

## How fundamental knowledge on mass transfer in bubbly flows will help process intensification

**Citation for published version (APA):**

Baltussen, M. W. (2023). How fundamental knowledge on mass transfer in bubbly flows will help process intensification. *Chemical Engineering and Processing : Process Intensification*, 187, Article 109344. <https://doi.org/10.1016/j.cep.2023.109344>

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**DOI:**  
[10.1016/j.cep.2023.109344](https://doi.org/10.1016/j.cep.2023.109344)

**Document status and date:**  
Published: 01/05/2023

**Document Version:**  
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

**Please check the document version of this publication:**

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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# Chemical Engineering and Processing - Process Intensification

journal homepage: [www.elsevier.com/locate/cep](http://www.elsevier.com/locate/cep)

## How fundamental knowledge on mass transfer in bubbly flows will help process intensification

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### ARTICLE INFO

#### Keywords:

Bubbly flow  
Mass transfer  
Mixing  
Process intensification

### ABSTRACT

We are in danger of losing our current standards of living due to the depletion of natural resources. It is essential to decrease the amount of waste produced. In reactor engineering, the waste production is caused by the formation of byproducts. To increase the selectivity of the reactor, the fluid dynamics should benefit the intrinsic reaction kinetics, which can only be done when each molecule has exactly the same experience. The mass transfer from a bubble is generally assessed solely by the overall mass transfer coefficient from the bubble. However, the concentration profile is far from uniform when the mass is transferred from the bubble. The obtained concentration profile seems to be confined by the hydrodynamic wake of the bubble. The obtained structures are quite stable and not easily disturbed by other bubbles. This leads to large concentration gradients in the bulk, which prevents the molecules in the liquid to have the same or a similar experience in the reactor. As this will have a large impact on the selectivity when consecutive reactors are considered. It would be essential in the future to study these local mixing profiles in bubbly flows in particular and in multi-phase flows in general.

To ensure that future generations can also have the current standards of living, we have the responsibility to decrease our environmental impact. As the consumption of natural resources, e.g. fossil fuels and minerals, has increased drastically since the industrial revolution. The depletion of some of these resources has been predicted in the next decades [1]. The most prominent case is the consumption of fossil fuels. Besides the depletion of these resources, the usage also increased the CO<sub>2</sub> emissions drastically leading to the greenhouse effects. The reason for the depletion is that the deposition of the minerals is slower than our current consumption rates.

To reduce the depletion of our natural resources, it is essential to ensure that the cycles for all natural resources are closed. This would imply that all materials need to be recycled or reused, leading to a circular economy. However, this can only be obtained when the production processes are selective to the desired products and suppress the formation of any side-products. This high selectivity is also one of the goals of process intensification [2–4]. Therefore, process intensification can be considered as an important tool for the circular economy and by that a more sustainable chemical industry.

The highest environmental impact of process intensification is expected to be in the development of reactor designs as these determine the amount of byproduct formation, i.e. lets ensure every molecule counts. To improve the reactor performance, the reactor design should ensure that the fluid dynamics inside the reactor exploit the intrinsic

reaction kinetics [3,5]. This can only be the case if every molecule has exactly the same experience in the reactor [4].

These improvements are generally created by either process intensification methods or process intensification equipment. First of all, the process intensification methods focus among others on improving the processes by combining functionalities, e.g. multifunctional reactors and hybrid separations, or the use of alternative energy sources, e.g. solar energy or ultrasound. [4]. In the process intensification equipment category, the current unit operations are changed to improve mass transfer or heat transfer.

Bubbly flows are often encountered in mineral processing, chemical and food processing industries [6]. Therefore, this paper will specifically focus on bubbly flows. In bubble columns, the bubbles are either used for mixing the liquid phase, or they are used to contact the gaseous reactant with the liquid reactants. In this second type, the mass transfer from the bubble to its environment is essential for the performance of the reactor. The current process intensification options for gas–liquid flows focuses on the creation of a large interfacial area and the creation of a plug flow like behavior in the reactor.

