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CFD aided investigation of single droplet coalescence

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ABSTRACT

This article describes the development of a coalescence model using various CFD work packages, and is validated using as toluene water model system. Numerical studies were performed to describe droplet interactions in liquid–liquid test systems. Current models use adjustable parameters to describe these phenomena. The research in the past decades led to different correlations to model coalescence and breakage depending on the chemical system and the apparatus geometry. Especially the complexity of droplet coalescence requires a detailed investigation of local phenomena during the droplet interaction. Computational fluid dynamics (CFD) studies of single droplet interactions were performed and validated with experimental results to improve the understanding of the local hydrodynamics and film drainage during coalescence. The CFD simulations were performed for the interaction of two differently sized droplets at industrial relevant impact velocities. The experimental verification and validation of the numerical results were done with standardized high-speed imaging studies by using a special test cell with a pendant and a free rising droplet. An experimental based algorithm was implemented in the open source code OpenFOAM to account for the contact time and the dimple formation. The standard European Federation of Chemical Engineering (EFCE) test system toluene/water was used for the numerical studies and the experimental investigations as well. The results of the CFD simulations are in good accordance with the observed coalescence behavior in the experimental studies. In addition, a detailed description of local phenomena, like film rupture, velocity gradients, pressures and micro-droplet entrainment could be obtained.

1. Introduction

Liquid–liquid extraction is applied in many chemical, petrochemical, biochemical, hydrometallurgical and nuclear separations processes. Column contactors are widely used for extraction due to the advantages of high throughput and low footprint [1] compared to alternative equipment options. An essential factor governing extraction-column performance is the competitive relationship between breakage and coalescence of droplets, which determines the interfacial area available for mass transfer. Characterization and prediction of both components are necessary, for process optimization purposes, in unit-scale modeling. Understanding the complex relationship between the various factors influencing coalescence, and increasing the accuracy and reliability of model predictions, have been an ongoing topic of scientific research [2–5]. All currently available models depend on the use of adjustable parameters in order to model specific test systems and apparatus geometries. Existing correlations neglect important factors such as electrostatic effects; which depend on the electrolyte ion species, concentration, pH value; as well as parameters characterizing relative droplet inertia, collision angle and the presence of additives. As such, previous studies have given inconsistent results. In order to increase the reproducibility of collision conditions, Eiswirth and Bart [5] and Villwock *et al.* [6] developed a standardized, automated experimental approach and used it to characterize a water/toluene test system. Generation of numerous collision sequences is of crucial importance in establishing a statistically valid coalescence probability database for the detailed characterization of test systems. The recently developed methodology is used, presently, in the development and validation of new coalescence models.

2. Theory

Coalescence is more complex than breakage, because there are hydrodynamic effects determined by the energy input and geometry of the apparatus, but also by the composition of the phases. Additionally the mass transfer has a decisive effect on the coalescence behavior. An early model describing coalescence of droplets is given by Coulaloglou and Tavlarides [3]. Here the first condition for successful coalescence is the knowledge of the collision frequency h_{coal}, depending on the number of droplets in the considered volume element. The determination of the collision frequency depends on swarm experiments and is not applicable to single droplet investigation. A description of the coalescence behavior with the collision frequency is not sufficient, since not every droplet interaction results in a coalescence event. It is necessary to introduce the coalescence efficiency λ to account for the observed

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behavior. A simple definition of the coalescence efficiency depending on the droplet sizes of the colliding droplets d_1 and d_2 is the ratio between coalescence events (N_{coal}) and droplet interactions (N_{int}):

$$\lambda_{d_1,d_2} = \frac{N_{coal}}{N_{int}}. \quad (1)$$

The focus in the following is on different physical and empirical approaches to calculate the coalescence efficiency. The prediction and the modeling of the droplet size distribution (DSD) with population balance equations (PBE) partially supported by CFD simulations depend mainly on the accuracy of the coalescence kernel. The DSD is influenced by physical properties, apparatus geometry and process parameters, but several factors, for instance the electrochemical effects or the impact conditions, are not considered in current kernels. The existing coalescence probability models can be roughly distinguished into two main groups. The basic film drainage model of the first group is given by Coulaloglou [7] and is characterized by the coalescence time needed for coalescence and the contact time between the two droplets:

$$\lambda_{d_1,d_2} = \exp\left(-\frac{t_{drainage}}{t_{contact}}\right). \quad (2)$$

Coalescence can only occur when the force which brings the droplets into contact is acting for a sufficient time ($t_{contact}$). The contact time must exceed the time to reach the critical film thickness ($t_{drainage}$). Due to the different definition of the characteristic times in various research groups several models were developed. One approach is the implementation of electrostatic effect in coalescence kernels by using the DLVO theory [8] giving:

$$\lambda_{DLVO} = \exp\left(-\frac{F_{el}}{|F_{vdW}|}\right). \quad (3)$$

An alternative is based on the consideration of the interfacial energy and the kinetic energy of the droplets [10]:

$$\lambda = \exp\left(-C_y\frac{E_\sigma}{E_{kin}}\right). \quad (4)$$

Different extensions with the droplet size and the relative velocity, leading to a momentum-based calculation of the kinetic energy are given by Simon [11]. A more detailed list of the existing coalescence models is published by Simon [11] and Liao and Lucas [12].

