Abstract

In many process engineering applications, the mass transfer across interfaces plays a crucial role. When a gaseous phase and a liquid phase are brought into contact, the mass transfer often represents a decisive, or even the rate-determining step. It therefore determines the performance of processes like absorption, oxidation, hydration, carbonylation, and chlorination. The objective to design and optimize industrial relevant processes regarding yield and selectivity makes multiphase flows an important research field. The scientific question of this study is how vortex structures influence mass transfer and local processes at the gas-liquid interface. For this purpose, experiments within a bubbly flow, with a rising bubble pair, and with a single fixed bubble are carried out. The measurement techniques used are Particle Image Velocimetry (PIV) to obtain velocity fields, and Laser-Induced Fluorescence (LIF) to obtain concentration fields. This study contributes to the scientific progress by means of providing detailed observations regarding hydrodynamics and mass transfer close to the interface. Experimental results as well as modeling is presented and discussed concerning momentum transporting vortices and their effect on boundary layer dynamics.

To identify and analyze momentum transporting vortices within bubble wakes, experiments within a bubbly flow are carried out. Continuing experiments with a freely rising bubble pair utilizing a novel scanning technique (TRS-PIV) make it possible to track the rising paths and the vortices induced by the rising bubbles in a quasi three-dimensional way. Since the aim of this study is to make detailed observations over a longer period of time, the findings of the preceding experiments are utilized to design a set-up with a single bubble within a duct flow. The bubble is fixed in place and impinged by momentum transporting vortices generated by a cylinder. Via these simplifications, it is possible for the first time to observe tangential velocity profiles close to the interface, as well as local boundary layer dynamics. By means of this methodical series of experiments, the impact of momentum transporting vortices on hydrodynamics and mass transfer is analyzed.

There is a significant influence of the momentum transporting vortices downstream the cylinder on the flow structure close to the bubble. In particular, the velocity fluctuations close to the interface and in the bubble wake are intensified. By evaluating instantaneous velocity fields, it is found that an inner shear layer is formed between cylinder and bubble. The vortices move along this shear layer; and they have to overcome it to directly approach the bubble. The evaluation of the concentration fields reveals that the point of vortex shedding is shifted downstream due to the vortices. The boundary layer fluctuations increase significantly and they are influenced to a larger extent by the cylinder wake than by the mean flow velocity. However, the global mass transfer from the whole bubble into the liquid is not significantly changed by the vortices. This leads to the conclusion that, in analogy to the hydrodynamic boundary layer, the concentration boundary layer can be sub-divided into a viscous sublayer and a flexible layer. While the viscous sublayer, which is the limitation for diffusive processes, is not influenced by the vortices, the vortices affect the flexible part. As a result, the cross-mixing within the bubble wake is enhanced.

Zusammenfassung

In vielen verfahrenstechnischen Anwendungen spielt der Stoffaustausch über Phasengrenzen hinweg eine wichtige Rolle. Wenn eine gasförmige und eine flüssige Phase in Kontakt gebracht werden, stellt der Stofftransport oftmals einen entscheidenden oder sogar den geschwindigkeitsbestimmenden Schritt dar und bestimmt so die Performance von industriellen Prozessen wie Absorption, Oxidation, Hydrierung, Carbonylierung und Chlorierung. Mehrphasenströmungen stellen somit einen wichtigen Forschungsbereich dar, um industriell relevante Prozesse hinsichtlich Selektivität und Ausbeute auszulegen und zu verbessern. Die wissenschaftliche Fragestellung dieser Arbeit ist, wie Wirbelstrukturen den Stofftransport und lokale Phänomene an der Gas-flüssig-Phasengrenze beeinflussen. Zu diesem Zweck werden Experimente in einer Blasenströmung, an einem Blasenpaar und an einer fixierten Einzelblase durchgeführt. Die zum Einsatz kommenden Messtechniken sind die Particle Image Velocimetry (PIV) zur Bestimmung von Geschwindigkeitsfeldern sowie die Laser-Induzierte Fluoreszenz (LIF) zur Bestimmung von Konzentrationsfeldern. In dieser Arbeit werden experimentelle Ergebnisse und Modellierungen zu impulstransportierenden Wirbeln und ihrem Einfluss auf Grenzflächendynamiken präsentiert. Mit detaillierten Betrachtungen zu Hydrodynamik und Stofftransport nah der Phasengrenze trägt diese Arbeit zum grundlegenden Verständnis von Prozessen an fluiden Phasengrenzen bei.

