Implementation of an Improved Bubble Breakup Model for TFM-PBM

Simulations of Gas-Liquid Flows in Bubble Columns

Xiaofeng Guo, Qiang Zhou, Jun Li, Caixia Chen*

Key Laboratory of Coal Gasification and Energy Chemical Engineering of Ministry
of Education, East China University of Science and Technology, Shanghai 200237,
China

Abstract

An improved bubble coalescence and breakup model was implemented in the inhomogeneous TFM-PBM model for buoyancy-driven gas-liquid bubbly flows. The bubble coalescence was modeled by considering bubble collisions induced by turbulent fluctuations, buoyancy driven, wake entrainment, and viscous shear, and the liquid film drainage model was used for the description of the coalescence efficiency of collisions. The bubble breakup was analyzed in terms of bubble interactions with turbulent eddies which coupled the restriction of surface energy with the capillary pressure. A generic TFM-PBM model was developed and applied for the simulations of bubble columns operated in different flow regimes. The evolutions of the bubble size distributions were simulated and validated for literature data of a $D=0.14$ m cylindrical bubble column at gas superficial velocities 0.03 and 0.45 m/s, respectively. Furthermore, a well documented $D=0.44$ m bubble column was simulated, and the predicted distributions of the gas holdup and liquid velocity were compared with the experimental measurements of Chen et al. (1998). The model showed the capacity of

* To whom correspondence should be addressed: Tel:86-021-64252057 E-mail address: exchen@ecust.edu.cn

describing the gas-liquid fluid dynamics of bubble columns operated in both bubbly and churn-turbulent flow regimes.

Keywords

Bubble coalescence and breakup; Bubble size distribution; TFM-PBM model; Bubbly and churn-turbulent bubble columns

1. Introduction

Computational fluid dynamics (CFD) has become an important approach for analyzing the hydrodynamics of buoyancy-driven gas-liquid flows, and the Two-Fluid Model (TFM) has been frequently employed in the simulations of the bubble columns (Sanyal et al., 1999; Wang et al., 2003; Chen and Fan, 2004; Chen et al., 2005; Cheung et al., 2007; Tabib et al., 2008; and Xu et al., 2014; among others). However, a generic TFM model for the gas-liquid bubble columns operated in different flow regimes has not been well established due to the lack of reasonable closure models that describe the gas-liquid interface transport phenomena. The interface interactions are mainly induced through the drag force that liquid exerts on the bubble surface due to viscosity, the lift force caused by the shear flows around the bubbles, and the turbulent dispersion force due to the collisions of liquid eddies. Therefore, the interface closure model relies on (1) the theoretical formulation of each interaction force, (2) the model of the bubble induced turbulence of the liquid phase, (3) the descriptions of bubble sizes, which are considered to be closely connected to the models of the interaction forces and the turbulence (Xu et al., 2013).

1 In most early TFM simulations of dispersed gas-liquid flow, the spatial
2 time-variation of the bubble sizes were not considered and a constant bubble size was
3 used instead (Sanyal et al., 1999; Oey et al., 2003; Zhang et al., 2005; Tabib et al.,
4 2008; and Rzehak and Krepper, 2013; among others). The simplification of the bubble
5 size model limits the TFM simulations to a bubbly flow regime, because the bubble
6 size distribution in such a condition is narrow and the interaction between bubbles is
7 relatively weak. However, for a bubble column operated in a churn-turbulent regime,
8 the flow structure becomes very complex. The bubble coalescence and breakup results
9 in a wide distribution of bubble sizes which is greatly different from the assumption
10 of a constant bubble size.

11 The dynamic variation of the bubble sizes and their distributions in buoyancy
12 driven gas-liquid flows have been modeled using the population balance model
13 (PBM). With PBM, the bubbles are represented by a number of size bins, and usually,
14 can be classified into two discrete size groups: a small size group (with bubbles
15 smaller than 5.8 mm) and a large size group (with bubbles larger than 5.8 mm)
16 according to the sign change of lift force (Tomiyama et al., 2002; Lucas et al., 2003;
17 and Krepper et al., 2008). The mass and momentum transfer between the two discrete
18 phases is accounted for by the computations of the birth and death rates due to the
19 coalescence and breakup of bubbles. In the past decades, substantial efforts have been
20 made to develop a bubble coalescence and breakup models (Prince and Blanch, 1990;
21 Luo and Svendsen, 1996; Martinez-Bazan et al., 1999; Lehr and Mewes, 2001; Wang
22 et al., 2003; Zhao and Ge, 2007; Liao and Lucas, 2010; Solsvik and Jakobsen, 2015;

1 among others), while a generic bubble coalescence and breakup model has not yet
2 been established in combination with the TFM-PBM simulations due to the
3 complexity of the transport phenomena resulting from bubble coalescence and
4 breakup.

5 For the bubble coalescence process, several criteria have been proposed, and the
6 film drainage model of Shinnar and Church (1960) was widely acknowledged among
7 others. According to the film drainage model, a liquid film is firstly formed when two
8 bubbles come close due to an overpressure; the liquid film begins to drain sequentially,
9 and finally results in film rupture and bubble coalescence. While, if the pressure force
10 is insufficient to overcome the viscosity force of the thin film, the bubbles bounce
11 back without coalescence. The coalescence probability depends on the intrinsic
12 contact time and drainage time between the bubbles. Howarth et al. (1964) suggested
13 that the attraction force between two colliding interfaces was too weak in comparison
14 with the turbulent force to control the coalescence probability, and concluded that
15 whether coalescence will occur or not depended on the impact of colliding fluid
16 particles. Note that during energetic collisions, when the approach velocity of two
17 colliding bubbles exceeds a critical value, the dominant mechanism is an immediate
18 coalescence without liquid film capturing and thinning. Lehr et al. (2002) introduced
19 an empirical model using a critical approach velocity based on the experimental
20 observation. In all mechanisms mentioned above, contact and collision is the premise
21 of coalescence. The collision between bubbles is usually caused by their relative
22 velocity, and the relative motion may occur due to a variety of mechanisms. Prince

1 and Blanch (1990) proposed a collision model considering effects of turbulent eddy
2 fluctuation, buoyancy-driven and liquid viscous shear. In some cases, the effects of
3 buoyancy-driven and liquid viscous shear are insignificant compared with the
4 turbulence eddy fluctuations, therefore, Luo and Svendsen (1996) proposed a collision
5 model which only took into account the collisions induced by turbulence fluctuations.
6 Lehr et al. (2002) considered the coalescence caused by turbulence random motion
7 that was directly caused by a critical velocity. Liao and Lucas (2010) summarized
8 almost all the published papers about bubble collision induced coalescence
9 mechanisms, namely, turbulence fluctuation, viscous shear stress, capture in turbulent
10 eddies, buoyancy and wake interaction. A baseline model was introduced and applied
11 to a dispersed bubbly flow simulation (Liao et al., 2015).

12 For the bubble breakup process, according to the theory of drop breakup, a fluid
13 particle's deformation and subsequent breakup are usually described by a stress or
14 kinetic energy balance. Coulaloglou and Tavlarides (1977) proposed the first
15 phenomenological model for drop breakup in turbulent flows, and assumed that
16 breakup would take place if the turbulent kinetic energy received by the drop from the
17 turbulent eddies exceeded the surface energy of the drop itself. Prince and Blanch
18 (1990) agreed that bubble breakup was caused by eddy collision and assumed that
19 only eddies of length scale larger than 0.2 times bubble diameter or equal to the
20 bubble diameter could induce a bubble breakup. The large eddies are merely
21 responsible for the translatory motion of bubbles, and the small eddies determine the
22 strain, thus dominate the deformation of the bubbles in the flow field. Luo and

1 Svendsen (1996) considered eddies not only with different length scale but also with
2 different amounts of energy. In their model, eddies that are responsible for breakup
3 are equal to or marginally smaller than the colliding bubbles. Large eddies simply
4 transport the bubble without causing breakup, while very small eddies do not contain
5 sufficient energy to induce breakup. The minimum length scale of eddies equals to
6 11.4 times Kolmogorov scale. For a particular size eddy hitting a bubble, the
7 probability for bubble breakup depends not only on the energy contained in the
8 arriving eddy, but also on the minimum energy required by the surface area increase
9 due to a bubble fragmentation. Lehr et al. (2002) breakup model is similar to that of
10 Luo and Svendsen (1996) in the calculation of the bubbles collision. The difference is
11 that the minimum length scale of eddies is determined by the size of the small bubble
12 fragment resulting from the breakup, and that the force balance between eddies and
13 bubble collisions result in breakups. In other words, the breakup of a bubble is
14 determined by the balance between the interfacial force of the bubble surface and the
15 inertial force of the colliding eddy. The interfacial forces depend on the shape of a
16 bubble and on the size of the resulting bubble fragments. Breakup occurs immediately
17 when the inertial force of the colliding eddy becomes larger than interfacial force of
18 the bubble surface. Based on the results of Luo and Svendsen (1996) and Lehr et al.
19 (2002), Wang et al. (2003) proposed a new breakup model which accounted for both
20 the energy constraint and a capillary constraint, and the model provided more
21 reasonable results than that of Luo and Svendsen (1996). However, the complicated
22 computation of the breakup kernel limited the use of the model, though it was applied

in a two-dimensional simulation of a bubble column. Still, the idea of coupling the energy constraint and the capillary constraint in the breakup criteria has been adopted in other recent work, like Zhao and Ge (2007) in a three-phase simulation, and Liao et al. (2015) in a dispersed bubbly flow.

