

## Influence of butanol addition on mass transfer and bubble diameter in a split-cylindrical airlift reactor

Mostafa Keshavarz Moraveji\*, Baharak Sajjadi, Reza Davarnejad & Saba Sharafoddin Zade  
Department of Chemical Engineering, Faculty of Engineering, Arak University, Arak 38156-8-8349, Iran

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This study is focused on the effect of butanol addition on mass transfer and bubble size dispersion in dilute butanol solutions [from 0 to 1% (v/v)], in a split-cylindrical airlift and compared with pure water. Bubble size distribution is measured by photography and picture analysis. Bubble diameter in dilute butanol solutions is about 3.5 times more than it in pure water. Also increasing of butanol concentration leads to decreasing of bubble diameter about 25%. Increasing of gas velocity inlet by increasing of coalescence rate leads to increasing of bubble diameter, so the biggest bubble observed in the highest gas velocity inlet and lowest butanol concentration. Although butanol provides an additional decrease of mass transfer coefficient, but by reducing bubble diameter, active surface for mass transfer increases that leads to increasing of volumetric mass transfer coefficient. Furthermore, a correlation for volumetric mass transfer coefficient is exhibited using butanol concentrations and gas velocity inlet as independent variables.

**Keywords:** Airlift reactor, Gas-liquid mass transfer, Bubble diameter, Butanol

The airlift reactors (ALR) are introduced as one of the best two-phase contactors, which are used in many chemical, petrochemical and biological processes such as wastewater treatment (nitrification and phenolic treatments) and aerobic fermentation<sup>1-5</sup>. In fact they are a specially modified type of bubble columns in which internal structure has been divided into two zones by a baffle split or draft tube, so there are four main zones in an ALR involving riser, down-comer, bottom and separator. Gas injection into riser region leads to a density difference between riser and down-comer which produce a driving force for liquid circulation<sup>3</sup>. Recently, many studies have been focused on hydrodynamic and mass transfer parameters in ALRs due to their advantages such as high and flexible capacity, simple conceptual design, low shear rate, high mixing performance, better contact between the phases, shorter reaction time, favorite heat transfer, low energy consumption, long life time and low maintenance cost<sup>5,6</sup>. But most of the reports are about systems containing water as the continuous phase and air as the dispersed phase and there are not enough data about systems containing salts, alcohols, surfactants or even oil micro-emulsions, which have different behaviours in different concentrations.

These are used in bioprocesses, waste water treatment like denitrifying<sup>7</sup>, dephosphatation, phenolic treatment<sup>8,9</sup>, waste gas treatment<sup>10</sup>, chemical and biotechnological industries like oxidation, chlorination<sup>11</sup>, production of biological origin like organic acids, alcohols, antifoams, alkaloids, antibiotics and proteins<sup>12,13</sup>. Also aqueous solutions of alcohols such as ethanol and butanol are encountered in the production of bio-fuels by fermentation process<sup>14,15</sup>. In some cases, alcohols are used as a carbon source in biological processes<sup>16</sup>.

Bubble size distribution, which is necessary for simulation, effective on volume fraction of gas in liquid and determine the active surface of mass transfer. Dhanasekharan *et al.*<sup>17</sup>, by investigation of bubble size distribution with population balance model and solving it, by computational fluid mechanics code, have investigated gas hold-up and mass transfer coefficient in different superficial gas velocities and could predict them in different superficial gas velocities. In addition, there are useful studies on break-up and coalescence model in pure water<sup>18-21</sup>. For example; Wang *et al.*<sup>18</sup>, have developed a theoretical break-up kernel function based on an eddy-bubble/droplet collision method, for bubbles/droplets in a fully turbulent flow, and have found an increase in the energy dissipation rate and mother bubble/droplet size increases the probability

\*Corresponding author (E-mail: m-moraveji@araku.ac.ir)

of unequal break-up. Wang *et al.*<sup>19</sup>, in another study based on the bubble size distribution by the population balance model (PBM), have predicted the flow regime transition in bubble columns theoretically. Bulk liquid properties such as density, viscosity, and surface tension, also interfacial properties of two-phase and sparger type are the key parameters which affect on bubble size distribution<sup>19</sup>. Thorpe *et al.*<sup>20</sup> have considered the effects of sparger design [peripheral sparger (PS) and a plunging jet sparger (PJS)] on the resulting dispersed bubble size and its distribution and the volumetric gas-liquid mass transfer coefficient. In this paper, we used a porous sparger that can produce uniform radial bubble distribution<sup>20</sup>.