Zur Identifikation und Analyse Impuls transportierender Wirbel in Blasennachläufen werden Experimente an einer Blasenströmung durchgeführt. In weiterführende Untersuchungen an einem frei aufsteigenden Blasenpaar gelingt die Nachverfolgung von Blasenaufstiegswegen und Wirbelstrukturen im Nachlauf. Hierzu kommt erstmals eine neuartige Scanning-Technik (TRS-PIV) zum Einsatz, die quasi-dreidimensionale Informationen liefert. Da das Ziel dieser Arbeit ist, detaillierte Betrachtungen über einen längeren Zeitraum durchzuführen, werden die Erkenntnisse der vorangegangenen Experimente genutzt, um einen Aufbau mit einer einzelnen Blase in einer Rohrströmung zu konzipieren. Die einzelne Blase ist örtlich fixiert und wird definiert erzeugten Wirbeln ausgesetzt, die durch die Umströmung eines Zylinders entstehen. Mit Hilfe dieser Vereinfachungen ist es erstmals möglich, tangentiale Geschwindigkeitsprofile nahe der Phasengrenze und lokale Grenzschichtdynamiken zu beobachten. Die methodische Reihe an Experimenten in dieser Arbeit liefert einen sys-

tematischen Ansatz zur Analyse des Einflusses von Impuls transportierenden Wirbeln auf Hydrodynamik und Stofftransport.

Der Einfluss der Wirbel auf die Strömungsstruktur nahe der Blase ist signifikant. Insbesondere die Geschwindigkeitsfluktuationen nahe der Blasenoberfläche sowie im Blasennachlauf werden durch die Wirbel beeinflusst. Durch die Auswertung momentaner Strömungsfelder wird festgestellt, dass sich eine Scherschicht zwischen Zylinder und Blase ausbildet. Entlang dieser Scherschicht bewegen sich die Wirbel; sie muss von den Wirbeln überwunden werden, um in direkten Kontakt zur Blase zu treten. Die Auswertung der Konzentrationsfelder zeigt, dass der Ablösepunkt der Konzentrationsgrenzschicht durch die Wirbel in Richtung der Blasenrückseite verschoben wird. Die Fluktuationen der Grenzschichtdicke nehmen erheblich zu und werden durch den Zylindernachlauf stärker beeinflusst als durch die anliegende Strömungsgeschwindigkeit. Der globale Stofftransport der gesamten Blase wird jedoch nicht signifikant durch die Wirbel beeinflusst. Dies führt zur Annahme, dass, ähnlich der hydrodynamischen Grenzschicht, auch die Konzentrationsgrenzschicht in eine viskose Unterschicht und eine flexible Schicht eingeteilt werden kann. Während die viskose Unterschicht, die die Limitierung für die Diffusion darstellt, durch die Wirbel nicht beeinflusst wird, wirken die Wirbel auf den flexiblen Teil der Grenzschicht sehr stark ein und erhöhen dadurch die konvektive Durchmischung im Blasennachlauf.

Chapter 1

Introduction

Transport processes with respect to momentum, heat, and mass transfer play a major role in multiphase flows. Since multiphase flows occur in countless applications like in chemical, pharmaceutical, biological and food industry as well as in waste water treatment wide-ranging knowledge concerning these transport processes in multiphase systems is necessary. When a multiphase system consists of a dispersed gas phase and a continuous liquid phase, there exists a fluidic interface between the bubble and the bulk phase. Decisive physical and chemical events happen close to this interface, and thus, this is the region of very high experimental interest. The small-scale events, like the mass transfer across the fluidic interface, will determine large-scale parameters that influence efficiency, reactor design, residence times, and many more important factors. When it comes to reactor design for bubble columns, macroscopic experiments are conducted and empirical correlations are used to estimate reference points for the choice of the reactor. However, basic knowledge about hydrodynamics and mass transfer close to the interface is necessary to not only estimate, but rather understand transport phenomena.

The scope of this work is to contribute to the scientific question how hydrodynamics and mass transfer is coupled in multiphase systems and how they are influenced by vortex structures. For this purpose, a methodical series of experiments is conducted. In a first set-up, a bubbly flow is investigated utilizing Particle Image Velocimetry (PIV). Within this set-up, a method to detect and analyze momentum transporting vortices is implemented. To gain deeper insight into the interaction of bubbles and bubble wakes, a second set-up is designed in which one or two rising bubbles can be investigated carefully. Videography is used to characterize bubble trajectories, and via Time-Resolved Scanning PIV, which has been conducted for the first time, quasi-3D information of rising bubbles and occurring vortices is gained. It is analyzed for which parameters an interaction of vortex structures with bubbles is observable. With the findings from these two set-ups, a third set-up is implemented

