International Journal of Heat and Fluid Flow xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

International Journal of Heat and Fluid Flow

journal homepage: www.elsevier.com/locate/ijhff

Numerical simulation of rising droplets in liquid–liquid systems: A comparison of continuous and sharp interfacial force models

Roland F. Engberg [a], Eugeny Y. Kenig [a,b,*]

[a] Chair of Fluid Process Engineering, University of Paderborn, Pohlweg 55, 33098 Paderborn, Germany
[b] Gubkin Russian State University of Oil and Gas, Moscow, Russian Federation

ARTICLE INFO

Article history:
Received 13 December 2013
Received in revised form 25 April 2014
Accepted 5 May 2014
Available online xxxx

Keywords:
Droplet
Liquid–liquid extraction
Terminal velocity
CFD
Level set
Interfacial force model

ABSTRACT

Simulations of single droplets rising in a quiescent liquid were performed for various initial droplet diameters. The continuous phase was water and the dispersed phase was either *n*-butanol, *n*-butyl acetate or toluene, thus resulting in three standard test systems for liquid–liquid extraction. For the simulations, a level set based code was developed and implemented in the open-source computational fluid dynamics (CFD) package OpenFOAM®. To prevent volume (or mass) loss during the reinitialisation of the level set function, two methods published recently were used in the code. The continuum surface force (CSF) model and the ghost fluid method (GFM) were applied to model interfacial forces, and their influence on grid convergence, droplet shapes and rise velocities was investigated. Grid convergence studies show a reasonable behaviour of the GFM, whereas the CSF model is less reliable, especially for systems with high interfacial tension. The results for droplet shape and terminal rise velocity are in excellent agreement with experimental and numerical investigations from literature. The onset of oscillations is correctly predicted, and the influence of the smoothing of interfacial forces on velocity oscillations is studied. Simulations of oscillating droplets remain stable, but the frequencies of the velocity oscillations differ from experimental results.

1. Introduction

Transport phenomena at moving boundaries are crucial in various unit operations of process engineering, e.g. condensation, absorption and liquid–liquid extraction. Therefore, moving boundaries have been studied extensively for several decades, both experimentally and theoretically, and remain subject of ongoing research. Mass transfer in liquid–liquid extraction columns occurs at moving boundaries of rising droplets. Thus, fluid dynamics of and mass transfer in droplet swarms are essential for the design of liquid–liquid extraction. To capture these complex phenomena properly, fluid dynamics of single moving droplets should be fully understood and mastered first.

Fluid dynamics of a single droplet is governed by physical properties of the system and the size of the droplet. Relatively small droplets remain spherical and reach their terminal rise velocity after a short period of acceleration. Increasing the droplet diameter leads to higher terminal velocities. Consequently, the forces acting upon the droplet grow and deform the droplet to an ellipsoidal or oblate shape. Droplets of even larger size begin to oscillate. Apart from the physical properties of both liquid phases, the terminal velocity also depends on the purity of the system. In pure systems, the interface remains mobile and the shear forces acting there lead to internal circulations, i.e. toroidal flow patterns in the droplet. In contrast, impurities accumulated at the interface inhibit the development of internal circulations. Thus, shear forces increase and the resulting terminal velocities are lower than in pure systems. For this reason, carefully purified systems were used in recent experimental investigations.

This work focusses on the numerical simulation of rising droplets. The most widely-used methods to simulate moving boundaries are moving mesh, front tracking and front capturing methods. Moving mesh methods (e.g. Bäumler et al., 2011; Tuković and Jasak, 2012) employ a boundary fitted grid which is moved to keep the grid nodes exactly at the interface. Thus, boundary conditions at the interface can be implemented directly, resulting in a very exact and sharp representation of the two-phase flow. Problems may arise when the interface undergoes significant topological changes, such as strong droplet deformations or the coalescence of two droplets. Front tracking methods (Unverdi and Tryggvason, 1992) describe the interface with a moving mesh

* Corresponding author at: Chair of Fluid Process Engineering, University of Paderborn, Pohlweg 55, 33098 Paderborn, Germany.
E-mail addresses: rolandengberg@gmx.de (R.F. Engberg), eugeny.kenig@upb.de (E.Y. Kenig).

http://dx.doi.org/10.1016/j.ijheatfluidflow.2014.05.003
0142-727X/

on a fixed grid. This approach is very accurate but requires dynamic restructuring of the interface mesh. Front capturing methods employ markers or indicator functions to locate the interface of a two-phase system. The movement of the interface is then described with a simple advection equation for the indicator function, which can be readily discretised with standard numerical methods, e.g. finite element methods (FEM) or finite volume methods (FVM). Hence, front capturing methods are robust and especially suitable for the simulation of strong interface deformations whereupon no time-consuming mesh movement or smoothing algorithms are necessary. While various front capturing methods have been proposed, the prevailing approaches of this category are the *volume of fluid* (VOF) method introduced by Hirt and Nichols (1981) and the *level set* method by Osher and Sethian (1988).

The interfacial forces are of utmost importance for the dynamic behaviour of moving boundaries. Consequently, the implementation of interfacial forces are crucial for the performance of a CFD code for two-phase flows, and various methods have been introduced. One of the earliest methods is the continuous surface stress (CSS) model of Lafaurie et al. (1994) and Gueyffier et al. (1999). Owing to its simplicity and robustness, the continuum surface force (CSF) model of Brackbill et al. (1992) is probably the most widely-used interfacial force model. By spreading the interfacial forces over a volume, the CSF model leads to a smooth transition in pressure at the interface. In contrast, the ghost fluid method (GFM) of Liu et al. (2000) and Kang et al. (2000) leads to a sharp pressure jump.