Ruen-ngam *et al.*<sup>21</sup> have investigated the influence of salinity on bubble size distribution and gas-liquid mass transfer in an airlift reactor and they have found in saline water, bubble diameter decreased with  $U_g$  due to the increasing of collision and break-up of bubbles at high gas holdup so specific area ( $a$ ) was high in the saline water systems, however, the mass transfer coefficient ( $K_L$ ) was higher in the fresh water system than saline water. Homayouni *et al.*<sup>22</sup> have investigated bubble size distribution in bubble containing three petroleum-based liquids but they have focused on developing population model balance.

According to the literature, diluted non-viscous aqueous alcohol solutions were used for liquid phase simulation in coal liquefaction process in a bioreactor<sup>23,24</sup>. The surface tension of alcohol

solutions considerably was different in comparison with pure water<sup>25</sup>. Furthermore, diluted aqueous alcohol solutions were used as surfactants to prevent the coalescence phenomenon. The aim of this work is to investigation of butanol addition and its concentration on bubbles diameter in different gas velocity inlet and the influence of it on volumetric mass transfer coefficient in a split cylindrical ALR. This information will be useful in the future design and scale up of the airlift reactors for specific applications because for scaling up and prediction of airlift reactor behavior, being aware from experimental and practical results are so important, nowadays, correlation and simulation are the best methods for scaling up which both of them need experimental data. The correlation and simulation decrease the cost and time consumption.

## Materials and Methods

### Materials

The liquid phase was aqueous solutions of butanol with the concentrations of alcohols ranged from 0.25 to 1% (v/v) that were compared with pure water. Butanol (99.9% and  $\rho=0.81 \text{ g/cm}^3$ ) was purchased from Merck Company (Germany) and its various concentrations were locally prepared. Since the diluted alcohol solutions behave as coalescence inhibitor (e.g. surfactant) after the transition concentrations<sup>26</sup>.

### Apparatus set-up

The split-cylinder airlift reactor and the flow scheme used are schematically shown in Fig. 1. The