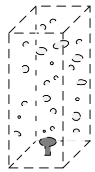
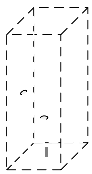
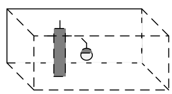
	pictogram	some phenomena occuring	optical access	experimental campaigns conducted	methodical series	reference to reality
n bubbles		- mass transfer - bubble size distribution - bubble-induced turbulence - wake interaction - rising paths - bubble bouncing - coalescence and break-up	very challenging	PIV		
2 bubbles		- mass transfer - wake interaction - rising paths - bubble formation	challenging	TRS-PIV		
1 bubble		- mass transfer - wake interaction	good	Video-graphy PIV LIF		

Table 1.1 Methodical approach of this study.

to make very detailed experiments on single fixed bubbles and to describe small-scale events at the interface. Here, Videography, PIV, and Laser-Induced Fluorescence (LIF) can be conducted precisely, but at the expense of reference to reality which is presented in table 1.1. By using the very well characterized flow around a circular cylinder, artificial wakes are created which impinge the single bubble held in place and in shape by a spherical cap. With this set-up, investigations very close to the fluidic interface are possible.

Chapter 2

State of the art and basic knowledge

2.1 Gas-liquid flows and transport phenomena

2.1.1 Gas-liquid flows and bubble columns

In many industrial processes, a dispersed gas phase and a continuous liquid phase are brought into contact to achieve process steps like absorption, oxidation, hydration, bioreactions, coal liquefaction, and many more (examples are listed e. g. in [Sha82]). For this purpose, reactors are designed to make a large contact area between the phases, high throughput, and preferably low costs possible. Bubble columns have the advantages of a simple design without installations needed, of high liquid residence time, and of a good mixing [Sha82]. They can be run in co- or counter-current flow as well as under elevated pressure [Rol15]. There are different construction methods which are described in [Dee12]. Basic knowledge on bubble columns can be found in [Dec77], [Dec85]. The disadvantage of this type of reactor is that the hydraulic conditions are difficult to control, which makes the scale-up and predictability of reactor performance difficult [Bot13]. Depending on the superficial gas velocity and the diameter of the column, different flow regimes can be obtained: homogeneous bubbly flow, heterogeneous bubbly flow, and slug flow [Sha82]. Within the heterogeneous regime, the axial mixing effect is remarkably higher than within the homogeneous regime [Zah96]. The behavior of bubbles within bubbly flows is influenced by bubble interactions, like coalescence, break-up, bouncing, and bubble-induced turbulence. While the first points only refer to the dispersed phase, the latter is about the influence of bubble interactions on the bulk phase flow. The bubble motion is found to influence the energy spectrum of the liquid flow [The82], [Lan91], [van98], and it is still a subject of current research [Rib13], [Haa17].

To estimate the mass transfer in bubble columns, empirical correlations are used. There are many correlations, which can be found in literature [Aki74], [Hon84], taking into account the flow regime by means of the Reynolds number

$$Re = \frac{u \cdot L_{char}}{\nu} \tag{2.1}$$

and the material properties by means of the Schmidt number

$$Sc = \frac{\nu}{D} \tag{2.2}$$

with u depicting the bubble velocity, the characteristic length L_{char} being the mean diameter of the fluid particles for which the Sauter diameter is used most of the times, ν depicting the kinematic viscosity, and D being the diffusion coefficient of the transferred species. The Sherwood number, which can be understood as a dimensionless mass transfer coefficient and is discussed in detail in section 2.1.2, is a function of the Reynolds number and the Schmidt number.

However, these correlations do not reveal the underlying principles of mass transfer, which are crucial to know for a more aimed design and scale-up of bubble columns. Furthermore, the role of the bubble wake is considered to be significant for turbulence effects [Brü99]. Therefore, investigations on single bubbles are still necessary to get fundamental knowledge about the transport processes at the gas-liquid interface.

