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Experimental and numerical investigation of a free rising droplet

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ABSTRACT

A toluene droplet rising in a continuous aqueous phase is studied both experimentally and numerically. The experiments have been performed in a small scale, high-speed measuring unit with a high magnification device to image the process in all details. Numerical simulations are performed with the aid of the level set method implemented in the commercial CFD tool COMSOL Multiphysics 3.3a by COMSOL AB. A new technique for quantitative comparison of experimental and numerical results with respect to the droplet shape is presented. Both terminal velocity and droplet shape in experiments and simulations are found to be in excellent agreement. Based on the experimental and numerical data obtained, an improvement of an existing terminal velocity correlation is suggested. The adjusted correlation can be used for the design and optimisation of liquid–liquid extraction units.

1. Introduction

Liquid–liquid extraction is a well established and widespread unit operation. Despite its extensive application in industry, optimal design and operation of liquid–liquid extraction requires pilot plant experiments which are usually expensive and time consuming. In this respect, model-based simulations can be considered as an attractive alternative or at least as a good support to experimental measurements.

To fully or partially replace pilot plant experiments, development of rigorous and accurate models and algorithms is necessary. The phenomena occurring in liquid–liquid extraction units are very complex and not yet fully understood. Furthermore, the different length scales of the involved phenomena make rigorous modelling of extraction units very difficult. As a first step towards the better understanding of the basic phenomena, smaller yet representative elements can be used. For liquid–liquid extraction, a single droplet can be considered as the smallest representative element.

During the last decades, the free droplet movement within a quiescent medium has been studied extensively, both experimentally and numerically, especially with respect to droplet terminal velocity. One of the first correlations for the calculation of the droplet terminal velocity was published by Hu and Kintner [1]. This correlation was developed for high surface tension liquid systems (e.g. toluene/water). On the other hand, Klee and Treybal [2] derived a correlation applicable for systems with medium surface tension (e.g. butylacetate/water). In this case, the different rising behaviour of oscillating and circulating droplets was taken into account.

Internal droplet recirculation is of crucial importance for the terminal velocity. This recirculation substantially affects the velocities near the droplet interface and hence the drag coefficient. The latter, in turn, results in a considerable change of the droplet terminal velocity. The effect of the internal recirculation on the terminal velocity has been considered, for example, by Calderbank and Korchinski [3] and by Horton et al. [4].

In all research works mentioned above, standard non-purified chemical systems were used. In later studies [5,6], purity of the investigated systems was higher, and higher terminal velocities were measured. This difference was found to be significant and could be attributed to the impurity influence which stabilised the interface and decreased the internal recirculation [7]. As a result, lower velocities near the interface were observed and this led to lower terminal velocities [8–11]. Similar effects were also observed in gas/liquid systems [12].

A commonly used correlation for the terminal velocity estimation of rigid droplets was developed by Pilhofer and Mewes [13]. Gourdon et al. [14] recommended a Morton-number (Mo) dependent combination of different correlations. For small Morton-numbers ($Mo < 10^{-11}$), droplets can be considered as rigid spheres and correlations similar to that by Pilhofer and Mewes [13] can be employed. For Morton-numbers in a range $10^{-11} < Mo < 10^{-7}$, Vignes law [15] is recommended, while for $10^{-7} < Mo < 10^{-5}$, the model published by Klee and Treybal [2] is preferred. For even higher Morton-numbers, $Mo > 10^{-5}$, the empirical correlation of Grace et al. [16] should be used. The latter correlation gives the most reliable results when it is applied for low surface tension

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systems (such as butanol/water). Critical reviews of these models were published by Steiner [17] and by Godfrey and Slater [18].

A more recent correlation for the droplet terminal velocity was published by Henschke [19]. It is a combination of two models, one of which describes the rising behaviour of rigid droplets, while the second one governs the behaviour of oscillating and deformed droplets. These two models are combined with the aid of a transfer function containing an adjustable parameter to determine whether the droplet behaves like a rigid or like an oscillating droplet. The whole model contains in total two constant and three adjustable parameters. For the estimation of these parameters, experimental data are necessary.

Wegener et al. [20] presented experimental results on transient droplet rise velocity and mass transfer. They worked with highly purified systems (toluene/water) and examined the influence of Marangoni convection on the terminal velocity. Without mass transfer (toluene/water system), the droplets with a diameter smaller than 2 mm remained spherical and in a short time (< 1 s) reached their terminal velocity. Droplets with a diameter larger than 2 mm accelerated to a certain pseudo-terminal velocity, but then started to deform. Due to this deformation, their rising velocity decreased and fluctuated around some lower velocity value.

Theoretical investigations of various problems in droplet-containing systems can be performed using the methods of computational fluid dynamics (CFD) that allow the simulation of multi-fluid flow in complex geometries and can handle phenomena at and around moving interfaces [21].