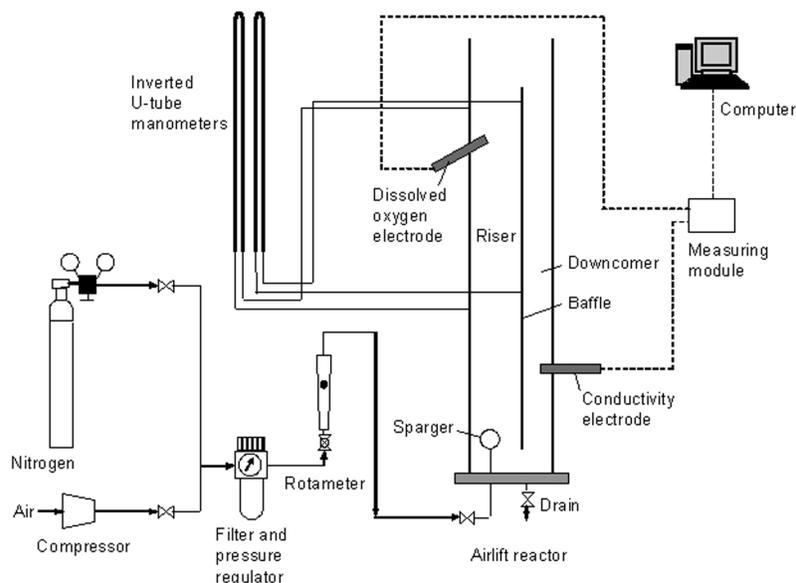


Fig. 1– Schematic diagram of the split-cylinder airlift reactor

reactor is involving a glass column with 1.5 m height and 0.136 m in diameter. A rectangular Plexiglass baffle with 0.129 m wide, 1.0 m height and 0.005 m thickness was inserted in the glass column to divide the cross-section into riser and down-comer zones (with the riser area of 86.115 cm<sup>2</sup> and down-comer area of 40.299 cm<sup>2</sup> and the ratio of 2.136 riser per down-comer). The baffle was also located at 0.1 m from the bottom of reactor. The gas-free liquid height in the reactor was about 1.23 m for all experiments. The gas sparger with 0.02 m diameter sintered ceramic ball located at the bottom of the riser. The volumetric flow rate of air in the riser zone was controlled by using a regulating valve and a calibrated rotameter. Also dissolved oxygen electrode was used for mass transfer measurement. Conductivity electrode and inverted U-tube manometers were used for liquid circulation velocity and hold-up measurement. All experiments were carried out at ambient conditions (atmospheric pressure and 25±0.5°C).

**Results and Discussion**

Table 1 shows the used solutions properties such as density and surface tension. The surface tension was measured by a tensiometer (KRUSGmbH, Hamburg, Germany), and the density was determined by using the buoyancy method<sup>27</sup>. According to Table 1, butanol addition to pure water and its concentration increment causes surface tension and density reduction.

**Overall volumetric gas liquid mass transfer coefficient**

Oxygen desorption method was used to measure the volumetric mass transfer coefficient<sup>3</sup>. According to this method the liquid phase is deoxygenated by stripping it with nitrogen, and then re-aerated. An oxygen electrode (model: WTWCelox325) located horizontally 0.1 m below the exit of the riser with angle of 30° to the horizontal position (for preventing oxygen bubbles gathering near the sensor). Output signals obtained from the dissolved oxygen are transmitted to the computer using a DO-meter (model:

WTWOXI197-S). Typical response signal is shown in Fig. 2. In the first section of this figure (desorption), nitrogen gas is injected into the liquid phase until oxygen concentration reaches 0 mg/L and in the second section (aeration), the oxygen-depleted liquid was re-aerated until relatively high dissolved oxygen concentration were reached.

The dissolved oxygen concentration is measured as a function of time using Eq. (1):

$$\frac{dC_L}{dt} = K_L.a.(C_L^* - C_L) \quad \dots (1)$$

where, C\* and C<sub>L</sub> are saturated oxygen concentration and the DO concentration at time t. K<sub>L</sub>a is independent of time and can be calculated using Eq. (2):

$$\ln\left(\frac{C^* - C_L}{C^* - C_0}\right) = -k_L a t \quad \dots (2)$$

Figure 3 shows the volumetric mass transfer coefficient, K<sub>L</sub>a, versus superficial gas velocity. As shown in this figure, K<sub>L</sub>a increased with adding butanol and increasing the superficial gas velocity. At low superficial gas velocity, the effect of butanol addition is not much on volumetric mass transfer coefficient while at high superficial gas velocity; butanol addition decreased the bubbles diameter and increased the interfacial area (a) increased. Therefore, volumetric mass transfer coefficient increased. This output is supported by literatures<sup>28-30</sup>, although Dhaoudi *et al.*<sup>31</sup> and El Azher *et al.*<sup>32</sup> have mentioned that alcohol addition decreases K<sub>L</sub>a amounts. They discussed that alcohol is adsorbed at air-water

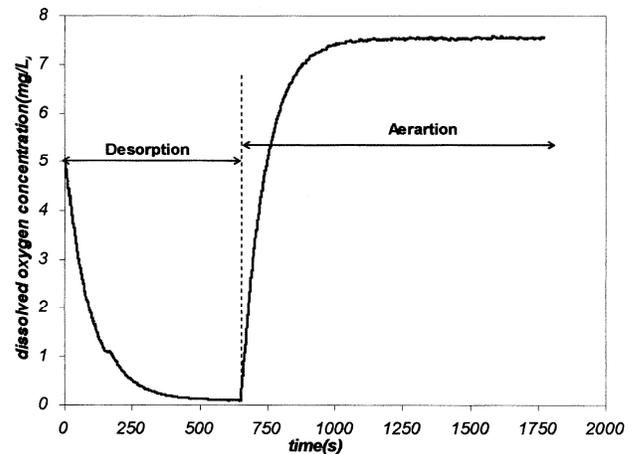


Fig. 2–Dissolved oxygen concentration versus time during deaeration and aeration

Table 1– Physical properties of system at 25°C

System	Density, ρ <sub>l</sub> , kg.m <sup>-3</sup>	Surface tension×10 <sup>3</sup> , σ <sub>l</sub> Pa
Tap water	998.2	72.75
Butanol 0.25%	997.8	68.323
Butanol 0.50%	997.4	60.077
Butanol 0.75%	997.0	54.768
Butanol 1.00%	996.6	50.221

interface of bubbles and a rigid layer forms around them which decreases  $K_L$  amounts.