In the present work, an improved bubble coalescence and breakup kernel function is implemented in the TFM-PBM model in the ANSYS Fluent solution platform. The work is a continuation of our previous effort aimed at implementing a generic hydrodynamic model for gas-liquid bubble column flows (Xu et al., 2013). The existing schemes of the TFM-PBM simulations are reviewed with an emphasis on the bubble coalescence and breakup models. Fully three-dimensional gas-liquid flow simulations are conducted for cylindrical bubble columns of laboratory scale, and the results of model validations are presented.

2. The population balance model (PBM)

2.1 The population balance equation (PBE)

Population balance model is used to describe a wide dispersed phase size distribution (bubbles or particles) based on the balance equation of time change rates of the number or the number density of bubbles. For the inhomogeneous PBM, the dispersed gas phase is further divided into a small bubble group and a large bubble group. The continuity equations for the two bubble groups are expressed as:

$$\begin{cases} \frac{\partial \alpha_{small}}{\partial t} + \nabla \cdot (\alpha_{small} \mathbf{u}_{small}) = \sum_{i=1}^{N_{small}} S_i \\ \frac{\partial \alpha_{large}}{\partial t} + \nabla \cdot (\alpha_{large} \mathbf{u}_{large}) = \sum_{i=1}^{N_{large}} S_i \end{cases} \quad (1)$$

Where, α , $\mathbf{u}$, N , represent the volume fraction, velocity and bins of bubble, respectively. S_i is the source term of i -th bubble group generated by bubble coalescence and breakup, which is expressed as:

$$\frac{S_i}{V_i} = C_{B,i} - C_{D,i} + B_{B,i} - B_{D,i} \quad (2)$$

Note, $C_{B,i}$, $C_{D,i}$, $B_{B,i}$, $B_{D,i}$ represent the source terms of dispersed phase bubble class i resulting from birth due to coalescence of bubble classes j and k , death due to coalescence with bubble j , birth due to breakup of higher bubble classes j and reassign of bubbles $i+1$ and i , and death due to breakup in present bubble class i , respectively. The notions of each source term and their physical representations are shown in **Fig. 1**.

$C_{B,i}$ (birth due to coalescence) $C_{D,i}$ (death due to coalescence)

$B_{B,i}$ (birth due to breakage) $B_{D,i}$ (death due to breakage)

Fig. 1. Bubbles birth and death due to coalescence and breakup.

No breakup for the smallest class ($i=1$), and no coalescence for the largest class ($i=N$) are assumed. The source terms of Eq. (2) are computed following the

1 Hageseather et al. (2002) method as below:

$$2 \quad (a) \quad C_{B,i} = \frac{1}{2} \sum_{j=1}^i \sum_{k=1}^i c(v_j, v_k) x_{jk} \xi_{jk} n_j n_k \quad (3)$$

$$3 \quad (b) \quad C_{D,i} = \sum_{j=1}^{N-1} c(v_i, v_j) n_i n_j \quad (4)$$

$$4 \quad (c) \quad B_{B,i} = \sum_{j=i+1, i \neq N}^N b(v_j : v_i) n_j + \sum_{j=1, i \neq N}^i y_{i+1,j} b(v_{i+1} : v_j) n_{i+1} \\ + \sum_{j=1, i \neq N}^{i-1} (1 - y_{i,j}) b(v_i : v_j) n_i \quad (5)$$

$$5 \quad (d) \quad B_{D,i} = \sum_{j=1}^{i-1} b(v_i : v_j) n_i \quad (6)$$

6 Where, v is bubble volume, n is the bubble number density. $c(v_i, v_j)$
7 represents the coalescence rate of two aggregate bubbles of volume i and j , and
8 $b(v_i : v_j)$ represents the breakup frequency of bubble of volume i breaking to volume
9 j , which will be described in next section. For present work, bubbles from 0.57 mm to
10 5.8 mm in diameters belong to the small bubble group, and 5.8 mm to 29.0 mm in
11 diameters belong to the large bubble group. All bubbles are divided into a total of 18
12 classes based on the study of Chen et al. (2004) and Ekambara et al. (2008). The sizes
13 of i -th and $(i-1)$ -th bins bubble classes satisfy $v_i = 2v_{i-1}$. Meanwhile, bubbles that
14 have been generated due to coalescence and breakup need to be reassigned or
15 redistributed to two size classes to guarantee the conservation of mass, as shown in

16 **Fig. 2.**

17

Fig. 2. Bubble coalescence and reassignment process.

The redistribution factors ξ , x and y appearing in Eqs. (3) - (6) are defined as follows:

$$\xi_{jk} = \begin{cases} 1 & ; \quad v_{i-1} < v_{jk} < v_{i+1} \\ 0 & ; \quad \text{else} \end{cases} \quad (7)$$

$$x_{jk} = \begin{cases} (v_{jk} - v_{i-1}) / (v_i - v_{i-1}) & ; \quad v_{i-1} < v_{jk} \leq v_i \\ (v_{jk} - v_{i+1}) / (v_i - v_{i+1}) & ; \quad v_i < v_{jk} < v_{i+1} \\ v_{jk} / v_N & ; \quad v_N < v_{jk} \end{cases} \quad (8)$$

$$y_{i+1,j} = \frac{(v_{i+1} - v_j) - v_{i+1}}{(v_i - v_{i+1})} = \frac{v_1 \cdot 2^j}{v_1 \cdot 2^i} = 2^{j-i} \quad (9)$$

2.2 Coalescence rate $c(v_i, v_j)$

Bubbles coalescence is induced by collisions, however, not every collision results in coalescence. The coalescence probability resulting from collisions is referred to as coalescence efficiency. Two parameters, namely the collision frequency w_{ij} and the coalescence efficiency $p_c(d_i, d_j)$ are used for describing the bubble coalescence rate and the total coalescence rate of bubbles of size d_i and d_j are computed as follows.

$$c(v_i, v_j) = w_{ij} \cdot p_c(d_i, d_j) = A_{ij} \cdot u_{ij} \cdot p_c(d_i, d_j) \quad (10)$$

The film drainage model is employed for the computation of coalescence

efficiency. Four mechanisms of collisions are included in the present model. As is shown in **Fig. 3**, the collisions due to turbulence fluctuation, buoyancy driven, wake entrainment and viscous shear are major causes of two bubbles colliding with each other. The collision frequency of each mechanism is described as follows.

Fig. 3. The illustration of bubble collision.