For example, Taylor bubbles are created in tubular microreactors. The confinement of the bubble elongates the bubble in the flow direction, which increases the interfacial area. In addition, the bubble and its

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<https://doi.org/10.1016/j.cep.2023.109344>

Received 7 April 2022; Received in revised form 8 March 2023; Accepted 11 March 2023

Available online 20 March 2023

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**Table 1**

Sherwood ( $Sh = kd_b/D$ ) correlations for spherical bubbles with their applicability.  $Re$  is the Reynolds number ( $Re = \frac{\rho_l v_b d_b}{\mu_l}$ ),  $Sc$  the Schmidt number ( $Sc = \frac{\mu_l}{D \rho_l}$ ),  $Pe$  the Peclet number ( $Pe = Re Sc$ ),  $\rho_l$  the liquid density,  $\mu_l$  the liquid viscosity,  $v_b$  the bubble rise velocity,  $d_b$  the bubble diameter and  $D$  the diffusion coefficient.

Reference	Correlation	Validity
Colombet et al. [11]	$Sh = 1 + \left(1 + \left(\frac{8}{3\pi} Pe_{max}\right)^{2/3}\right)^{3/4}$ with $Pe_{max} = \frac{Pe}{2} \frac{16+3.315\sqrt{Re+3Re}}{16+3.315\sqrt{Re+Re}}$	All $Re$ and $Pe$
Leclair and Hamielec [12]	$Sh = (0.65 + 0.06\sqrt{Re}) \sqrt{Pe}$	$Re < 10$
Feng and Michaelides [13]	$Sh = 0.651 \sqrt{Pe} \left(1.032 + \frac{0.61Re}{Re+21}\right) + \left(1.60 - \frac{0.61Re}{Re+21}\right)$	$Re > 1$
Takemura and Yabe [14]	$Sh = \frac{2}{\sqrt{\pi}} \left(1 - \frac{2}{3} \frac{1}{(1+0.09Re^{2/3})^{3/2}}\right)^{1/2} (2.5 + \sqrt{Pe})$	$Re < 100, Pe > 1$
Winnikow [15]	$Sh = \frac{2}{\sqrt{\pi}} \sqrt{Pe} \left(1 - \frac{2.89}{Re^{1/2}}\right)^{1/2}$	$Re > 50$
Potential flow [16]	$Sh = \frac{2}{\sqrt{\pi}} \sqrt{Pe}$	$Re \rightarrow \infty$

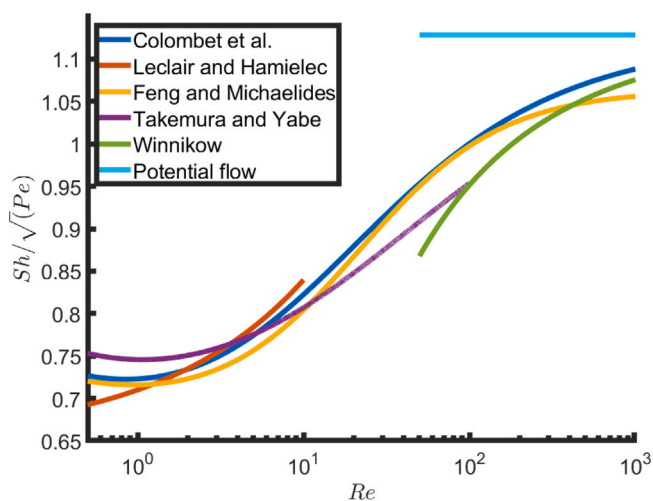


Fig. 1. A selection of the available Sherwood correlations for spherical bubbles found in Table 1 [11–16].

surrounding liquid move as a single plug through the reactor. Although this improves the similarity between several bubbles in the reactor, it does not imply that the experience of each molecule in the reactor is similar, which is clearly visible in the wake structures reported by Kexel et al. [7,8]. However, these local effects of the mass transfer and its interaction with the wake are poorly understood, while these are essential for the further intensification of reactors using bubbly flows. Therefore, this paper will discuss the current missing links in our understanding of mass transfer from bubbles. This effort will be split into two main parts: the overall mass transfer coefficients for bubbly flows and the micro-mixing in the fluid. Besides a small discussion on the effects for process intensification operations will be discussed.

