k_o	overall mass transfer coefficient, m/s
K^*	partition coefficient, –
\dot{m}	mass flux, $kg/(m^2 s)$
M	mass of the drop, kg
R	drop radius, m
t	time, s
T	temperature, K
v	velocity, m/s
V	drop volume, m^3

Greek letters

α	diffusivity factor, –
β	mass transfer coefficient, m/s
δ	boundary layer thickness, m
κ	virtual mass coefficient, –
μ	dynamic viscosity, $Pa s$
μ^*	viscosity ratio μ_d/μ_c
ρ	density, kg/m^3
ρ^*	density ratio ρ_d/ρ_c , –

$\Delta\rho$	density difference $ \rho_d - \rho_c $, kg/m^3
γ	interfacial tension, N/m

Subscripts

A	transfer component
B	buoyancy
c	continuous phase
cr	critical
d	dispersed phase, droplet
G	gravitation
hi	high
i	inertia
I	interface
lo	low
MC	Marangoni convection
o	overall
P	particle, droplet
ra	reacceleration
s	solute
$s0$	solute at initial conditions
t	terminal

Dimensionless numbers

Eo	Eötvös number	$Eo = g\Delta\rho d_p^2/\gamma$
Fo	Fourier number	$Fo = D_s \times t/R^2$
Mo	Morton number	$Mo = g\mu_c^4\Delta\rho/(\rho_c^2\gamma^3)$
Pe	Peclet number	$Pe = v_t d_p/D_s$
Re	Reynolds number	$Re = v_t d_p \rho_c/\mu_c$
Sc	Schmidt number	$Sc = \mu_c/(\rho_c D_{s,c})$
Sh	Sherwood number	$Sh = \beta d_p/D_s$
We	Weber number	$We = v_t^2 d_p \rho_c/\gamma$

ratio for the latter two examples is extreme (0 and ∞ , respectively) which makes modeling issues more accessible (which does not mean that these systems are not complex!). Another look into the literature also reveals that a vast amount of work has been done over the last decades to measure, correlate, model, and predict mass transfer coefficients and terminal drop rise velocities in a likewise vast amount of different liquid/liquid systems for various initial and boundary conditions. This makes it even more difficult to choose the correct correlation (if available at all) for a given system.

So let us assume the mass transfer coefficient β and the terminal drop velocity v_t (hence the drag coefficient) are required for a specific liquid/liquid system. A literature survey may have shown that there are no reliable results from other groups available, and also there are not enough capacities to perform own laboratory experiments. The question arises whether it is yet possible to confine and to derive from available data a reasonable estimate for the current liquid/liquid system with respect to β and v_t ?

Fig. 1 introduces the structure of the present paper and may be regarded as a selection chart for a specific problem. Starting point in Fig. 1 is a single droplet immersed in an ambient quiescent liquid which rises or falls according to the forces exerted at the interface of the droplet. In the case without additional mass transfer, see Section 2, the droplet velocity only depends on the fluid dynamic behaviour. In general, the droplets may be of spherical, deformed or oscillating shape according to their Reynolds/Eötvös/Morton number combination. One extreme case is represented by the fully mobile interface which can be defined by a complete absence of impurities or interfacial active molecules such as surfactants. In this case, the droplet would reach the maximum possible

terminal velocity for a given set of physical properties. For partly mobile interfaces, the terminal velocity reduces but inner circulations are still possible to a certain extent. The other extreme case is a droplet with immobile interface, or in other words the tangential velocity component is always zero. This may be the case in contaminated systems or in systems with presence of surfactants which completely block the whole interface. The terminal velocity is minimum. Note that normal components of the interfacial velocities can still be non-zero, e.g. due to shape deformation.

Section 2 focuses on how to estimate the terminal velocity of a single droplet in a quiescent ambient liquid. The standard drag curve for rigid spheres is briefly mentioned followed by a more detailed look on the different regimes of single droplets (spherical, transition and oscillatory regime). Equations and correlations available in literature are then introduced for each regime and eventually compared to experimental terminal velocity data of five different liquid/liquid systems together with some CFD simulation results available in literature.

In the case of mass transfer, Section 3, one should first estimate where the main mass transfer resistance resides: inside the droplet only (internal problem, $K^* \sqrt{D^*} \ll 1$), outside the droplet only (external problem, $K^* \sqrt{D^*} \gg 1$) or in both phases (conjugate problem, $K^* \sqrt{D^*} \approx 1$), K^* being the partition coefficient and D^* the ratio of diffusivities. In the corresponding Sections 3.2–3.5, a spherical fully mobile interface is assumed. For the external problem, an example is given which describes simultaneous adsorption of surfactants (Section 3.3). Only in the case of the conjugate problem, differences in the concentration of the transferred component may occur which may lead to a change in the interfacial tension γ . This may eventually trigger Marangoni convection which

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