Collision frequency due to turbulent fluctuation. The collision frequency of bubble of diameter d_i with a bubble of diameter d_j is expressed analogous to the gas kinetic theory (Prince and Blanch, 1990) by:

$$w_{ij}^T = A_{ij} \bar{u}_{ij} = A_{ij} (u_{ti}^2 + u_{tj}^2)^{1/2} = \frac{\pi}{4} (d_i + d_j)^2 \cdot \sqrt{2} \varepsilon^{1/3} (d_i^{2/3} + d_j^{2/3})^{1/2} \quad (11)$$

The collision frequency is estimated as the volume that impact area A_{ij} passes by in unit time. For consistency, minor change has been made for the factor of Eq. (11) than the original ones appeared in the literature. The turbulent velocity u_{ti} in the inertial sub-range of isotropic turbulence according to Rotta (1972) is expressed as:

$$u_{ti} = \sqrt{2} (\varepsilon d_i)^{1/3}.$$

Collision frequency due to buoyancy driven. The difference in rise velocities of

1 bubbles of different sizes was introduced by Friedlander (1977). The buoyant
 2 collision frequency is given by:

$$3 \quad w_{ij}^B = 0.5 \cdot A_{ij} \Delta \bar{u}_{bij} = 0.5 \cdot \frac{\pi}{4} (d_i + d_j)^2 \cdot |u_{bi} - u_{bj}| \quad (12)$$

4 The event of two bubbles colliding with each other will happen only when the
 5 trailing bubble is larger than the leading one. Due to the randomness of the bubble
 6 size distribution, the probability of the collision due to buoyancy driven would be
 7 reduced by a factor of 0.5, following Liao et al. (2015). The rise velocity can be
 8 expressed as a function of bubble size. Clift et al. (1978) provided an expression
 9 $u_{bi} = (2.14\sigma / \rho_l d_i + 0.505 g d_i)^{1/2}$ that covered the size range typical of gas-liquid
 10 contactors operating in the turbulent flow regime.

11

12 ***Collision frequency due to wake entrainment.*** Wang et al. (2006) indicated that the
 13 wake of large bubbles, generating negative pressure which sucks a small bubble, may
 14 significantly increase the frequency of collision. Liao et al. (2015) corrected wake
 15 entrainment velocity $u_{wi} = u_{bi} C_D^{1/3}$ and considered the randomness of wakes of two
 16 collision bubbles. Finally, the frequency of wake entrainment induced collision
 17 between two bubbles is derived as (Liao et al. 2015):

$$18 \quad w_{ij}^W = 0.5 \cdot A \bar{u}_w = 0.5 \cdot (A_i u_{wi} + A_j u_{wj}) = 0.5 \cdot \frac{\pi}{4} (d_i^2 \Theta_i u_{wi} + d_j^2 \Theta_j u_{wj}) \quad (13)$$

19 Where, a coefficient Θ_i was introduced by Wang et al. (2006) to represent the
 20 capability of a bubble wake entraining a small bubble:

$$21 \quad \Theta_i = \begin{cases} (d_i - 0.5d_c)^6 / [(d_i - 0.5d_c)^6 + (0.5d_c)^6]; & d_i \geq 0.5d_c \\ 0 & \text{otherwise} \end{cases} \quad (14)$$

22 Furthermore, a critical bubble diameter, which is the volume equivalent diameter

1 of bubbles corresponding to spherical caps, can be estimated according to Ishii and
 2 Zuber (1979) $d_c = 4 \left(\sigma / \left[g \cdot (\rho_l - \rho_g) \right] \right)^{1/2}$.

3

4 **Collision frequency due to viscous shear.** The final contribution to the bubble
 5 collision frequency is due to instantaneous velocity gradients in the liquid phase. It is
 6 well recognized that bubbles are placed in a variety of liquid strain field, and the
 7 velocity gradient may generate differential pressure leading to bubble collisions,

$$8 \quad w_{ij}^v = 0.5 \cdot A_{ij} \bar{u}_{vij} = 0.5 \cdot \frac{\pi}{4} (d_i + d_j)^2 \left[\frac{0.5}{\pi} (d_i + d_j) \gamma_l \right] \quad (15)$$

9 The liquid velocity gradient u_{vi} which induces collision in the turbulence
 10 according to Liao et al. (2015) can be expressed as, $u_{vi} = \frac{0.5}{\pi} (d_i + d_j) \gamma_l$. Where,

$$11 \quad \gamma_l = \left(2 \left(\frac{\partial u_l}{\partial x} \right)^2 + 2 \left(\frac{\partial v_l}{\partial y} \right)^2 + 2 \left(\frac{\partial w_l}{\partial z} \right)^2 + \left(\frac{\partial u_l}{\partial y} + \frac{\partial v_l}{\partial x} \right)^2 + \left(\frac{\partial v_l}{\partial z} + \frac{\partial w_l}{\partial y} \right)^2 + \left(\frac{\partial w_l}{\partial x} + \frac{\partial u_l}{\partial z} \right)^2 \right)^{1/2} \quad (16)$$

12

13 **Coalescence efficiency.** The coalescence efficiency adopts film drainage model
 14 introduced by Shinnar and Church (1960), which depends on the intrinsic contact time
 15 and drainage time between the bubbles. For air-water system, an estimate of the initial
 16 thickness of the film h_0 is in an order of 10^{-4} m and the critical film thickness h_f is
 17 typically taken an order of 10^{-8} m, respectively.

$$18 \quad p_c(d_i, d_j) = \exp \left(- \frac{t_{drainage}}{t_{contact}} \right) = \exp \left(- \frac{r_{ij}^{5/6} \rho_l^{1/2} \varepsilon^{1/3}}{4\sigma^{1/2}} \ln \frac{h_0}{h_f} \right) \quad (17)$$

19 The equivalent radius can be derived according to Chesters (1991)

$$20 \quad r_{ij} = d_i d_j / (d_i + d_j).$$

The final equation for bubble coalescence rate including four collision mechanisms is expressed as follows:

$$c(v_i, v_j) = (w_{ij}^T + w_{ij}^W + w_{ij}^B + w_{ij}^V) \cdot p_c(d_i, d_j) \quad (18)$$

The relationship between bubble diameter and collision frequency due to different collision factors and coalescence efficiency in air-water system is presented in **Fig. 4**. The impact area between bubbles grow as the bubble diameter increases, leading to increases in the collision frequency and decreases in the coalescence efficiency. Compared with other mechanisms of collisions, the contribution of turbulence eddy fluctuation is significantly large.

Fig. 4. Effect of bubble diameter on collision frequency and coalescence efficiency in air-water system.

2.3 Breakup frequency $b(v_i : v_j)$

In turbulent dispersion systems, the fluid dynamics and breakup processes are complex due to the existence of a large number of eddies with various length scales and different amounts of energy. Velocity fluctuations at a point can be caused by the arrival of specific eddies. The arriving eddies have an uneven pressure distribution on a bubble surface, thus causing bubble breakup. In other words, bubble breakup occurs through bubble interactions with turbulent eddies, and the bubbles are usually deformed in the turbulent flow field and stretched in one direction. This leads to a necking that contracts further, finally resulting in the breakup. Actually, when breakup happens, the breakup volume fraction is assumed to be a stochastic variable, and the daughter bubble size has a certain distributions. Therefore, breakup frequency and daughter bubble size distributions are two important parameters in a breakup model as presented in many bubble breakup models (Luo and Svendsen, 1996; Lehr and Mewes, 2001; Wang et al., 2003; Zhao and Ge, 2007; Martinez-Bazan et al., 2010; Solsvik and Jakobsen, 2015; among others).

For the computation of a breakup kernel function, some assumptions include: (1) the breakup of a bubble in the turbulent dispersion systems is due to the arrival of eddies of different length scales and different amounts of energy onto the surface of the bubbles. (2) Only the binary breakup into two daughter bubbles is considered. (3) Only eddies with length scales smaller than the bubble diameters can induce breakup,

1 larger eddies lead to bubble transport instead. According to Lehr et al. (2002), the
 2 length scale of the eddy λ leading to bubble breakup has to be smaller than the
 3 diameter of parent bubble d and larger than the smaller bubble fragment $d_{\min}$. This
 4 gives a simple relationship $d_{\min} \leq \lambda \leq d$.

5 The breakup frequency is calculated from the frequency of arriving eddies onto
 6 the surface of the bubble and from the probability that collisions lead to breakup,

$$7 \quad b(v_i : v_j) = \int_{d_{\min}}^{d_i} w_i(d_i, \lambda) \cdot p_b(d_i : d_{j,k}) d\lambda \quad (19)$$

8 Where, $w_i(d_i, \lambda)$ denotes the frequency of collisions between a parent bubble
 9 with the volume $v_i = \pi d_i^3 / 6$ and an eddy with length scale λ . $p_b(d_i : d_{j,k})$
 10 denotes the probability of collisions resulting in a breakup and leading to the
 11 formation of a daughter bubble with size d_j and d_k . Specifically, the lower limit of
 12 integral can be expressed as $d_{\min} = \min(d_j, d_k)$.

