Chapter 1

Introduction

Bubbly flows are important in many applications in the chemical and biochemical industry. In oxidation processes for example, oxygen has to be dissolved in the reactive liquid phase. Further, oxygen is of importance in fermentation and cell culture processes to supply cells and microorganisms. In all these applications the relevant chemical and biochemical reactions take place in the liquid phase, thus the mass transfer from the gaseous phase into the liquid phase is of highest relevance for the whole process. The mass transfer is defined by the mass transfer coefficients, the available interfacial area, which can be described by the bubble size distribution (BSD), and the concentration difference between the gaseous and the bulk phase.

In the Collaborative Research Centre 1615 "SMART Reactors" (CRC 1615), funded by the German Research Foundation, new reactor types are investigated which will face climate change by operating **S**ustainably, for **M**ultipurpose, **A**utonomously, **R**esiliently, and **T**ransferably. Clearly, for this kind of reactors the prediction of the multiphase fluid dynamics is indispensable and especially the bubble size distribution is dominating in buoyancy driven flows the mixing and the mass transfer performance.

This work focuses on the development, improvement, and application of a breakup model for gaseous bubbles dispersed in a liquid phase [Wei24]. In the past, many groups have worked on the determination of the BSDs which occur during selected processes. In 2009 and 2010, Liau and Lucas have published two review articles, summarising and classifying many attempts to describe the bubble breakup [Lia09] and the bubble coalescence [Lia10]. Population balance models (PBM) have been proven to perform rather well for predicting BSDs. First mentioned by Hulburt and Katz in 1964 [Hul64] they received ever increasing attention. A detailed book about PBMs was published by Ramkrishna in 2000, delivering an in-depth insight into the mechanism and an overview over the fields of application [Ram00]. More recently, several research groups such as Marchisio et al. and Fox et al. [Mar03a, Mar06, San05, Gav10], Maaß et al. [Maa12, Maa07] or Alopaeus et al. [Buf16b, Buf16a, Zha18], just to mention a few, have worked intensively with PBMs, improved and coupled them with Computational Fluid Dynamics (CFD). While PBMs predict the development of a BSD by taking into account selected kernels which describe e.g. breakage and coalescence probabilities, other bubble breakup models, originating in the theory of isotropic turbulence [Tay35], have also been used widely during the past decades. Hinze proposed in 1955 an expression to calculate the largest possible diameter a drop can assume [Hin55], taking into account quantities such as the turbulent eddy dissipation rate, originating from the theory of isotropic turbulence. For this purpose, measurements from Batchelor [Bat51] have been used to estimate arising proportionality constants. This relation was later, in 1990, extended and further examined by Kawase and Moo-Young [Kaw90] and resulted in a model developed by Martínez-Bazán et al. [Mar99a, Mar99b] describing both, the breakup frequency and the distribution of daughter bubbles, according to a statistical view. Most recently, this model was used by Jamshidian et al. to predict bubble sizes and the mass transfer in stirred tank reactors simulatively [Jam25]. Also in 1990, Prince and Blanch have proposed a phenomenological model to estimate both, the bubble coalescence and breakup in air sparged bubble columns. For this purpose, they have taken into account the bubble-bubble collision rates, the resulting collision efficiencies, and the sizes of turbulent eddies encountering the bubbles, respectively [Pri90]. Luo and Svendson, Lehr et al., and later Xing et al. have investigated the whole range of turbulent eddies and their contained kinetic energy bombarding the observed bubble to develop criteria for bubble breakup and give expressions for the resulting daughter size distribution [Luo96, Leh02, Xin15]. Following the approach of Luo and Svendson, van den Hengel et al. have developed a model to predict the breakup and coalescence of gas bubbles in bubble columns [Hen05]. The group of Kuipers further investigated the influence of the Weber number as ratio between inertial and interfacial forces as dominant parameter for the bubble breakup [Lau14, Jai14]. Another attempt to predict the bubble breakup in such vessels was carried out by Mast and Takors, implementing findings from Luo and Svendson [Luo96] and Lehr et al. [Leh02] into selected CFD programmes [Mas23]. Following a different approach, Coulaloglou and Tavlarides developed a correlation, describing the Sauter mean diameter as function of the diameter of a stirred vessel and the Weber number [Cou77], taking into account the turbulent kinetic energy of the liquid surrounding the bubbles and comparing those with a critical value. Further, several other groups have focused on retrieving or predicting bubble sizes in stirred tanks in dependency of either gaseous or liquid phase properties or geometric characteristics, such as Wilkinson et al. [Wil93], Zhang et al. [Zha23], Zhou and Kresta [Zho98], Martín et al. [Mar08], Alves et al. [Alv02], and Parthasarathy and Ahmed [Par94]. Tsouris and Tavlarides have improved and combined several of the aforementioned models and used them for the solution of Population Balance Equations [Tso94]. Based on findings of numerous previously mentioned works, Sarimeseli and Kelbaliyev investigated the deformation of gas bubbles and its influence on the bubble breakup [Sar04]. Further than the aforementioned models, highly resolved numerical simulations have been conducted for example by Baltussen [Bal15], yielding very detailed information about the breakup of bubbles cut by a wire. The knowledge from those simulations can be applied to give more insight into the breakup process itself. A direct implementation of these highly resolved simulation to large scale systems on the other hand, is not feasible due to the high computational cost.

While many of the models that do predict BSDs have either the necessity to solve integrals, which can be numerically expensive, to determine proportionality constants which arise in the development of the correlation models or to adjust breakup or coalescence kernels, this work follows a different approach.