By applying the regression analysis, a simple correlation for  $K_L a$  as a function of butanol concentration and input gas velocity is investigated as Eq. (3):

$$K_L a = 0.01603 \times U_G^{0.871603} \left[ 1 + C_A^{0.848354} \right]^{0.323719} \quad \dots (3)$$

For data fitting, LABFIT software (V 7.2.47) was applied. As shown in Fig. 4, experimental data were in good agreement with the correlated data (with  $R^2=0.98441$ ).

#### Bubble diameter

The steady state bubble diameter size was determined with photographic technique by a digital

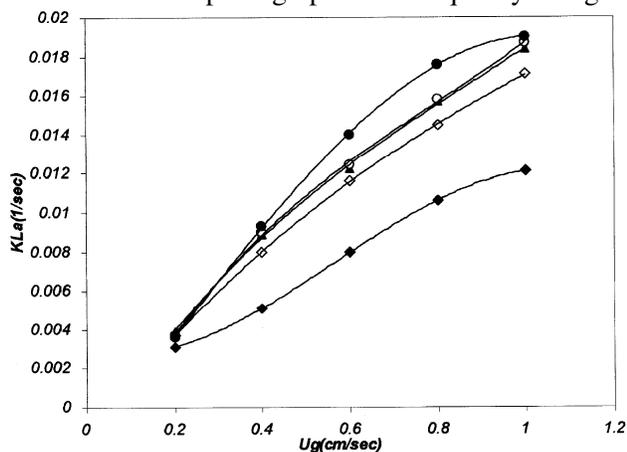


Fig. 3– Overall volumetric oxygen mass transfer coefficient,  $K_L a$ , versus superficial gas velocity,  $U_g$ , in riser (◆—pure water, ◇—0.25% butanol, ▲—0.5% butanol, ○—0.75% butanol, ●—1% butanol)

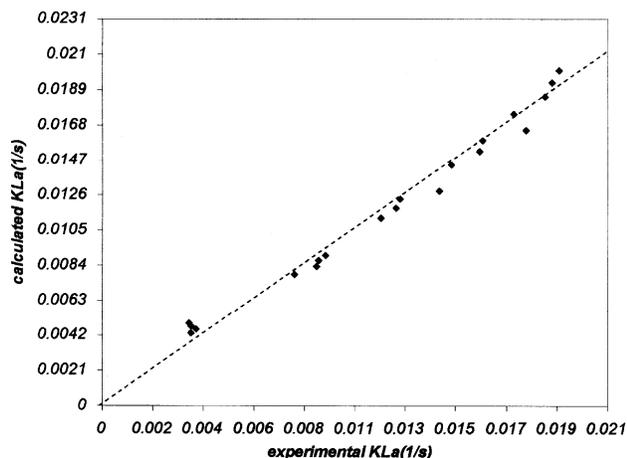


Fig. 4– Correlated data for  $K_L a$  versus the experimental data

camera (CANON S51S, with resolution of 8 M pixels). It was placed at 0.6 m above the bottom of ALR. The camera was used for riser and down-comer. The moving average method was used to account number of bubbles which were more than 300 bubbles that were chosen randomly in ten pictures that were captured at the middle of reactor (0.6 m above the bottom). Then, bubble diameter was measured by WINDIG software (version 2.5). In spherical bubbles  $d_1$  is equal to  $d_2$  (so  $d_v=d_1=d_2$ ) while elliptical bubbles were measured according to the maximum and minimum diameters of bubbles as following:

$$d_v = \sqrt[3]{d_1^2 d_2} \quad \dots (4)$$

where,  $d_v$ ,  $d_1$  and  $d_2$  in Eq. (4) introduce equivalent, maximum and minimum diameters, respectively.