13 For the breakup mechanisms, two categories, namely, turbulence fluctuation and
 14 viscous shear are considered. Where, the interfacial instability of large bubbles is not
 15 included, because of lacking of a sound theoretical description of the mechanism and
 16 specific experimental data. According to Luo and Svendsen (1996), the collision
 17 frequency due to turbulence fluctuation can be expressed as,

$$18 \quad w_T(d_i, \lambda) = A_i \bar{u}_\lambda \dot{n}_\lambda = \frac{\pi}{4} (\lambda + d_i)^2 \cdot \sqrt{2} (\varepsilon \lambda)^{1/3} \cdot \frac{0.822(1 - \alpha_g)}{\lambda^4} \quad (20)$$

19 The viscous shear, as pointed out by Sporleder et al. (2012), is also considered,

$$20 \quad w_V(d_i, \lambda) = A_i \bar{u}_v \dot{n}_\lambda = \frac{\pi}{4} (\lambda + d_i)^2 \cdot \left[\frac{0.5}{\pi} (d_i + \lambda) \dot{\gamma}_l \right] \cdot \frac{0.822(1 - \alpha_g)}{\lambda^4} \quad (21)$$

21 In one view, breakup is assumed to occur when the disruptive stresses causing

1 bubble deformation are not sufficiently balanced by the stresses that counteract
 2 deformation. The breakup probability resulting from collisions between eddies and
 3 bubbles is determined by using a critical stress force computed below:

$$4 \quad p_b(d_i : d_{j,k}) = \exp \left(- \frac{\tau_{critical}}{\frac{1}{2} \rho_l \bar{u}_\lambda^2} \right) \quad (22)$$

5 As is shown in **Fig. 5a**, the critical stress force is determined by two different
 6 criteria; one is based on the increase in surface energy as introduced by Luo and
 7 Svendsen (1996) and further developed by Hageseather et al. (2002), the other one is
 8 based on the interaction force balance proposed by Lehr et al. (2002). The same idea
 9 has been adopted by Wang et al. (2003), Zhao and Ge (2007), Liao et al. (2015).

$$10 \quad \tau_{critical} = \max(\tau_{energy}, \tau_{pressure}) = \left(\frac{6c_f \sigma}{d_i}, \frac{\sigma}{\min(d_j, d_k)} \right); c_f = \left(\frac{d_j}{d_i} \right)^2 + \left(\frac{d_k}{d_i} \right)^2 - 1 \quad (23)$$

11 The surface energy and the pressure energy changes as functions of bubble
 12 breakup volume are drawn in **Fig. 5b**, respectively. When the volume difference of
 13 two daughter bubbles is significant, high pressure energy is needed, otherwise, high
 14 surface energy is needed.

15 The breakup frequency can be obtained by substituting Eqs. (20) - (23) into Eq.
 16 (19),

$$17 \quad b(v_i : v_j) = (1 - \alpha_g) \int_{f_{bv}^{1/3}}^1 \left[0.913 \left(\frac{\varepsilon}{d_i^2} \right)^{1/3} \frac{(1 + \xi)^2}{\xi^{11/3}} + 0.103 \frac{(1 + \xi)^3}{\xi^4} \dot{\gamma}_l \right] \exp \left(- \frac{\tau_{critical}}{\rho_l (\varepsilon \xi d_i)^{2/3}} \right) d\xi \quad (24)$$

19 Where, $\xi = \lambda / d_i$ is the size ratio between an eddy and a parent bubble;

20 $f_{bv} = (d_{min} / d_i)^3$ is the dimensionless bubble volume fraction of breakup.

Similar to the model of Luo and Svendsen (1996), the present breakup model does not need to explicitly define the daughter bubble size distributions, because the model directly gives the bubble breakup frequency for bubbles of diameter d_i breaking into the daughter bubbles with a given f_{bv} . The daughter particle size distributions can be obtained directly as a result of the computation.

Fig. 5. The illustration of bubble breakup and the situation of two compete stresses.

3. Governing equations of TFM and the closure models

3.1 Governing Equations of two-fluid model (TFM)

For the gas-liquid flow system, liquid is treated as a continuous phase, and the gas bubble is regarded as a dispersed secondary phase. The continuity and momentum equations of two fluids are represented in the Eulerian formulation as following:

$$\frac{\partial(\alpha_i \rho_i)}{\partial t} + \nabla \cdot (\alpha_i \rho_i \mathbf{u}_i) = 0 \quad (25)$$

$$\frac{\partial(\alpha_i \rho_i \mathbf{u}_i)}{\partial t} + \nabla \cdot (\alpha_i \rho_i \mathbf{u}_i \mathbf{u}_i) = -\alpha_i \nabla p + \nabla \cdot \boldsymbol{\tau}_i + \alpha_i \rho_i \mathbf{g} + \mathbf{F}_{i,j} \quad (26)$$

Where, α_i , ρ_i , $\mathbf{u}_i$, $\boldsymbol{\tau}_i$, and $\mathbf{F}_{i,j}$ represent volume fraction, density, velocity,

stress-strain tensor and inter-phase momentum exchange term of the i -th phase, respectively. The sum of the volume fractions of all phases is equal to 1.

3.2 The turbulence closure model

The dispersed renormalization group (RNG) k - ε turbulence model (Orszag et al., 1993) is used due to its capability of capturing the flow characteristics of bubble columns (Laborde-Boutet et al. 2009). The turbulence kinetic energy k and dissipation rate ε of liquid phase are computed by the following equations:

$$\frac{\partial(\alpha_l \rho_l k_l)}{\partial t} + \nabla \cdot (\alpha_l \rho_l \mathbf{u}_l k_l) = \nabla \cdot \left(\alpha_l \rho_l C_\mu \frac{k_l^2}{\varepsilon_l \sigma_k} \nabla k_l \right) + \alpha_l G_{k,l} - \alpha_l \rho_l \varepsilon_l + \alpha_l \rho_l \Pi_{k,l} \quad (27)$$

$$\frac{\partial(\alpha_l \rho_l \varepsilon_l)}{\partial t} + \nabla \cdot (\alpha_l \rho_l \mathbf{u}_l \varepsilon_l) = \nabla \cdot \left(\alpha_l \rho_l C_\mu \frac{k_l^2}{\varepsilon_l \sigma_\varepsilon} \nabla \varepsilon_l \right) + \alpha_l \frac{\varepsilon_l}{k_l} (C_{1\varepsilon} G_{k,l} - C_{2\varepsilon} \rho_l \varepsilon_l) + \alpha_l \rho_l \Pi_{\varepsilon,l} \quad (28)$$

Where, σ_k , σ_ε , C_μ , $C_{1\varepsilon}$ and $C_{2\varepsilon}$ are constants, and the default parameter settings of the RNG k - ε model are used in the present work. $G_{k,l}$ is the production rate of turbulent kinetic energy. $\Pi_{k,l}$ and $\Pi_{\varepsilon,l}$ are source terms that take the bubble induced turbulence into the account, and computed as Simonin and Viollet (1990):

$$\Pi_{k,l} = \frac{K_{gl}}{\alpha_l \rho_l} (k_{gl} - 2k_l + (\mathbf{u}_g - \mathbf{u}_l) \cdot \mathbf{u}_{dr}) \quad (29)$$

$$\Pi_{\varepsilon,l} = C_{3\varepsilon} \frac{\varepsilon_l}{k_l} \Pi_{k,l} \quad (30)$$

Where, K_{gl} is inter-phase momentum exchange coefficient which will be discussed in the next section. k_{gl} is covariance of the velocities of liquid and bubble. $\mathbf{u}_{dr}$ is the drift velocity. Especially, $C_{3\varepsilon}$ uses the default constant 1.3.

The turbulence quantities for the gas phase are not obtained from transport

equations. Time and length scales that characterize the motion are used to evaluate dispersion coefficients, correlation functions, and the turbulent kinetic energy of each dispersed phase. Further details of the theory are available elsewhere (ANSYS Inc., 2011).