## 1. Mass transfer coefficients

If we want improve the mass transfer rate from the gas-phase to the liquid, it is essential that we can determine the mass transfer coefficient from a bubble to its environment. As bubbly flows are often encountered, one might expect that for the most simple case, an unconfined single rising bubble in a clean liquid, the mass transfer is readily available. In literature, several correlations can be found, which are based on either experimental results, theoretical derivations, simulations or a combination of these. For all of these correlations, it should be noted that they have a limited range of applicability [9,10].

For spherical bubbles, a selection of the available correlations is shown in Fig. 1. The figure clearly shows that the Sherwood numbers predicted by the correlations are within 10%, which is the error margin of most of the correlations. From this it could be concluded that the mass transfer from a single spherical bubble is known. However, the spherical bubbles are generally small and therefore not that relevant in the current process industry. If the bubble size is increased, the bubble path instabilities occur and the bubble shape changes to wobbling, spherical cap or skirted. For these bubbles, mass transfer correlations are also created, but comparing these might result in differences up to 100% [10]. Concluding for a single bubble the Sherwood number can be predicted as long as the bubbles are relatively small, while for larger bubbles it is difficult to predict the overall mass transfer coefficients.

In addition, it would be essential to understand how the current mass transfer correlations are obtained. First of all, some correlations are based upon theoretical derivations in either potential flow or creeping flow, which is generally not the case for rising bubbles. The other correlations are based on experimental results. To determine the amount of mass that is transferred two methods are used: bubble shrinking or interaction with a dye. Both methods have problems in accurately determining the mass transfer coefficient.

To understand the problems with the problems regarding the experimental measurements, the species transport in the liquid can be described using an advection diffusion equation:

$$\frac{\partial c}{\partial t} + (\mathbf{u} \cdot \nabla) c = D \nabla^2 c + \phi_{gas \rightarrow liquid} - R \quad (1)$$

where  $c$  is the concentration,  $t$  the time,  $\mathbf{u}$  the velocity,  $D$  the diffusion coefficient,  $\phi_{gas \rightarrow liquid}$  the flux of the transferred component from the gas to the liquid and  $R$  the amount of the component consumed by a reaction. The mass transfer from the gas phase to the liquid phase is generally located in the liquid phase and can be expressed by:

$$\phi_{gas \rightarrow liquid} = k(Hc_{gas} - c)A_{interface} \quad (2)$$

where  $A_{interface}$  is the gas–liquid interfacial area and  $H$  the Henry coefficient.  $k$  is the mass transfer coefficient, which is also used in the Sherwood numbers ( $Sh = \frac{k d_b}{D}$ ) in the correlations of Table 1.

When using the bubble size for the determination of the mass transfer, the amount of mass transferred from the bubble is measured by the decrease in the bubble size. However, the problem with this method is that the measurements of bubble size assume that the bubble size is only changed due to the mass transfer from the gas phase to the liquid phase, which might be the case for the component in the gas phase initially. However, any dissolved gas in the liquid phase also behaves according to Eq. (1). When the concentration in the liquid is higher than the concentration in the gas phase times the Henry coefficient, it should be noted that the flux is negative (i.e. from the liquid phase to the gas phase) as given in Eq. (2). As it is generally difficult to ensure that

the liquid phase is free of any dissolved gas components, this counter-diffusion can never be fully prevented in experiments resulting in large uncertainties. In addition, the determination of the size of the bubble is difficult when non-axi-symmetric bubbles are measured.

When using dyes to determine the mass transfer, the dye changes color due to a reaction with the transferred molecules. This leads to two problems with this method. First of all, the coloring of the dye depends on the local concentration, which means that the color intensity has to be calibrated. In addition, the local concentration in the liquid of the transferred component will decrease due to the reaction in the liquid. This will result in a relatively larger driving force in the flux (Eq. (2)) compared to the visualized concentration in the liquid. Therefore, the obtained mass transfer rates are higher than the gas to liquid mass transfer rates, which are not representative of the pure gas to liquid mass transfer.

To further improve our reactors, it is essential to predict the mass transfer from a bubble accurately. As the discussion above shows, the current knowledge on mass transfer from single bubbles is limited. If bubble swarms or clusters are considered, there is only a limited understanding of the effects on the mass transfer, as they are obtained by simplified experiments of interacting bubbles [17] or simulations at low Schmidt numbers [18]. Therefore, the mass transfer across the gas-liquid interface should be of key interest. However, even if this mass transfer could be predicted, the same route for every molecule can only be ensured if we have perfect micro-mixing in the liquid phase.