Average of bubbles diameter is calculated:

$$d_{ave} = \frac{\sum_{i=1}^N d_i^3}{\sum_{i=1}^N d_i^2} \quad \dots (5)$$

$d_i$  is bubble diameter and  $i$  is number of bubbles (from 1 to  $N$ ).

Figure 5 shows bubbles diameter average versus number of bubbles using moving average method<sup>33</sup>.

Effect of superficial gas velocity inlet on bubbles diameter for both riser and down-comer are shown in Figs 6 and 7. As shown in these figures, bubble diameters curve in the riser is flatter than it in the down-comer. Its reason is due to exploding bubbles and also coalescence rate enhancement. Therefore, bubbles with various diameters are produced. The bubbles with size of less than 0.8 mm can be imported the down-comer and the bigger ones leave the reactor.

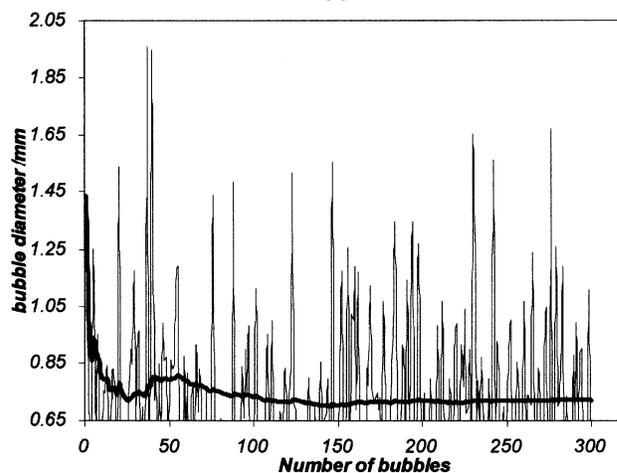


Fig. 5– Bubbles diameter average versus number of bubbles

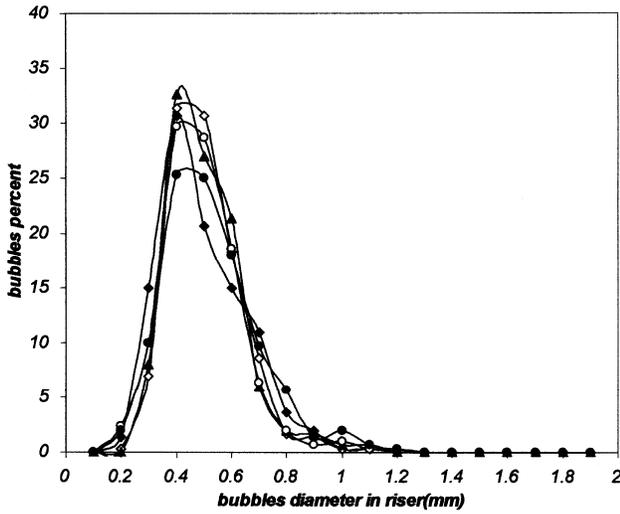


Fig. 6– Percentages of bubbles with the same diameter in riser with different  $U_g$  at butanol concentration of 1% (◆– $U_g$  0.2 cm/s, ◇– $U_g$  0.4 cm/s, ▲– $U_g$  0.6 cm/s, ○– $U_g$  0.8 cm/s, ●– $U_g$  1 cm/s)

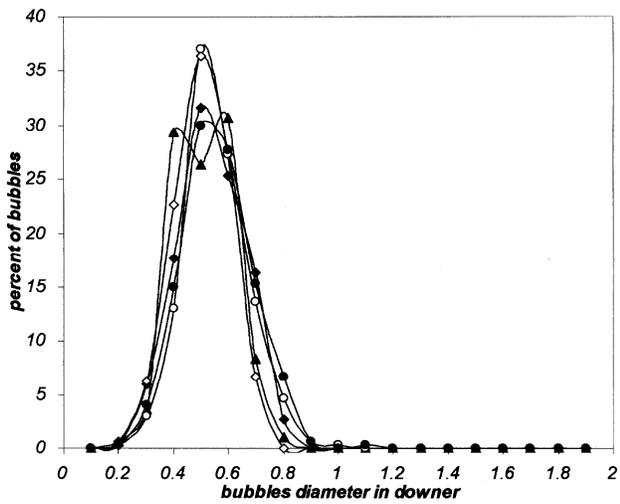


Fig. 7– Percentages of bubbles with the same diameter in riser with different  $U_g$  at butanol concentration of 1% (◆– $U_g$  0.2 cm/s, ◇– $U_g$  0.4 cm/s, ▲– $U_g$  0.6 cm/s, ○– $U_g$  0.8 cm/s, ●– $U_g$  1 cm/s)

The buoyancy force which is greater than the other forces causes this effect.