In this work, each and every bubble present in an observed system is treated individually. The bubbles' deformations are computed along their respective trajectory, hence the history of each bubble with regard to the local conditions is taken into account. The relevance of the history of a fluid particle along its trajectory has already been shown by Nachtigall et al. [Nac16] for droplets. Further, Risso et al. and Lalanne et al. [Ris98, Lal19] have formulated a differential equation, describing the bubble deformation along its trajectory by taking into account the velocity fluctuations in the near-bubble reagion. Krakau and Kraume have observed bubbles along their respective trajectories and during the breakup event experimentally to derive daughter bubble sizes and a deeper understanding of the bubble disintegration in stirred tank reactors [Kra19a, Kra19b]. Vela-Martín and Avila have linked the energy a droplet or bubble gains or loses on its way along the respective trajectory to the rate of strain or the rate of compression, respectively, obtainable from the rate-of-strain tensor [Vel21]. The work stresses the effect of the history of deformation of the bubble onto its breakup behaviour. The deformation and the consecutive breakup of single bubbles in a quiescent medium and in artificial turbulence were also recently observed experimentally and reviewed by the group of Ni et al. in a very detailed way [Qi20, Qi22, Ni15, Ni24]. This group has conducted experiments in which they recorded the shape and deformation of bubbles with multiple cameras and a high resolution. The results are then used to extend an already existing model for the evolution of the elliptical bubble shape and its orientation along its trajectory developed by Maffettone and Minale [Maf98]. Ni et al. introduced additional terms into the model that take into account the slip velocities between bubbles and carrier fluid as well as the coarse grained strain [Mas21b, Mas21a]. Further, the deformation and breakup of gas bubbles have also been investigated by Zedníková et al. experimentally [Vob23, Vej18].

To take into account the effect of the carrier flow on the bubble deformation in this work, the bubbles are assumed to behave like Kelvin[1]-Voigt[2] elements [Wei24], first mentioned by Thomson [Tho65] and Voigt [Voi92] and used for the description of plastic material properties and rheological phenomena. The modelling of bubbles as Kelvin-Voigt element has already been proposed by Lagisetty et al. [Lag86] and Nambiar et al. [Nam92], where the breakup criteria was derived from global variables such as the stirrer speed in the case of stirred vessels. In this work, the local phenomena of the carrier fluid causing the bubble deformation are based on Lagrangian[3] deformation theory, which has been intensively studied during the last decades to analyse transport barriers especially in oceanographic flows [Hal00, Hal01], but has also been applied recently to multi- and singlephase flows [Kam19, Lla20, Kur22, Wei23], which are relevant for chemical or biochemical processes. In the model, namely the Trajectory-Based Breakup Model (TBBM), the size of the daughter bubbles is retrieved by conserving the energy the bubble has gained due to deformation [Wei24]. Altogether, the TBBM is based on first principles from physics and mathematics. Neither fitting constants have to be determined by fitting data, nor tedious or computationally expensive integrals have to be solved. Summarising, in the development of the TBBM the following assumptions are made:

- Every bubble is either a prolate or an oblate ellipsoid with an initially spherical shape and is modelled with a Kelvin-Voigt element. This might lead to errors when strictly wobbling bubbles are to be expected, compare Clift et al. [Cli92].

- The tension which is relevant for the deformation is only based on the largest strain or the largest compression the bubble experiences at its position and the bubble rotates immediately in a way that it is always deformed in the same, initial direction, which could lead to a slight overprediction of the deformation, hence an overprediction of the breakup rate could occur.

[1]William Thomson, 1st Baron Kelvin OM, GCVO, PC, PRS, FRSE, British mathematician, mathematical physicist, and engineer, 1824-1907

[2]Woldemar Voigt, German physicist, 1850-1919

[3]Joseph-Louis Lagrange, born Giuseppe Luigi Lagrangia, Italian mathematician, physicist, and astronomer, 1736-1813

- The bubble breaks in the very moment in which it is so strongly deformed that the interfacial force is smaller than the inertial forces. Since the deformation of the bubbles already takes into account the time leading to the breakup this assumption is deemed to not be very error prone.

- The inertial forces are set equal to the drag force which is, in accordance to Mersmann [Mer77], approximated with the buoyancy force. This assumption might lead to both, over- and underestimation of breakup events.

- Every bubble which breaks undergoes a binary breakup while the daughter bubbles conserve the energy of the mother bubble in its deformed state. This assumption will lead to slight errors where ternary or higher order breakup is relevant. Those errors are not expected to be too large, since the observed bubble could potentially break again in a rather short time, leading to a pseudo higher order breakup.

Measures to encounter the other limitations introduced with the listed assumptions in future works are summarised in Chapter 6 at the end of this thesis. The TBBM is implemented and tested using M-Star CFD (M-Star Simulations, LCC.) for a wide range of scales, ranging from the small scale of 3 L, containing about a hundred bubbles at a point in time, to the large scale of $15\,\text{m}^3$, containing about a million bubbles at a point in time. The validation shown in this work is limited to a mid-size stirred tank reactor with a working volume of 200 L. The model is validated with experimental data and compared with the default model in M-Star CFD based on the critical diameter defined by Kawase and Moo-Young [Kaw90] coupled with a statistical expression for the daughter size distribution by Xing et al. [Xin15] as described by Thomas et al. [Tho22]. In contrast to Weiland et al. [Wei24], in its current state the TBBM includes the possibility for both, prolate and oblate deformation rather than be restricted to pure prolate deformation. To further analyse the capabilities of the TBBM, simulations regarding the mass transfer of oxygen are conducted, respecting the influence of the bubble sizes, the local flow conditions, and the local concentration differences.