## 2. Micro-mixing in the fluid

Generally, it is assumed that as soon as mass is transferred from the bubble to the liquid phase the molecules are equally distributed in the liquid surrounding the bubble especially for bubbles in a bubble swarm. In this section, this distribution of the concentration in the liquid is discussed in more detail for both single bubbles and interacting bubbles.

A contour plot for the mass transfer from a spherical bubble and a wobbling bubble is shown in Fig. 2. The figure shows that most of the mass transfer occurs at the nose of the bubble, because the local concentration gradient is relatively high at the nose. However the obtained concentration profile in the wake is clearly different for both types of bubbles [7,8,10,17]. As the main transport mechanism for the component is convective transport, the difference occurs due to the different wake structure, i.e. spherical bubbles have a closed wake while larger bubbles generally show unsteady wakes with vortex shedding [19,20].

Upon closer examination, the concentration profiles both show that the recirculation pattern in the wake of the bubble is confining the transferred components, which is clearly visible in the comparison of the velocity profile and the concentration profile in Fig. 3. It is expected that the micro-mixing of the transferred component is less for a spherical bubble. However, the concentration profile of the larger bubble shows a far from uniform concentration distribution. If realistic reaction mechanisms are considered including consecutive reactions, this non-uniform profile will lead to a lower selectivity. In general this will be problematic in all cases in which the time scales for mass transport are comparable to the time scales of the reaction or when the mass transfer is considerably faster than the reaction [8,22].

In general a large scale circulation would be required to break these structures in the wake of a single rising bubble [17,22]. These large scale circulations can be obtained by adding more bubbles to the system. However experiments with two interacting bubbles show that the structures of the concentration wakes are maintained. When the amount of interactions is increased in a bubble train, the same structures are still visible, meaning that the trailing bubbles do not sufficiently disturb the concentration profiles created by the preceding bubbles [23]. Similar results are obtained numerically for a bubble swarm by Roghair et al. [18].



Fig. 2. Visualizations of the concentration profile for a spherical bubble (Eötvös number of 1, Morton number of  $10^{-3}$  and Peclet number of  $10^6$ ) and a wobbling bubble (Eötvös number of 10, Morton number of  $10^{-11}$  and Peclet number of  $9 \cdot 10^5$ ) on the left and right, respectively. The concentration value increases from yellow to orange and the blue contour is the bubble.

Source: Reproduced from Claassen et al. [10].

## 3. Intensification of the mass transfer

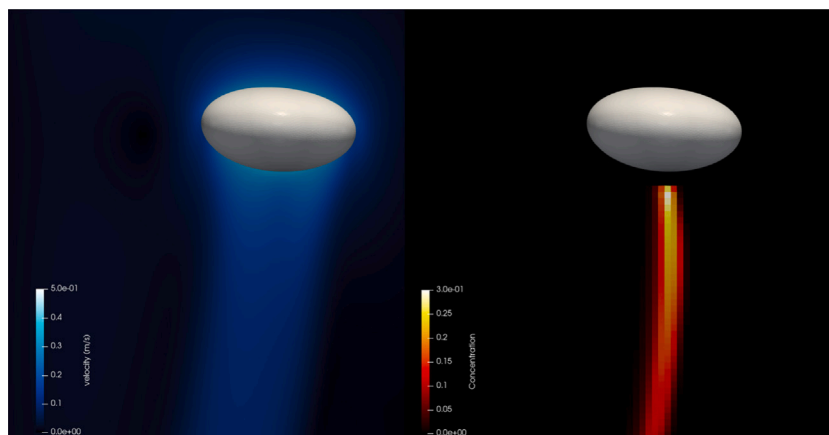
Although the mass transfer from a bubble to the liquid has been studied in large detail, the local mixing surrounding a bubble is largely unknown. As stated above the main research is focused on a single bubble or a small cluster of bubbles. In dense bubbly flows, the available information is limited to flows with low Schmidt numbers. The current research efforts regarding the mass transfer in process intensification are generally focused solely on increasing the mass transfer. However this might not be sufficient to enable tuning the selectivity of the reactor towards one single product.