2.1.2 Single rising bubbles

Bubble shape and rising velocity

The behavior of single rising bubbles differs from the behavior of bubble swarms since there is no interaction of bubbles leading to wake effects, bubble induced turbulence, coalescence or break-up. When a single bubble is released, a terminal rise velocity v_t occurs after a certain initial period. This velocity is dependent on the bubble shape, initial shape deformations, and material system [Tom02]. The interdependency of bubble shape, interfacial effects, rising velocity, and rising path is complex, since they are inextricably linked to each other. To make statements concerning the shape regimes for bubbles and drops, three dimensionless numbers are used, Eötvös

$$Eo = \frac{(\rho_c - \rho_d) \cdot g \cdot d_b^2}{\sigma}, \tag{2.3}$$

Reynolds

$$Re_b = \frac{v_t \cdot d_b \cdot \rho_c}{\eta_c}, \tag{2.4}$$

and Morton number

$$Mo = \frac{g \cdot \eta_c^4 \left(\rho_c - \rho_d\right)}{\rho_c^2 \cdot \sigma^3}. \tag{2.5}$$

ρ_c depicts the density of the continous phase, ρ_d the density of the dispersed phase, g is the constant of gravitation, d_b is the bubble diameter, σ is the interfacial tension, v_t is the terminal bubble velocity, and η_c is the dynamic viscosity of the continuous phase. For d_b, the volume equivalent diameter of a sphere

$$d_b = \left(\frac{V_b \cdot 6}{\pi}\right)^{1/3} \tag{2.6}$$

is used. The Reynolds number depicts the ratio of the inertial force to viscous force, the Eötvös number depicts the ratio of gravitational force to surface force, and the Morton number is only dependent on the material system and depicts the ratio of viscous force to surface tension. From these three numbers, the shape of the bubble can be estimated using the well-known diagram by Clift et al. [Cli78] (figure 2.1: spherical without (A in figure 2.2) and with (B) inner circulation, ellipsoidal (C), cap shaped, or irregularly shaped (D)). From a

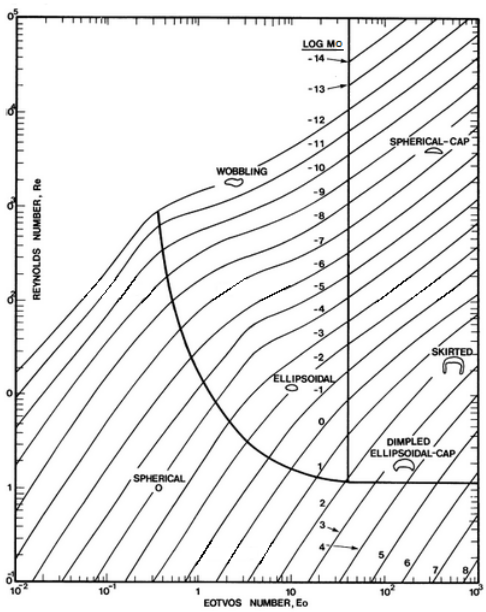


Fig. 2.1 Shape regimes for rising bubbles, [Cli78]

balance of forces acting on a bubble which is not accelerated, the general equation for the bubble rise velocity is obtained, which can be set equal to the terminal rise velocity

$$v_b = v_t = \sqrt{\frac{4}{3} \cdot \frac{|\rho_d - \rho_c|}{\rho_c} \cdot g \cdot d_B \cdot \frac{1}{C_D}} \tag{2.7}$$

assuming that a steady state is reached. From equation 2.7, it becomes clear that the bubble rise velocity v_b is dependent on the drag coefficient C_D

$$C_D = \frac{F_D}{\frac{1}{2}\rho_c \cdot v_b^2 \cdot A_P}, \tag{2.8}$$

which is defined as the ratio of the drag force F_D to the dynamic pressure and the projection area A_P. For low Reynolds numbers, $Re < 1$, when Stokes' law

$$F_D = 3 \cdot \pi \cdot d_B \cdot \eta_c \cdot v_B \tag{2.9}$$

is applicable, the relation between the Reynolds number and the drag coefficient can be described explicitly. It is for rigid particles

$$C_D^{rigid} = \frac{24}{Re_p}. \tag{2.10}$$

For bubbles, the drag coefficient is dependent on the shape regime (A to D in figure 2.2, superscripts in the following equations). Hadamard and Rybcynski [Had11], [Ryb11] were the first who considered the drag coefficient for bubbles based on Stokes' law, however, neglecting the inertial terms in the equations of motion. Levich [Lev62] proposed the expression

$$C_D^A = \frac{48}{Re_b}, \tag{2.11}$$

which is based on the boundary layer theory. For higher Reynolds numbers, when the shape of fluid particles is controlled by the outer flow, the drag coefficient C_D varies depending on the bubble shape. Koebe [Koe04] gives a carefully researched overview of equations for the drag coefficient according to the bubble shape regimes. For regime B in figure 2.2, left, he cites the work of Maxworthy et al. [Max96]

$$C_D^B = \frac{11.1}{Re_b^{0.71}} \tag{2.12}$$