Figures 8 and 9 shows the bubbles diameter in pure water and butanol solutions with different concentrations. According to these figures, bubbles diameter decreased with butanol addition to the pure water and its concentration increment. Further, in pure water, bubbles diameter curve was too flat. For pure water in the riser, around 8% of bubbles had the same size while in butanol solutions, around 30% of bubbles had the same size. For pure water in the down-comer, around 13% of bubbles had the same size while in

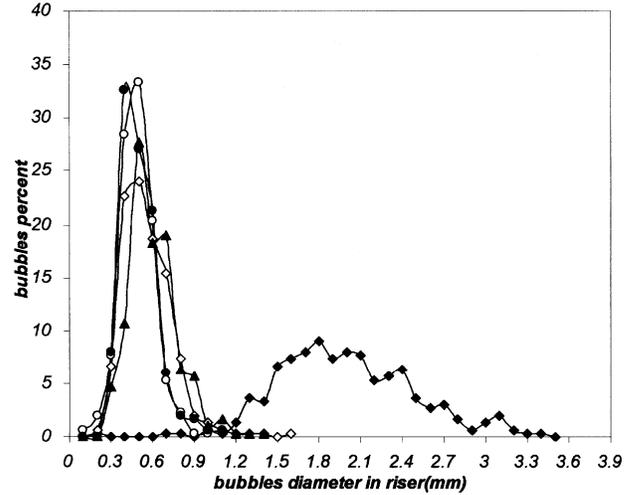


Fig. 8– Percentages of bubbles with the same diameter in riser with different butanol concentration at  $U_g$  of 0.6 cm/s (◆–pure water, ◇–0.25% butanol, ▲–0.5% butanol, ○–0.75% butanol, ●–1% butanol)

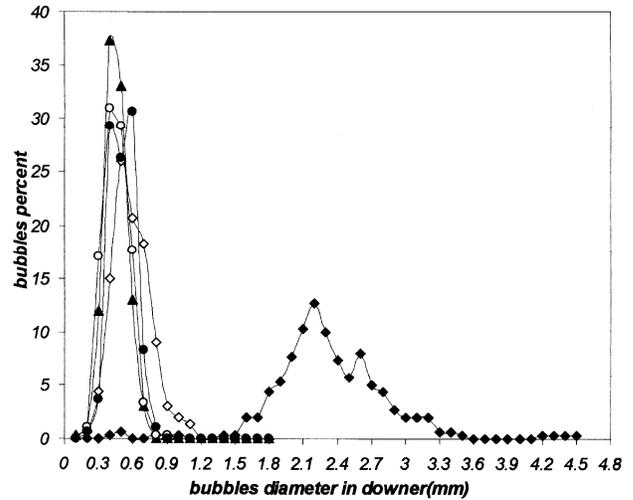


Fig. 9– Percentages of bubbles with the same diameter in down-comer with different butanol concentration at  $U_g$  of 0.6 cm/s (◆–pure water, ◇–0.25% butanol, ▲–0.5% butanol, ○–0.75% butanol, ●–1% butanol)

butanol solutions; around 36% of bubbles had the same size.

Since in the riser, bubbles with the same size are more, so the curve seems to be sharper while a regular trend is not observed in the down-comer. Albijanic *et al.*<sup>28</sup> and El Azher *et al.*<sup>32</sup> in their studies on airlift reactor have observed reduction of bubbles diameter with dilute alcohol solutions.