For Taylor bubbles in a microreactor, there are some experimental results for the local mixing behind a Taylor bubble [7,8]. These results show that there are large local concentration gradients in the wake of the Taylor bubble. When comparing experiments with only physical absorption and a reaction, it is clear that the reaction in the bulk enhanced the mass transfer from the bubble. In addition, they showed that in the case of a consecutive reaction the time scale of mixing in the bulk compared to the reactive timescale is important. Improving the mixing in the vicinity of the bubbles will ensure that the selectivity towards a certain product is only dependent of the residence time, enabling tuning the reactor to a certain product.

## 4. Conclusion

In process intensification, the goal is to reduce the amount of waste create in products. An important way to decrease the amount of waste is the improvement of the selectivity of the reactor, which is largely dependent on the hydrodynamics and the mass transfer in the reactor. The discussion above shows that the mass transfer in simple gas-liquid flows are not even fully understood. When increasing the complexity of the system by path instabilities or interactions with other bubbles, it is difficult to find a representative mass transfer coefficient.

In addition, the discussion also shows that the selectivity is also largely dependent on the mixing in the vicinity of the bubble. The local wake structures largely determine the effective mixing of the transferred components to the bulk of the liquid, i.e. the local recirculation patterns seem to confine the transferred components to the wake of the bubble.



**Fig. 3.** Visualizations of the velocity profile (right) and concentration profile (left) for a wobbling bubble (Eötvös number of 1, Morton number of  $10^{-9}$  and Peclet number of  $2.9 \cdot 10^3$ ). It should be noted that the concentration in the vicinity of the bubble cannot be captured with the current method, which uses the subgrid model near the gas-liquid interface [21].

To proceed with a further intensification of bubbly flows, it is thus essential to study the mass transfer coefficients from the bubble to the liquid as well as the local mixing in the liquid. Only if the time scales for mixing are lower than the time scales for the reaction, the selectivity towards a certain product can be steered in any direction.

This work focused solely on the mass transfer from bubbles to their environment and a reaction in the bulk. However, the same arguments also hold for any mass transfer problem in a multiphase flow system. As soon as the mixing time in a certain medium is larger than the reaction time, the non-uniform distribution of a certain component will lead to difficulties in stirring the selectivity towards a desired product.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

No data was used for the research described in the article

#### Acknowledgments

This work is part of the research program “First principles based multi-scale modeling of transport in reactive three phase flows” with project number 716.014.001, which is financed by the Netherlands Organization for Scientific Research (NWO) TOP grant. The authors also thank SURF ([www.surf.nl](http://www.surf.nl)) for the support in using the National Supercomputer Snellius.