Dijkhuizen et al. (2005) used a front tracking model to simulate toluene droplets with initial diameters of 4–12.5 mm rising in water. Compared with the experimental data of Wegener et al. (2010) for the same system, velocity oscillations occured for larger diameters in the simulations and showed different amplitudes and frequencies. Moreover, the rise velocities were lower in the simulations than in the experiments of Wegener et al. (2010).

Wang et al. (2008) studied mass transfer from droplets in a *n*-butanol/water system. The moving boundary was described with the level set method and the interfacial forces were implemented with the CSF model. For one droplet diameter, numerical results for the rise velocity and a comparison of the simulated and experimental droplet rise height were shown.

Bertakis et al. (2010) published terminal velocities of *n*-butanol droplets rising in water obtained with the academic code DROPS (cf. Gross et al., 2006). This code employs a level set formulation of the two-phase flow discretised with finite elements; the interfacial forces are implemented with the CSF model. The results were in excellent agreement with the authors' own experimental data. However, the simulated rise time was rather short and covered only the onset of velocity oscillations.

Eiswirth et al. (2011) studied toluene droplets rising in water using the commercial finite-element code COMSOL Multiphysics 3.3a. The two-phase flow was described with a modified level set method suggested by Olsson and Kreiss (2005) which is, in fact, very similar to a VOF approach. The CSF model was used for interfacial forces. Comparing the terminal velocities with their own experimental results and those published by Wegener et al. (2007), Eiswirth et al. (2011) found very good agreement for small droplets (<1.5 mm) and large droplets (>3 mm) as well as satisfactory agreement for medium-size droplets. Moreover, experimental and simulated droplet shapes were in excellent accordance. However, the simulated rise time was too short to show oscillations which were, consequently, not discussed.

Bäumler et al. (2011) investigated the rise of single droplets experimentally and numerically. The simulations of three standard test systems for liquid–liquid extraction were carried out with moving mesh method implemented in the academic code NAVIER. The code uses finite element discretisation, and interfacial conditions, including the interfacial forces, are implemented with a subgrid projection method. Experimental results were taken from Wegener et al. (2010) (toluene/water) and Bertakis et al. (2010) (*n*-butanol/water). Additionally, experiments for *n*-butyl acetate droplets rising in water were performed. The agreement of numerical and experimental results was excellent for all systems and over the whole range of droplet diameters. However, oscillations of large droplets led to strong mesh deformation and, eventually, to a collapse of the simulations.

The results presented in this publication were obtained with a CFD-code based on a level set formulation of the two-phase flow. The code was implemented in the open source CFD package OpenFOAM® (Open Source Field Operation and Manipulation, www.openfoam.com, cf. Weller et al. (1998)). It includes two recent methods to overcome the volume loss frequently found in level set based methods. Simulations of rising droplets in three standard standard test systems for liquid–liquid extraction (*n*-butanol/water, *n*-butyl acetate/water and toluene/water) were performed with the continuous interfacial force model of Brackbill et al. (1992) and with the sharp model of Kang et al. (2000). The influence of the interfacial force model on grid convergence, droplet shape, rise velocity and the onset of oscillations was studied. Furthermore, the simulation results were compared with recent numerical and experimental investigations. In this context, a comparison with the precise boundary fitted moving mesh method of Bäumler et al. (2011) is particularly interesting.

2. Mathematical model and numerical method

The CFD code used for this publication was recently employed to study the influence of Marangoni convection on single rising droplets (Engberg et al., 2014a). In the following sections, the mathematical framework of the level set approach, the governing equations and the numerical details are outlined.

2.1. Level set method

The fundamental idea of the level set approach is to employ a function to indicate whether a point $\mathbf{x}$ is located in the continuous phase Ω_c or in the dispersed phase Ω_d. For this purpose, the indicator or level set function is defined as follows:

$$\psi(\mathbf{x},t) < 0 \quad \text{if } \mathbf{x} \in \Omega_d \tag{1}$$

$$\psi(\mathbf{x},t) > 0 \quad \text{if } \mathbf{x} \in \Omega_c \tag{2}$$

Thus, the interface $\partial\Omega = \partial\Omega_c \cap \partial\Omega_d$ is defined implicitly by the isocontour $\psi(\mathbf{x}_I) = 0$, where $\mathbf{x}_I$ is an arbitrary point on the interface.

In principle, any sufficiently smooth function could be chosen as a level set function. However, it has been shown that a *signed distance function* is a particularly advantageous choice (cf. Osher and Fedkiw, 2003). It has the following properties:

$$|\psi(\mathbf{x})| = \min(|\mathbf{x} - \mathbf{x}_I|) \quad \text{for all } \mathbf{x}_I \in \partial\Omega \tag{3}$$

$$|\nabla\psi| = 1 \tag{4}$$

The movement of an interface in a velocity field $\mathbf{v}$ can be described with the following advection equation for the level set function ψ:

$$\frac{\partial\psi}{\partial t} + \mathbf{v}\cdot\nabla\psi = 0 \tag{5}$$

Due to the advection, the level set function becomes distorted and loses its properties as a signed distance function (Eqs. (3) and (4)).

The procedure to restore ψ as a signed distance function is called reinitialisation and is realised by solving the following partial differential equation:

Please cite this article in press as: Engberg, R.F., Kenig, E.Y. Numerical simulation of rising droplets in liquid–liquid systems: A comparison of continuous and sharp interfacial force models. Int. J. Heat Fluid Flow (2014), http://dx.doi.org/10.1016/j.ijheatfluidflow.2014.05.003