3.3 The inter-phase momentum closure

Drag force. The drag force reflects the resistance caused by the relative motion of bubbles to the surrounding liquid, which is computed according to:

$$\mathbf{F}_{lg}^{drag} = -\mathbf{F}_{gl}^{drag} = K_{lg} (\mathbf{u}_l - \mathbf{u}_g) = \frac{3C_d}{4d_b} \alpha_g \rho_l |\mathbf{u}_l - \mathbf{u}_g| (\mathbf{u}_l - \mathbf{u}_g) \quad (31)$$

Where, $\mathbf{F}_{lg}$ denotes the momentum transfer terms from the gas phase to the liquid phase and vice versa for $\mathbf{F}_{gl}$. When the population balance model is used, the Sauter mean diameters of bubbles change in a wide range. The drag force coefficient model is slightly modified considering the influence of gas holdup than the original Ishii-Zuber drag model (nonspherical bubble drag model) (1979):

$$\text{spherical: } C_{d,spherical} = \frac{24}{\text{Re}} (1 + 0.1 \text{Re}^{0.75}) \quad (32)$$

$$\text{distorted: } C_{d,distorted} = \frac{2}{3} \text{Eo}^{0.5} \left(\frac{1 + 17.67(1 - \alpha_g)^{9/7}}{18.67(1 - \alpha_g)^{1.5}} \right)^2 \quad (33)$$

$$\text{cap: } C_{d,cap} = \frac{8}{3} (1 - \alpha_g)^2 \quad (34)$$

If $C_{d,spherical} > C_{d,distorted}$, $C_d = C_{d,spherical}$ else $C_d = \min(C_{d,distorted}, C_{d,cap})$

Where, Re is Reynolds number of bubbles, Eo is Eötvös number of bubbles, $\text{Eo} = g(\rho_l - \rho_g) \cdot d_b^2 / \sigma$.

Lift force. Due to velocity gradients in radial and azimuth directions, bubbles rising in

1 a liquid are subjected to a lateral lift force. The force can be correlated to the relative
2 velocity and the local liquid vorticity as:

$$3 \quad \mathbf{F}_{lg}^{lift} = -\mathbf{F}_{gl}^{lift} = -C_{lift} \rho_l \alpha_g (\mathbf{u}_l - \mathbf{u}_g) \times (\nabla \times \mathbf{u}_l) \quad (35)$$

4 For buoyancy-driven gas-liquid bubble flows, the Tomiyama et al.'s (2002)
5 model is used for the computation of the lift coefficient and can be calculated as:

$$6 \quad C_{lift} = \begin{cases} \min[0.288 \tanh(0.121 \text{Re}), f(\text{Eo}_d)] & \text{Eo}_d < 4 \\ f(\text{Eo}_d) & 4 \leq \text{Eo}_d \leq 10 \\ -0.27 & 10 < \text{Eo}_d \end{cases} \quad (36)$$

$$7 \quad f(\text{Eo}_d) = 0.00105 \text{Eo}_d^3 - 0.0159 \text{Eo}_d^2 - 0.0204 \text{Eo}_d + 0.474 \quad (37)$$

8 Where, $\text{Eo}_d = g(\rho_l - \rho_g) \left(d_b \sqrt[3]{1 + 0.163 \text{Eo}^{0.757}} \right)^2 / \sigma$ is the modified Eötvös
9 number computed using the long axis of the deformable bubble. The aspect ratio of
10 the maximum vertical dimension to the maximum horizontal dimension of the
11 spheroidal bubble was evaluated using Wellek et al.'s (1966) empirical correlation.

12 The TFM-PBM simulation is conducted through coupling the gas-liquid two
13 fluid dynamics with the population balance model (PBM). The PBM solution
14 provides the bubble size distribution, and the bubble breakup and coalescence are
15 strongly linked with kinetic energy dissipation rate of the liquid phase. Once the
16 bubble size distribution is obtained, the Sauter mean diameters (SMD) of the two
17 bubble groups are computed as follows:

$$18 \quad d_b = \frac{\sum_{i=1}^{N_{gas}} n_i d_i^3}{\sum_{i=1}^{N_{gas}} n_i d_i^2} \quad (38)$$

19 Of note, the SMD of the small bubble group is smaller than 5.8 mm, and the
20 large bubble group is larger than 5.8 mm. The values of SMD are important for the

computation of the interphase momentum exchanges including the drag force and the lift force.

4. Results and discussions

4.1. Description of the simulation cases and the operation conditions

In order to quantitatively verify the present bubble coalescence and breakup model, a series of simulations were performed based on the experiment setups of Kagumba and Al-Dahhan (2015) and Chen et al. (1998). In Kagumba's experiments, a bubble column of 0.14 m diameter and 1.83 m height was operated maintaining a constant level of 1.56 m with superficial gas velocities of 0.03 and 0.45 m/s, respectively. A four-point optical probe was used to measure the bubble sizes. Note that only the mean and the variance of the bubble diameters were recorded in their experiments, and the reported bubble size distributions were modeled using a log-normal probability density function shown below (Akita and Yoshida, 1974).

$$pdf = f(d_b) = \frac{1}{\sigma d_b \sqrt{2\pi}} \exp \left[-\frac{(\ln d_b - \mu)^2}{2\sigma^2} \right] \quad (39)$$

$$\mu = \log \frac{m^2}{\sqrt{v + m^2}}; \sigma = \sqrt{\log \left(\frac{m^2 + v}{m^2} \right)}$$

Although the gas holdup and bubble sizes were measured at H/D=5.1 with and without internals, only the data of an empty column will be used for comparison in the present work.

Another experiment of Chen et al. (1998) was conducted in a 0.44 m diameter column with a height of 2.43 m. The superficial gas velocities of 0.02 and 0.1 m/s

1 were used to represent the bubbly and the churn-turbulent flow regimes, respectively.
2 For each experimental case, the axial liquid velocity was measured by a computer
3 automated radioactive particle tracking technique (CARPT) at different axial
4 positions, and the local gas holdup was obtained by computed tomography (CT). The
5 experiment data has been well-documented and used as a benchmark in several
6 simulation works (Chen et al., 2005; Xu et al., 2013). Further details of the
7 experimental setup and procedures of measurement are referred to Chen et al. (1998).

8 Firstly, the new bubble coalescence and breakup model was validated at different
9 superficial gas velocities for a 0.14 m diameter column followed with a numerical test
10 of the grid independence. The bubble size distributions were obtained from the
11 simulations and compared with experimental data (section 4.2). The inhomogeneous
12 population balance model was adopted and the interfacial drag and lift forces were
13 included. Secondly, the generic TFM-PBM model was further applied in a 0.44 m
14 column operated at different gas velocities, and the gas holdup and axial liquid
15 velocities were simulated and compared with literature data (section 4.3). In addition,
16 the effect of the lift force was investigated in section 4.4.

17 The numerical simulations were conducted using the ANSYS Fluent 14.0. The
18 QUICK discretization scheme was used for volume fraction and momentum equations,
19 and the second order upwind scheme was specified to discretize other equations.
20 Water and air at ambient temperature and pressure were used as continuous and
21 dispersed phases, respectively. For all simulations, a velocity inlet was used for the
22 bottom of the column and a pressure outlet was used for the top of the column. The

1 wall boundary conditions of gas and liquid in a cylindrical bubble column were
2 free-slip and no-slip, respectively. A time step of 0.01 s was used and the simulation
3 results were averaged over 80 s after the flow pattern reached a quasi-steady state.

4 5 4.2. Verification of the bubble coalescence and breakup model with data of $D=0.14$ m 6 bubble column

7 A coarse grid I and a fine grid II were used in the simulations of a 0.14 m column
8 with a superficial velocity 0.45 m/s. Hexahedral, structured computational cells were
9 employed, and the near wall grid was refined. **Fig. 6** shows the cross-sectional mesh
10 structures of two grids used in the simulations. For grid I, the axial direction
11 (z-direction) size of the grid is uniformly set to 0.0154 m, and the radial size of the
12 grid is about 0.0045 m. The total grid number is 76,800. For grid II, the axial direction
13 size of the grid is set to 0.0128 m, and the radial direction size is about 0.0032 m. The
14 total grid number is 144,000. The simulation results of the time-averaged radial
15 distributions of gas holdup and axial liquid velocity are shown in **Fig. 7a** and **7b**. It is
16 confirmed that the predicted gas holdup and the axial liquid velocities using the two
17 meshes are nearly the same. Therefore, the grid I set-up of grid number 76800 is
18 employed in the following simulations.

Grid I

Grid II

Fig. 6. The cross-sectional mesh set-ups of Grid I and Grid II used in the simulations of Fig.7. Number of grid I: 76,800; Number of grid II: 144,000.