3. Experimental Setup

Due to the sensitivity of the coalescence process to impurities, pH-value, *etc.* a firm definition of the experimental conditions is necessary for a precise understanding of the occurring phenomena. Reproducible and statistically reliable single droplet investigations were performed with a specially designed test cell. A detailed description of the experimental setup with all features and the basic control routine is described by Kamp and Kraume [9]. The test cell has been improved and all changes are described in detail by Villwock *et al.* [6]. The experiments were recorded with a high speed camera (Photron Fastcam APX RS) equipped with a macro lens and an adjustable bellow to register the coalescence with 30,000 frames per second (fps) and a high magnification (Fig. 1). The physical properties of the used EFCE test system are given in Table 1.

4. CFD Setup

The volume of fluid (VoF) approach called multiphaseInterFoam with the additional extension of adaptive mesh refinement based on the OpenSource toolbox OpenFOAM® version 2.3.0 was used for the simulations. The numerical framework to specify the experimental single droplet test cell is built with a purely structured hexahedral

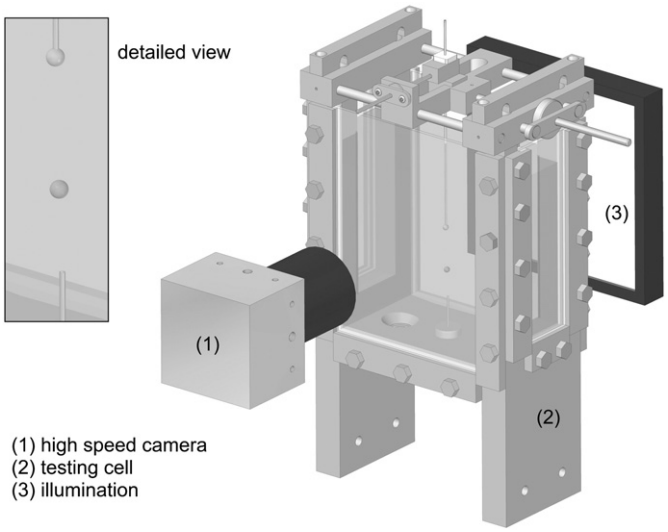

Fig. 1. CAD draft of the single droplet test cell based on Villwock *et al.* [6].

Table 1
Test system physical properties of the laboratory scale experiments, at 20 °C [13]

Substance	Toluene	Water
ρ/kg·m^{-3}	862.3	997.02
μ/mPa·s	0.552	0.8903
γ/mN·m^{-1}	35	
Purity	>99.99%	<0.5 μS·cm^{-1}

mesh created with the pre-processing utility blockMesh. The boundary conditions for velocity and phase fraction in the simulation framework are set to zeroGradient with the exception for the velocity at the inlet, which is defined as a fixed value (0 0 0). The initialization of the continuous phase (water) and the droplet regions (toluene) is done with the *setFlieds* OpenFOAM® utility. The solver needs to adapt the time step to keep the Courant number below 0.5. The refinement conditions and initial solve options are given in Table 2. Setting a maximum cell number is necessary to limit the computational time. The mesh refinement at each time step improves the accuracy of the interface tracking. Simulations were performed on a single core (Xeon E5345, 2.33 GHz, with a maximum requested 8 GB RAM) using the ELWE-Cluster from the University of Kaiserslautern.

Table 2
Refinement conditions and solution options

Boundary	Value
Refinement	Every step
Maximum cell number	2,000,000
Maximum refinement	3
Initial time step	0.00001 s
Max Courant number	0.5
Write interval	0.00005 s

Taking into account the drainage time given in Eq. (2) an additional algorithm is implemented in the CFD code to avoid a meaningless numerical coalescence. A three phase system with two different phases for the droplets and one for the continuous phase is used. The coalescence is initiated by an algorithm that defines the second dispersed phase from then on as the first dispersed phase droplet by using an experimentally derived contact time. The triggering of the algorithm requires a definition for the initial contact of the droplets. The contact is