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Numerical simulation of species transfer across fluid interfaces in free-surface flows using OpenFOAM

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ABSTRACT

This paper presents the Continuous-Species-Transfer (CST) method, which enables interface capturing techniques – a group among Computational Multi-Fluid Dynamics (CMFD) methods that rely upon a smooth interface representation – to deal with species transfer. In this study we examine realistic species transfer across fluid interfaces, taking into account both the steep interfacial concentration gradients (at high Schmidt numbers) and the sharp interfacial concentration jump (at high Henry coefficients due to different species solubilities). Thus, the main objective is to establish the CST method for species transfer across fluid interfaces of arbitrary morphology in free-surface flows at high viscosity and density ratios.

Detailed numerical simulations of single rising bubbles have been performed at high resolutions. Results were compared to experimental data and correlations derived thereof.

1. Introduction

Chemical reactors with bubbly flows are widely used in chemical reaction engineering for a variety of processes, e.g., bubble columns in the chemical, petroleum, metallurgical and energy industries. Common examples for the applications of bubble columns are liquid-phase oxidations, hydrogenations, chlorination, gas scrubbing, waste water treatment and various biotechnological applications.

One pivotal point of interest concerns the flow regimes in bubble columns, as this significantly influences the reactor's performance – its conversion, site time yield and selectivity. Though it is well-known that different flow regimes can be obtained by variation of the bubble column's operating parameters (such as superficial gas velocity, pressure, etc.) or design parameters (aspect ratio, sparger, internals, etc.), there is still a pivotal lack of detailed knowledge concerning the inherently complex nature of the underlying fluid dynamics in bubble columns: flow structures are intrinsically transient and characterized by very different spatial and temporal scales as depicted in Fig. 1.

It is therefore of major importance to develop both understanding and predictive simulation tools in order to obtain better and economically viable (efficient) technologies for process intensification and optimization of bubble column reactors.

For this purpose, a detailed understanding of the influence of mixing characteristics within the continuous liquid phase as well as of the dynamics of the dispersed gaseous phase upon *species transport within a phase* and *species transfer across phase interfaces* is crucial. This clearly becomes even more important for fast chemical reactions, where conversions take place in close vicinity to the bubble surface, greatly influenced by its surrounding local mixing pattern and corresponding diffusive and convective transport of the chemical species involved with these reactions.

In this work we perform detailed three-dimensional simulations covering the fluid dynamics and species transfer in single bubble systems. Emphasis is put upon the underlying physical background as well as the mathematical model and governing equations derived thereof (Sections 2 and 3), the basic framework of the employed solution methodology (Section 4), and detailed validation of both bubble dynamics and species transfer across its interface (Section 5).

2. Physical background

2.1. Bubble dynamics

In the past considerable research effort has been devoted to fluid dynamics in bubbly flows (bubble dynamics) focusing on the bubble's shape, wakes and velocities in various liquids

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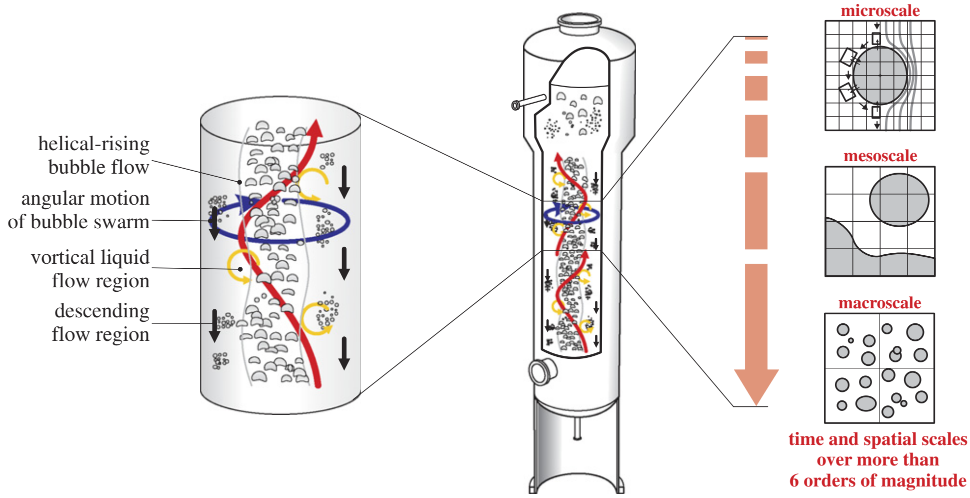

Fig. 1. Flow structures and physico-chemical phenomena in bubble column reactors.

(Habermann and Morton, 1953; Lindt, 1971, 1972; Grace et al., 1976; Clift et al., 1978; Brauer, 1979; Bhaga and Weber, 1981; Fan and Tsuchiya, 1990; Brücker, 1999; Tse, 2000).