Fig.7. Effects of numerical grids on the simulated radial profiles of the time-averaged (a) gas holdup, and (b) liquid axial velocity.

The bubble size distribution represents the results of competitions due to bubble

1 coalescence and breakup occurring simultaneously. In a given system, bubble
2 coalescence and breakup are primarily influenced by the turbulent kinetic energy
3 dissipation rate of the liquid phase especially when the column is operated in a
4 churn-turbulent regime. **Fig. 8** shows the simulated bubble number density
5 distributions at two superficial velocities 0.03 and 0.45 m/s, respectively. The
6 log-normal distributions computed using equation (4-1) is shown in the same plot for
7 comparison. Based on the measurements of Kagumba and Al-Dahhan (2015), the
8 mean bubble sizes were 5.1 and 8.5 mm for the superficial gas velocities 0.03 and
9 0.45 m/s.

10 As is shown in **Fig. 8**, the bubble size distribution with a high superficial gas
11 velocity is flatter than that with a low superficial gas velocity. The reason is that as the
12 superficial gas velocity increases, the turbulent kinetic energy dissipation rates
13 increase significantly (shown in **Fig. 9**), hence, the mean bubble sizes decrease
14 dramatically due to the increase in the bubble breakup rates. **Fig. 10** shows the radial
15 distributions of the simulated time-averaged mean bubble sizes of the large and small
16 bubble groups for different superficial gas velocities. At a higher superficial gas
17 velocity, the mean bubble sizes of both groups are constantly smaller than the case of
18 a lower superficial gas velocity, confirming the dominating effects of the bubble
19 breakup due to increased turbulent dissipation rates of the liquid phase. In addition,
20 the mean sizes of the bubbles larger than 10 mm based on the simulated bubble size
21 distributions as shown in **Fig. 8** are also calculated, and the diameters are 14.3 and
22 12.7 mm for the superficial gas velocities 0.03 and 0.45 m/s, respectively. The mean

1 bubble size of the large bubble group is significantly smaller than the size computed
2 by using the correlation of Krishna et al. (2001), where the large bubbles were defined
3 as having diameters larger than 10 mm. Because the correlation of Krishna et al. was
4 obtained based on a two-dimensional bubble column of air/paraffin oil slurry and the
5 effects of the slurry viscosity was not included, the discrepancy between the present
6 simulation and the empirical correlation is incomparable.

7 In **Fig. 8**, the simulation results of bubble coalescence and breakup model, which
8 only take the turbulence into the account, are presented. At a higher gas velocity, the
9 bubble size distribution does not show obvious differences, indicating that compared
10 with other factors, turbulence plays a more important role, indeed. However, the
11 phenomenon that more large bubbles are produced with a low superficial velocity
12 reveals that the effect of velocity gradient shear induced bubble breakup becomes
13 remarkable gradually, especially when the turbulence is relatively weak.

14 The gas holdup is another important parameter describing the hydrodynamics
15 behavior of a bubble column, and is closely correlated with superficial gas velocities.
16 As has been reported using the same gas velocity, predicted gas holdup distribution
17 with a smaller bubble size was higher than a larger one (Xu et al, 2013); the
18 simulation results of the bubble size distribution play a critical role for a reliable
19 prediction of the gas holdup of a bubble column. **Fig. 11** compares the simulated
20 radial profiles of time-averaged gas holdup with the experiment results, at superficial
21 gas velocities 0.03 and 0.45 m/s, respectively. The simulation results show reasonable
22 agreement with the experimental data. The simulated gas holdup of the large and

1 small bubble groups are shown in the same plot. The contribution of the large bubble
 2 group is significantly greater than the small bubble group, indicating that the
 3 movement of large bubbles plays an important role in determining the gas-phase
 4 structure and distributions of the gas holdup.

5

6

7 Fig. 8. Predicted and measured bubble number density distributions for different
 8 superficial gas velocities. (Experimental data given by Kagumba and Al-Dahhan,
 9 2015)

10

11

12 Fig. 9. Radial profiles of time-averaged turbulent kinetic energy dissipation rate for
 13 different superficial gas velocities.

14

Fig. 10. Radial profiles of time-averaged mean bubble sizes of the large and small bubble groups for different superficial gas velocities.

Fig. 11. Radial profiles of time-averaged gas holdup for different superficial gas velocities: (red circles) Exp., $U_g=0.45$ m/s; (red solid line) Sim., $U_g=0.45$ m/s, Local total gas holdup; (red dashed line) Sim., $U_g=0.45$ m/s, Large bubble gas holdup; (red dotted line) Sim., $U_g=0.45$ m/s, Small bubble gas holdup; (green squares) Exp., $U_g=0.03$ m/s; (green solid line) Sim., $U_g=0.03$ m/s, Local total gas holdup; (green dashed line) Sim., $U_g=0.03$ m/s, Large bubble gas holdup; (green dotted line) Sim., $U_g=0.03$ m/s, Small bubble gas holdup. (Experimental data given by Kagumba and Al-Dahhan, 2015)

4.3. Application of TFM-PBM model in the simulation of $D=0.44$ m bubble column

The above verified coalescence and breakup model in a small bubble column is further applied in the simulation of a well-documented 0.44 m air-water bubble

1 column at superficial gas velocities of 0.02 and 0.1 m/s, respectively. 76800 grids are
2 used in the simulations based on previous grid sizes set-up (Chen et al., 2005). The
3 simulation results are shown in **Fig. 12** and **Fig. 13**.

4 As is shown in **Fig. 12a**, the gas holdup decreases along the radial direction. The
5 gas holdup is high in the center and low near the wall of the column and this leads to a
6 gross liquid circulation throughout the column with liquid ascending in the center and
7 descending near the wall (**Fig. 12b**). The present model prediction is in a good
8 agreement with experimental results, especially for the case of superficial gas velocity
9 0.1 m/s. For the case of superficial gas velocity 0.02 m/s, a distinct difference is noted
10 between the simulated gas holdup and the literature data. In the present bubble
11 coalescence and breakup models, the bubble induced turbulence has been modeled as
12 a major driving force inducing bubble collisions, and the subsequent bubble
13 coalescence and breakup. The results indicate that the RNG k- ϵ model used in the
14 present simulations worked better for some cases with high superficial gas velocities,
15 where the bubble induced turbulence is strong and the flow structure close to isotropic.
16 For the case of a very low superficial gas velocity with large scale bubble column
17 diameter, the bubble induced turbulence is very weak and probably close to
18 anisotropic bubbly flow (Chen et al., 1998), the RNG k- ϵ model might not be
19 applicable. Besides, in the same plot, the radial profiles of time-average gas holdup
20 and liquid axial velocities have been predicted using the empirical correlations of
21 Al-Dahhan and coworkers (2001a, 2001b), and the present simulation results are in
22 line with the empirical correlations.

Fig. 12. Radial profiles of time-averaged (a) gas holdup, and (b) liquid axial velocity at different superficial gas velocities. (Experimental data given by Chen et al. 1998; Predicted data given by Wu and Al-Dahhan, 2001a, 2001b)

Fig. 13a illustrates the effects of superficial gas velocity on the bubble size distributions. The mean diameter at a high gas velocity is smaller than that of a low velocity. **Fig. 13b** shows that in the central region the variations of the bubble size distributions are not significant. However, in the wall region, the mean bubble size becomes smaller, indicating that small bubbles tend to move toward the wall region.

Recalling that the simulation results for a column of diameter 0.14 m running at a superficial gas velocity 0.03 m/s agreed reasonably well with the experiments, as

1 shown in **Fig. 8** and **Fig. 11**, we speculate that either superficial gas velocity or bubble
2 column diameter is a critical parameter for the turbulence intensity. To confirm, a
3 simulation using a superficial gas velocity 0.03 m/s for the 0.44 m column is
4 performed, the predicted radial profile of time-averaged gas holdup and the bubble
5 size distribution are shown in **Fig.12a** and **Fig.13a**, respectively. As the superficial gas
6 velocity increases from 0.02 m/s to 0.03 m/s, the time-averaged gas holdup increases
7 significantly, and the bubble sizes decrease dramatically due to much higher rates of
8 bubble breakup. It is noted that the time-averaged gas holdup profiles for the two
9 columns running at the same superficial gas velocity 0.03 m/s are comparable;
10 demonstrating the superficial gas velocity is the most important parameter
11 determining the level of the bubble induced turbulence intensity. Therefore, it is
12 concluded that the present simulation model is applicable to a superficial velocity as
13 low as 0.03 m/s. Further work is needed for simulating the anisotropic turbulence of
14 bubble column running at very low superficial gas velocities lower than 0.03 m/s.