#### References

- [1] S.M. Jowitt, G.M. Mudd, J.F.H. Thompson, Future availability of non-renewable metal resources and the influence of environmental, social, and governance conflicts on metal production, *Commun. Earth Environ.* 1 (1) (2020) 1–8.
- [2] A.I. Stankiewicz, J.A. Moulijn, Process intensification strategy, *Chem. Eng. Prog.* 96 (1) (2000) 22–34.
- [3] D. Reay, C. Ramshaw, A. Harvey, Chapter 1 - A brief history of process intensification, in: D. Reay, C. Ramshaw, A. Harvey (Eds.), *Process Intensification*, second ed., in: *Isotopes in Organic Chemistry*, Butterworth-Heinemann, Oxford, 2013, pp. 1–25.
- [4] A.C. Dimian, C.S. Bildea, A.A. Kiss, Chapter 10 - Process intensification, *Comput.-Aided Chem. Eng.* 35 (2014) 397–448.
- [5] D. Reay, C. Ramshaw, A. Harvey, Chapter 2 - Process intensification – An overview, in: D. Reay, C. Ramshaw, A. Harvey (Eds.), *Process Intensification*, second ed., in: *Isotopes in Organic Chemistry*, Butterworth-Heinemann, Oxford, 2013, pp. 27–55.
- [6] A. Panda, Y.E.J. Weitkamp, A.H. Rajkotwala, E.A.J.F. Peters, M.W. Baltussen, J.A.M. Kuipers, Influence of gas fraction on wall-to-liquid heat transfer in dense bubbly flows, *Chem. Eng. Sci.* X 4 (2019) 100037.
- [7] F. Kexel, S. Kastens, J. Timmermann, A. von Kameke, M. Schlüter, Experimental investigation of reactive bubbly flows - influence of boundary layer dynamics on mass transfer and chemical reactions, in: M. Schlüter, D. Bothe, S. Herres-Pawlis, U. Niekens (Eds.), *Reactive Bubbly Flows: Final Report of the DFG Priority Program 1740*, Springer International Publishing, Cham, 2021, pp. 267–307.
- [8] F. Kexel, A. v. Kameke, M. Hoffmann, M. Schlüter, The influence of fluid dynamics on the selectivity of fast gas-liquid reactions in methanol, *Chem. Eng. Process. Process Intensif.* 180 (2022) 108650.
- [9] D. Deising, D. Bothe, H. Marschall, Direct numerical simulation of mass transfer in bubbly flows, *Comput. & Fluids* 172 (2018) 524–537.
- [10] C.M.Y. Claassen, S. Islam, E.A.J.F. Peters, N.G. Deen, J.A.M. Kuipers, M.W. Baltussen, An improved subgrid scale model for front-tracking based simulations of mass transfer from bubbles, *AIChE J.* 66 (4) (2020) e16889.
- [11] D. Colombet, D. Legendre, A. Cockx, P. Guiraud, Mass or heat transfer inside a spherical gas bubble at low to moderate Reynolds number, *Int. J. Heat Mass Transfer* 67 (2013) 1096–1105.
- [12] B.P. LeClair, A.E. Hamielec, Viscous flow through particle assemblages at intermediate Reynolds numbers — a cell model for transport in bubble swarms, *Can. J. Chem. Eng.* 49 (6) (1971) 713–720.
- [13] Z. Feng, E.E. Michaelides, Mass and heat transfer from fluid spheres at low Reynolds numbers, *Powder Technol.* 112 (1–2) (2000) 63–69.
- [14] F. Takemura, A. Yabe, Gas dissolution process of spherical rising gas bubbles, *Chem. Eng. Sci.* 53 (15) (1998) 2691–2699.
- [15] S. Winnikow, Letter to the editors, *Chem. Eng. Sci.* 22 (3) (1967) 477.
- [16] J. Boussinesq, Calcul du poivoir refroidissant des courants fluide, *J. Math. Pures Appl.* 60 (1905) 285–332.
- [17] G. Kong, K.A. Buist, E.A.J.F. Peters, J.A.M. Kuipers, Dual emission LIF technique for pH and concentration field measurement around a rising bubble, *Exp. Therm Fluid Sci.* 93 (October 2017) (2018) 186–194.
- [18] I. Roghair, M. Van Sint Annaland, J.A.M. Kuipers, An improved Front-Tracking technique for the simulation of mass transfer in dense bubbly flows, *Chem. Eng. Sci.* 152 (2016) 351–369.
- [19] J.G. Khinast, Impact of 2-D bubble dynamics on the selectivity of fast gas-liquid reactions, *AIChE J.* 47 (10) (2001) 2304–2319.
- [20] J.G. Khinast, A.A. Koynov, T.M. Leib, Reactive mass transfer at gas-liquid interfaces: Impact of micro-scale fluid dynamics on yield and selectivity of liquid-phase cyclohexane oxidation, *Chem. Eng. Sci.* 58 (17) (2003) 3961–3971.
- [21] A. Weiner, C.M.Y. Claassen, I.R. Hierck, J.A.M. Kuipers, M.W. Baltussen, Assessment of a subgrid-scale model for convection-dominated mass transfer for initial transient rise of a bubble, *AIChE J.* 68 (7) (2022) e17641.
- [22] A. Kameke, S. Kastens, S. Rüttinger, S. Herres-Pawlis, M. Schlüter, How coherent structures dominate the residence time in a bubble wake: An experimental example, *Chem. Eng. Sci.* 207 (2019) 317–326.
- [23] G. Kong, *Hydrodynamics and Mass Transfer in Bubbly Flow: An Experimental Study* (Ph.D. thesis), Technische Universiteit Eindhoven, 2019, Proefschrift.