There are two general strategies dealing with moving interfaces. In the first one, a moving mesh is used to track the interface. As the topology of the interface changes, the mesh is deformed in accordance with the change. Methods falling under this strategy are called *front tracking methods*. The second approach uses a fixed (Eulerian) mesh, while the interface is tracked using different procedures, e.g. special markers or functions. Such methods are called *front capturing methods*.

In the front tracking methods, the interface is resolved directly. It always lies between two neighbouring mesh elements. As the interface moves, the neighbouring mesh elements are adjusted in order to track the movement of the interface. For the description of the interface movement, an additional explicit equation is necessary. Usually, the latter can be derived from an integral consideration of the transport phenomena around the interface. For instance, when the change of the topology of the interface is caused by the mass transfer, the total mass conservation condition can be used as an additional equation describing the mesh deformation [22]. Another possibility is given by relations describing physicochemical phenomena like condensation or boiling; they can also be used as an additional equation [23]. It is worth mentioning that, in the front tracking methods, each phase is considered separately, by using an individual set of equations describing the flow field. The front tracking methods are extremely accurate and robust yet rather complex to implement. Difficulties arise when two additional equations are needed to describe the mesh movement in three dimensions, because, in most cases, the second equation cannot be readily found in terms of the physical description of the system under study.

In the front capturing methods, the interface is described using special markers or indicator functions in a fixed mesh. Depending on the procedure used to capture the interface, they can be categorised in point-based or marker-based, surface capturing and volume capturing methods.

One of the oldest methods to describe interfacial phenomena is the marker particle method. In this method, massless particles are used to capture the motion of one fluid and thus the motion of the interface. Although its implementation is easy and it is capable of resolving the interface accurately, it fails to describe significant deformations of the interface. This is mainly because of the necessary redistribution of the markers, due to deformation or, in cases where the interface expands, the required addition of new particles. Besides, this method can be very demanding in terms of computational power and time for 3-D simulations.

One of the most popular surface capturing method is the level set (LS) method. In this method, an indicator function is used to locate the interface, which takes positive and negative values on different sides of the interface and zero at the interface. The interface is therefore called *zero level set*. An overview on the LS methods is presented by Osher and Fedkiw [24]. This method is conceptually simple and easy to implement. Its main drawback is the possible loss of mass (or volume), especially for significantly deformed interfaces [25].

The volume of fluid (VOF) method introduced by Hirt and Nichols [26] represents a typical volume capturing method. The basic idea of the VOF method is the definition of a volume function which takes values zero for the first phase, one for the second phase, and between zero and one for the cells containing the interface. A detailed review on the VOF method is published by Rider and Kothe [27]. This method is capable of handling problems with significant interface topology change and does not suffer from mass (or volume) losses. The extension of the method to 3-D simulations is straightforward and no special algorithm is needed for the case of merging or break-up of the interface. However, the interface is smeared out and merges “numerically”. Thus, up to now, it is not possible to use the VOF method for a rigorous analysis of the droplet or bubble coalescence probability [28]. The main drawback of this method is the inherent numerical smearing.

An alternative method is suggested by the group of Tryggvason [29,30]. They use a combination of both front tracking and front capturing methods, whereby a fixed grid is used to describe the motion of the fluid flow and another moving grid with a lower dimension is used to track the interface. Since for both phases a fixed grid is used, they are treated together, by solving a single set of governing equations for the whole flow field. The implementation of the hybrid method of Tryggvason is rather complex. The complexity of this method is due to the necessary dynamic remeshing of the moving sub-grid and the mapping of the data transferred from the moving grid to the fixed one. In addition, for the merging and breakage of the interface, a special sub-grid algorithm is required.

In most of the works on moving droplets found in the literature, only limited comparison between experimental and numerical results can be found. Quantitatively, only terminal velocities are compared.

In our work, a CFD model for the free interface movement is presented. This model is validated against results of a thorough experimental rising droplet investigation performed with a high-speed camera. Along with the terminal velocity comparison, a new method is presented for the quantitative comparison between experimental and numerical results in terms of the droplet shape, and an excellent agreement is found for both characteristics. Using the validated model, the effect of the internal droplet recirculation and the droplet shape on the droplet terminal velocity is addressed. This model can be further employed for the better understanding of other interfacial phenomena, e.g. interfacial mass transfer, Marangoni convection and droplet–droplet coalescence.

2. Experimental

To study the behaviour of rising droplets, the standard test system toluene/water, recommended by the European Federation of Chemical Engineers (EFCE) [31] is selected. The toluene/water system is most commonly used for testing the performance of liquid–liquid extraction columns and a large database is available in literature. The physicochemical properties of the selected system are given in Table 1.