15

16

Fig.13. (a) Effects of superficial gas velocity on the bubble size distributions; (b) Bubble size distributions sampled at various positions over a column $H/D=1-4$.

4.4. The effects of lift force

In order to predict the hydrodynamics behavior effectively in a bubble column, the effect of lift force was examined. For a buoyancy-driven bubbly flow, lift force plays an important role in determining the hydrodynamics behavior of reactor. **Fig. 14a** and **14b** compared the results of simulations with and without the lift force using the inhomogeneous model. It is clear that when the lift force is excluded in the simulations, the predicted profiles of time-averaged gas holdup and liquid axial velocity shows large discrepancies compared to experiment data. The simulation result is improved when the lift force is included. In fact, the lateral lift force is determined by both the velocity gradient and bubble diameter, which makes large bubbles move toward the central region and small ones toward the wall region. The aggregate of large bubbles in center region forms more larger bubbles, and these fast ascending large bubbles lead to a decrease of time-averaged gas holdup. In addition, the direction of lift force changing with bubble sizes implies that use of the

1 inhomogeneous PBM model is more reasonable for capturing the gas phase fluid
 2 dynamics when the effect of lift force is included.

3

4

5

6 Fig.14. Effects of lift force on time-averaged radial distributions (a) gas holdup, and
 7 (b) liquid axial velocities. (Experimental data given by Chen et al. 1998)

8

9 5. Conclusion

10 A generic TFM-PBM simulation scheme was developed in combination of
 11 bubble breakup and coalescence models. The bubble coalescence was modeled by
 12 considering bubble collisions induced by turbulent fluctuation, buoyancy driven,
 13 wake entrainment, and viscous shear. An improved bubble breakup model was
 14 implemented in the ANSYS Fluent 14.0 software package. By taking into account

1 different mechanisms of bubble coalescence and breakup, the bubble size
2 distributions were obtained from the simulations and compared with experimental
3 data. It was found that simulated log-normal bubble size distributions using the
4 present model agreed well with literature data of a 0.14 m bubble column at different
5 superficial gas velocities. The simulation results showed that the bubble size
6 distribution at a high superficial gas velocity was flatter than that at a low superficial
7 gas velocity. The model was further applied in a 0.44 m column operated at different
8 gas velocities, and the gas holdup and axial liquid velocities were simulated and
9 compared with well documented literature data. The model showed the capacity of
10 describing the gas-liquid fluid dynamics of bubble columns operated in a
11 churn-turbulent flow regime, and a bubbly regime with a superficial gas velocity
12 greater than 0.03 m/s. In addition, the effect of lift force was also investigated and the
13 results showed that by taking the lift force into account, the distribution of gas holdup
14 improved significantly at r/R equal to 0.6-0.8, indicating that the lateral lift force was
15 indispensable, especially in the case of the inhomogeneous PBM model.

16

17 **Notation**

18 *Roman letters*

19	A	bubble cross-sectional area, m^2
20	A_{ij}	bubble impact area, m^2
21	$b(v_i : v_j)$	breakup frequency, s^{-1}
22	B_B	production of bubble by breakup, $kg^{-1} m^{-3} s^{-1}$

1	B_D	destruction of bubble by breakup, $\text{kg}^{-1} \text{m}^{-3} \text{s}^{-1}$
2	c_f	surface area increase coefficient
3	$c(v_i, v_j)$	coalescence rate, $\text{m}^{-3} \cdot \text{s}^{-1}$
4	$C_{1\varepsilon}, C_{2\varepsilon}, C_{3\varepsilon}, C_\mu$	constants, 1.42, 1.68, 1.3, 0.0845
5	C_B	production of bubble by coalescence, $\text{kg}^{-1} \text{m}^{-3} \text{s}^{-1}$
6	C_D	destruction of bubble by coalescence, $\text{kg}^{-1} \text{m}^{-3} \text{s}^{-1}$
7	C_d	drag coefficient
8	C_{lift}	lift coefficient
9	d	bubble diameter, m
10	d_b	Sauter mean bubble diameter, m
11	D	bubble column diameter, m
12	Eo	Eötvös number
13	f_{bv}	bubble breakup volume fraction
14	$\mathbf{F}_{gl}, \mathbf{F}_{lg}$	interfacial momentum exchange term, $\text{kg m}^{-2} \text{s}^{-2}$
15	g	acceleration due to gravity, 9.81 m s^{-2}
16	$G_{k,l}$	production of turbulent energy, $\text{kg m}^{-1} \text{s}^{-3}$
17	h_0, h_f	liquid film thickness, m
18	H	liquid height, m
19	k	turbulent kinetic energy, $\text{m}^2 \text{s}^{-2}$
20	k_{gl}	covariance of the velocities of liquid and bubble, $\text{m}^2 \text{s}^{-2}$
21	K_{gl}, K_{lg}	inter-phase momentum exchange coefficient, $\text{kg m}^{-3} \text{s}^{-1}$
22	n	number of bubbles per unit volume, m^{-3}

1	$\dot{n}_\lambda$	number of eddies per unit volume, m^{-4}
2	N	number of bubble bins
3	$p_c(d_i, d_j)$	coalescence efficiency
4	$p_b(d_i : d_{j,k})$	breakup efficiency
5	pdf	bubble size distribution, m^{-1}
6	r_{ij}	equivalent radius, m^{-1}
7	Re	Reynolds number
8	S_i	Source term of i-th bubble group, $\text{m}^{-3} \text{s}^{-1}$
9	t	time, s
10	$t_{\text{contact}}, t_{\text{drainage}}$	contact/drainage time between bubbles, s
11	$\mathbf{u}$	velocity vector, m s^{-1}
12	$\mathbf{u}_{dr}$	drift velocity vector, m s^{-1}
13	u_b	bubble rise velocity, m s^{-1}
14	u_t	turbulent velocity, m s^{-1}
15	u_v	liquid velocity gradient, m s^{-1}
16	u_w	wake entrainment velocity, m s^{-1}
17	$\bar{u}_\lambda$	eddies turbulent velocity, m s^{-1}
18	U_g	superficial gas velocity, m s^{-1}
19	u, v, w	velocity component, m s^{-1}
20	v	bubble volume, m^3
21	w	collision frequency, $\text{m}^{-3} \text{s}^{-1}$
22	x, y, z	spatial coordinates, m

1

2 *Greek letters*

3 α void fraction

4 ε turbulent dissipation rate, $\text{m}^2 \text{s}^{-3}$

5 γ shear strain rate, s^{-1}

6 λ eddy size, m

7 μ dynamic viscosity, $\text{kg m}^{-1} \text{s}^{-1}$

8 π ≈ 3.1415926

9 ρ density, kg m^{-3}

10 σ surface tension, kg m^{-2}

11 $\sigma_k, \sigma_\varepsilon$ dispersion Prandal number, 0.75, 0.75

12 τ stress, kg m^{-3}

13 ξ eddy size divided by parent bubble size

14 $\Pi_{k,l}$ bubble-induced turbulence term, $\text{m}^2 \text{s}^{-3}$

15 $\Pi_{\varepsilon,l}$ bubble-induced turbulence term, $\text{m}^2 \text{s}^{-4}$

16 Θ coefficient for wake entraining

17

18 *Subscripts*

19 b bubble index

20 g gas index

21 l liquid index

22 i, j phase index, number index

1

2 **Acknowledgements**

3 This work is supported by China's National Science Foundation (NSFC) under
4 grant 21276085.

5

6 **References**

7 ANSYS Inc., 2011. ANSYS FLUENT-14.0 Theory Guide.

8 Akita, K., Yoshida, F., 1974. Bubble size, interfacial area, and liquid-phase mass
9 transfer coefficient in bubble columns. *Ind. Eng. Chem.* 13, 84-91.

10 Chen, C., Fan, L. S., 2004. Discrete simulation of gas-liquid bubble columns and
11 gas-liquid-solid fluidized beds. *AIChE J.* 50, 288-301.