The average of bubbles diameter in different butanol concentrations and different  $u_g$  in riser region are given in Table 2. Increase in  $U_g$  by increasing of coalescence and break-up rate leads to increase in

Table 2– The average of bubbles diameter in riser

Gas velocity	0.2 (cm/s)	0.4 (cm/s)	0.6 (cm/s)	0.8 (cm/s)	1 (cm/s)
Butanol concentration					
0% (pure water)	1.95094	2.31036	2.25435	3.16003	3.16578
0.25% (v/v)	0.50832	0.74984	0.70004	0.67057	0.71606
0.50%(v/v)	0.68755	0.71048	0.71359	0.70994	0.78777
0.75%(v/v)	0.67754	0.56581	0.55703	0.60445	0.71490
1.00%(v/v)	0.59057	0.57359	0.58841	0.59014	0.63862

Table 3– The average of bubbles diameter in downcomer

Gas velocity	0.2 (cm/s)	0.4 (cm/s)	0.6 (cm/s)	0.8 (cm/s)	1 (cm/s)
Butanol concentration					
0% (pure water)	1.10617	1.71204	2.54939	2.76523	3.63954
0.25%(v/v)	0.55975	0.58463	0.68161	0.62889	0.61183
0.50%(v/v)	0.48656	0.56631	0.49842	0.54235	0.59355
0.75%(v/v)	0.50403	0.56193	0.51440	0.52165	0.65436
1.00%(v/v)	0.58351	0.54037	0.55175	0.60201	0.61769

bubbles diameter, but coalescence rate in pure water is more than butanol solutions (than break-up). Bubbles moving in liquid phase lead to absorbing butanol molecules in their surfaces that change liquid-gas surface properties and change bubble surface too soft, flexible and sensitive to liquid pressure fluctuation and eddies. So, the minimum energy required for bubble break-up decreases and leads to increasing of bubble break-up. Although these molecules create a rigid layer which reduces the bubbles coalescence rate too, this mechanism has been explained elsewhere<sup>34-37</sup>. Also this rigid layer by reducing the surface renewal rate leads to decreasing of  $K_L$  but active surface ( $a$ ) increases by decreasing of bubbles diameter that is more effective and leads to increasing of volumetric mass transfer coefficient (Fig. 3). Also by increasing of butanol concentration which leads to decreasing of surface tension (Table 1) and bubbles diameter decrease overall.

Buoyancy force and drag force are two effective forces which act opposite of each other and compete too. Reducing of bubbles diameter because of increasing butanol concentration leads to decreasing of buoyancy force. Thus, these smaller bubbles can import to down-comer and some of them cover a distance in the down-comer. In the other hand they are usually similar and symmetrical and small, as we can be seen in Table 3. With increasing of  $U_g$ , liquid circulation velocity increase too<sup>3</sup>, this matter reduce the liquid and bubble delay time in separator region, so help bubble to import to down-comer, in this

situation some bigger bubbles import to down-comer lead to increasing average of bubble diameter in higher aeration velocity.

### Conclusions

The effects of butanol addition and its concentration increment on bubbles diameter and mass transfer coefficient in a split-cylindrical airlift bioreactor was studied. According to this study,  $K_L a$  increased with adding butanol to pure water and increasing its concentration when superficial gas velocity increased. Further, bubbles diameter decreased with increasing butanol to the pure water because surface tension in butanol solutions was less than it in pure water. For pure water in the riser around 8% of bubbles had the same size while for butanol solutions around 30% of bubbles had the same size. An accurate correlation was shown for volumetric mass transfer coefficient as a function of butanol concentration and superficial gas velocity.

### Nomenclature

- $C_L$  = concentration of dissolved oxygen at any time  $t$  ( $\text{kg}/\text{m}^3$ )
- $C_0$  = initial concentration of dissolved oxygen ( $\text{kg}/\text{m}^3$ )
- $C^*$  = saturation concentration of dissolved oxygen ( $\text{kg}/\text{m}^3$ )
- $K_L$  = liquid film mass transfer coefficient ( $\text{m}/\text{s}$ )
- $K_L a$  = overall volumetric gas-liquid mass transfer coefficient ( $\text{s}^{-1}$ )
- $DO$  = dissolved oxygen
- $d_1$  = maximum bubble diameter (mm)
- $d_2$  = minimum bubble diameter (mm)
- $d_v$  = equivalent of bubble diameter (mm)

$d_{ave}$  = average diameter of bubbles (mm)

$N$  = number of bubbles (-)

#### Greek symbols

$\rho_G$  = density of air (kg/m<sup>3</sup>)

$\rho_L$  = density of liquid (dilute alcohol solution) (kg/m<sup>3</sup>)

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