From the wide base of experiments examining the rising of single bubbles in a quiescent liquid Clift et al. (1978) presented a diagram as shown in Fig. 2. This illustratively allows to distinguish among different bubble shape regimes depending on characteristic dimensionless numbers. These are the bubble Reynolds number $Re_b \equiv U_\infty d_b / \nu_l$ representing the ratio of inertia to viscous forces, the Eötvös number $Eo \equiv g(\rho_l - \rho_b) d_b^2 / \sigma$ representing the ratio of buoyancy to surface tension forces and the Morton number $Mo \equiv g(\rho_l - \rho_b)\eta_l^4 / \rho_l^2 \sigma^3$, which is defined by the ratio of viscous to surface tension forces. Note that this is not a complete set describing single bubble dynamics in a quiescent liquid. Dimensional analysis reveals two more dimensionless numbers, the density ratio $\Pi_\rho \equiv \rho_l / \rho_b$ and viscosity ratio $\Pi_\nu \equiv \nu_l / \nu_b$. Moreover, the system purity plays a major role. Furthermore, other dimensionless groups might be used in a complementary manner, i.e., $We = Re^2 \sqrt{Mo/Eo}$ or $Fr = \sqrt{We/Eo}$. However, it is common practice to base parametric studies upon this set of non-dimensional numbers, when examining the bubbles' shapes, rising velocities and/or trajectories and wake phenomena.

It seems evident that the interplay of these characteristics of bubbly flows and their underlying physical phenomena is utmost complex and cannot be considered decoupled. However, the above characteristics are useful for comparison with the bubble dynamics and the species transfer across the bubble's surface, and consequently serve as a validation base in this study.

Bubble shape. According to Fig. 2 bubble shapes can be generally categorized into three types, namely (1) spherical/ellipsoidal, (2) cap/skirted and (3) irregular/wobbling. As the bubble interfacial area (or its specific area defined as bubble interfacial area per bubble volume) characteristically varies among these shape regimes – which as a consequence has a significant influence on the overall species transfer across the bubble surface – it is advisable to examine bubbles that pertain to different regimes. Thus bubbles of different size and shapes were subject to this study.

Bubble rising velocity. Mainly depending on material properties and the purity of the system under consideration the terminal rising velocity of bubbles provides another feature to be considered. The terminal rising velocity significantly influences the overall contact time and mean residence time available for the species transfer. Thus, it must be captured correctly by the numerical simulations.

Fig. 3 shows the terminal rising velocities of bubbles within different shape regimes for the air/water system. This velocity mainly depends on the surface tension, the liquid phase viscosity and surfactants present in the system, each of which is influencing the bubbles' inner circulation, shape deformation

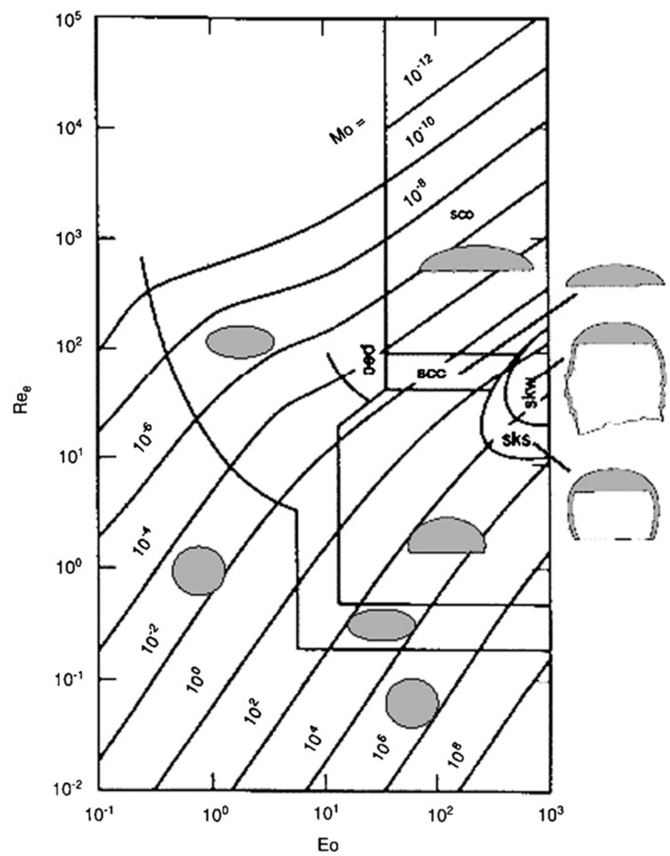

Fig. 2. Bubble shape depending on Reynolds, Eötvös and Morton number (Koynov and Khinast, 2006).