12 Chen, J., et al., 1998. Gas holdup distributions in large-diameter bubble columns
13 measured by computed tomography. *Flow Measurement and Instrumentation.* 9,
14 91-101.

15 Chen, P., Sanyal, J., Dudukovic, M. P., 2004. CFD modeling of bubble columns flows:
16 implementation of population balance. *Chem. Eng. Sci.*, 59, 5201-5207.

17 Chen, P., Dudukovic, M. P., Sanyal, J., 2005. Three-dimensional simulation of bubble
18 column flows with bubble coalescence and breakup. *AIChE J.* 51, 696-712.

19 Chesters, A. K., 1991. The modelling of coalescence processes in fluid-liquid
20 dispersions: a review of current understanding. *Trans. IChemE.* 69, 259-270.

21 Cheung, S. C. P., Yeoh, G. H., Tu, J. Y., 2007. On the numerical study of isothermal
22 vertical bubbly flow using two population balance approaches. *Chem. Eng. Sci.* 62,

1 4659-4674.

2 Clift, R., Grace, J. R., Weber, M. E., 1978. Bubbles Drops and Particles. Academic
3 Press, New York.

4 Coulaloglou, C. A., Tavlarides, L. L., 1977. Description of interaction processes in
5 agitated liquid-liquid dispersions. Chem. Eng. Sci. 32, 1289-1297.

6 Ekambara, K., Nandakumar, K., Joshi, J. B., 2008. CFD simulation of bubble column
7 reactor using population balance. Ind. Eng. Chem. Res. 47, 8505-8516

8 Friedlander, S. K., 1977. Smoke, Dust and Haze, Wiley, New York.

9 Hageseather, L., Jakobson, H. A., Svendsen, H. F., 2002. A model for turbulent
10 binary breakup of dispersed fluid particles. Chem. Eng. Sci. 57, 3251-3267.

11 Howarth, W. J., et al., 1964. Coalescence of drops in a turbulent flow field. Chem.
12 Eng. Sci. 19, 33-38.

13 Ishii, M., Zuber, N., 1979. Drag coefficient and relative velocity in bubbly, droplet or
14 particulate flows. AIChE J. 25, 843-855.

15 Kagumba, M., Al-Dahhan, M. H., 2015. Impact of internals size and configuration on
16 bubble dynamics in bubble columns for alternative clean fuels production. Ind. Eng.
17 Chem. Res. 54, 1359-1372

18 Krepper, E., et al., 2008. The inhomogeneous MUSIG model for the simulation of
19 polydispersed flows. Nucl. Eng. Des. 238, 1690-1702.

20 Krishna, R., et al., 2001. Design and scale-up of a bubble column slurry reactor for
21 Fischer-Tropsch synthesis. Chem. Eng. Sci. 56, 537-545.

22 Laborde-Boutet, C., et al., 2009. CFD simulation of bubble column flows:

1 Investigations on turbulence models in RANS approach. Chem. Eng. Sci. 64,
2 4399-4413.

3 Lehr, F., Mewes, D., 2001. A transport equation for the interfacial area density
4 applied to bubble columns, Chem. Eng. Sci. 56, 1159-1166.

5 Lehr, F., Millies, M., Mewes, D., 2002. Bubble-size distributions and flow fields in
6 bubble columns. AIChE J. 48, 2426-2443.

7 Liao, Y., Lucas, D., 2009. A literature review of theoretical models for drop and
8 bubble breakup in turbulent dispersions. Chem. Eng. Sci. 64, 3389-3406.

9 Liao, Y., Lucas, D., 2010. A literature review on mechanisms and models for the
10 coalescence process of fluid particles. Chem. Eng. Sci. 65, 2851-2864.

11 Liao, Y., et al., 2015. Baseline closure model for dispersed bubbly flow: Bubble
12 coalescence and breakup. Chem. Eng. Sci. 122, 336-349.

13 Lucas, D., Krepper, E., Prasser, H. M., 2003. Evolution of flow patterns, gas fraction
14 profiles and bubble size distributions in gas-liquid flows in vertical tubes. Trans. Inst.
15 Fluid-Flow. 112, 37-46.

16 Luo, H., Svendsen, H. F., 1996. Theoretical model for drop and bubble breakup in
17 turbulent dispersions. AIChE J. 42, 1225-1233.

18 Martinez-Bazan, C., et al., 1999. On the breakup of an air bubble injected into a fully
19 developed turbulent flow. Part 1. Breakup frequency. J. Fluid Mech. 401, 157-182.

20 Martinez-Bazan, C., et al., 2010. Considerations on bubble fragmentation models. J.
21 Fluid Mech. 661, 159-177.

22 Oey, R. S., et al., 2003. Sensitivity study on interfacial closure laws in two-fluid

- 1 bubbly flow simulations. AIChE J. 49, 1621-1636.
- 2 Orszag, S. A., et al, 1993. Renormalization Group Modeling and Turbulence
- 3 Simulations. International Conference, Tempe, Arizona.
- 4 Prince, M. J.; Blanch, H. W., 1990. Bubble coalescence and break-up in air-sparged
- 5 bubble columns. AIChE J. 36, 1485-1499.
- 6 Rotta, J.C., 1972. Turbulence. Stromungen, B.G.Teubner, Stuttgart.
- 7 Rzehak, R., Krepper, E., 2013. CFD modeling of bubble-induced turbulence. Int. J.
- 8 Multiph. Flow 55, 138-155.
- 9 Sanyal, J., et al., 1999. Numerical simulation of gas-liquid dynamics in cylindrical
- 10 bubble column reactors. Chem. Eng. Sci. 54, 5071-5083.
- 11 Shinnar, R., Church, J. M., 1960. Predicting particle size in agitated dispersions. Ind.
- 12 Eng. Chem. 52, 253-256.
- 13 Simonin, C., Viollet, P. L., 1990. Predictions of an oxygen droplet pulverization in a
- 14 compressible subsonic coflowing hydrogen flow. Num. Multiph. Flow. FED91,
- 15 65-82.
- 16 Solsvik, J., Jakobsen, H. A., 2015. Single air bubble breakup experiments in stirred
- 17 water tank. Int. J. Chem. React. Eng. 10, 1-15.
- 18 Sporleder, F., et al., 2012. On the population balance equation. Rev. Chem. Eng. 28,
- 19 149-169.
- 20 Tabib, M. V., Roy, S. A., Joshi, J. B., 2008. CFD simulation of bubble column-An
- 21 analysis of interphase forces and turbulence models. Chem. Eng. J. 139, 589-614.
- 22 Tomiyama, A., et al., 2002. Transverse migration of single bubbles in simple shear

1 flows. Chem. Eng. Sci. 57, 1849-1858.

2 Wang, T., Wang, J., Jin, Y., 2003. A novel theoretical breakup kernel function for
3 bubbles/droplets in a turbulent flow. Chem. Eng. Sci. 58, 4629-4637.

4 Wang, T., Wang, J., Jin, Y., 2006. A CFD-PBM model for gas -liquid flows. AIChE J.
5 52, 125-139.

6 Wellek, R. M., Agrawal, A. K., Skelland, A. H. P., 1966. Shapes of liquid drops
7 moving in liquid media. AIChE J. 12, 854-860.

8 Wu, Y., Al-Dahhan, M. H., 2001a. Prediction of axial liquid velocity profile in bubble
9 columns. Chem. Eng. Sci. 56, 1127-1130.

10 Wu, Y., Ong, B. C., Al-Dahhan, M. H., 2001b. Predictions of radial gas holdup
11 profiles in bubble column reactors. Chem. Eng. Sci. 56, 1207-1210.

12 Xu, L., et al., 2013. Numerical simulation of bubble column flows in churn-turbulent
13 regime: comparison of bubble size models. Ind. Eng. Chem. Res. 52, 6794-6802.

14 Xu, L., et al., 2014. Application of population balance model in the simulation of
15 slurry bubble column. Ind. Eng. Chem. Res. 53, 4922-4930.

16 Zhang, D., Deen, N. G., Kuipers, J., 2005. Numerical simulation of the dynamic flow
17 behavior in a bubble column: A study of closures for turbulence and interface forces.
18 Chem. Eng. Sci. 61, 7593-7608.

19 Zhao, H., Ge, W., 2007. A theoretical bubble breakup model for slurry beds or
20 three-phase fluidized beds under high pressure. Chem. Eng. Sci. 62, 109-115.