

## Investigation of Surfactant Effect on the Operational Characteristics of a Packed Bed Internal Loop Airlift Reactor

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**Abstract:** Investigation of turbulency on the rate of induced liquid circulation, gas hold-up, mixing time and overall gas-liquid volumetric oxygen mass transfer coefficient. The various types of surfactants (containing Brij58, TritonX-405, Tween40, HCTBr) with various concentrations of 1-5 ppm were examined on the operational characteristics of the reactor. In surfactant solutions (in comparison with pure water), surface tension of the liquid bulk decreased and smaller bubbles were produced. Therefore, surfactants existence increases gas hold-up and mixing time although it decreased the liquid circulation velocity and the rate of oxygen mass transfer. HCTBr which is a cationic surfactant was the most effective surfactant. Packing installation increased mass transfer by increasing flow turbulency and Reynolds number. Further, gas hold-up increased and liquid velocity decreased when gas bubbles movement increased. In the packed bed system, homogenous flow regime was highly observed while in the unpacked bed system the transition flow regime overcame at the high superficial gas velocities.

**Key words:** Airlift reactor • Gas hold-up • Liquid circulation • Mixing time • Gas-liquid mass transfer • Surfactants

### INTRODUCTION

Airlift reactors are one of the most important of two phase contactors which are increasingly used in chemical industries, biological processes, aerobic fermentation, wastewater treatment and other gas-liquid contacting applications [1, 2]. Approximately 25% of all chemical processes occur between a gas phase and a liquid phase [3]. There are some advantages such as suitable heat and mass transfer and shorter reaction time because of closer contact between the phases, low shear rate, high mixing performance, high and flexible capacity. For optimizing such reactor operations many different geometric configurations of airlift reactors were described [4, 5].

Surfactants are the materials consist of molecules containing both polar and non-polar parts (amphiphilic). These molecules locate their hydrophilic head groups in the aqueous phase and allow the hydrophobic hydrocarbon chains to escape from water phase [6]. These materials are widely using as antifoam agent,

wetting agent, detergent, film coating, emulsifying agent, chemical and petrochemical productions [7]. These materials exist in many factories wastewater and are one of the most important of water pollutant. As there is a balance between hydrophobic and hydrophilic parts of the surfactant molecule, these systems have special properties such as accumulation at various interfaces and association in solution, changing in physical chemistry properties of water, sludge growth and aquatics dead rate enhancement. More than 2 million Liters of such wastewater per year import the Anzali pond (where is a rare ecosystem in Iran) and they decrease water depth from 10 to less than 2 [8].

Kalekar *et al.* investigated the adsorption of various surfactants at gas-liquid interface [9].

Kothekar *et al.* studied the foamability, foam stability, emulsifying power, surface tension and interfacial tension of Tween 20, Tween 60, Tween 80, Arlacel 60 and arlacel 80. They found the Arlacel 60 has the best emulsifying power and foamability [10].

Several researchers investigated the critical micelles concentration (CMC) for different surfactants in different solutions [11].

Bubbles diameter reduction and oxygen concentration in some systems containing different surfactants were reported in the literature [12].

Nikakhtari *et al.* increased the volumetric mass transfer coefficient by inserting a small quantity of nylon mesh packing in the riser section of an external loop airlift bioreactor (ELAB) [13]. They also used stainless steel meshes as packings (with 99.0 % porosity) in the riser of the same reactor to increase the volumetric mass transfer coefficient [5].

Chisti *et al.* used two separate blocks of static mixer elements in the riser of an ELAB for the oxygen mass transfer enhancement [14].

Vychodilova *et al.* used glass spheres (with the voidage of 0.4 and diameter of 0.01 m) as packings [15]. They reached the volumetric mass transfer coefficient of  $0.05 \text{ s}^{-1}$ .

In this research, oxygen mass transfer coefficients in a packed bed internal loop airlift reactor were investigated when different surfactants (containing Brij58, TritonX-405, Tween40, HCTBr) at various concentrations were added into the water. The effects of these surfactants on the bubbles diameter, gas hold-up, liquid velocity, mixing time and flow regime were studied.

## MATERIALS AND METHOD

**Experiment:** HCTBr (Ammonium hexadecyltrimethyl bromide), Tween 40 (Polysorbate 40), Triton X-405 (Polyethylene glycol octylphenyl ether) and Brij58 (Polyoxyethylene (20) cetyl ether) were purchased from Merck Company (Germany) and their various solutions with various concentrations (1-5 ppm) were locally prepared. The properties of these surfactants are shown in detail in Table 1.

**Apparatus Set Up:** The split-cylinder airlift reactor and the flow scheme used are shown schematically in Figure 1.

The reactor consisted of a glass column (with 1.3 m height and 13.6 cm diameter). A rectangular Plexiglas baffle (with 0.129 m width, 1.0 m height and 0.005 m thickness) was inserted in the glass column to divide the cross section into a riser zone and a down-comer zone (the riser area:  $86.115 \text{ cm}^2$  and the down-comer area:  $40.299 \text{ cm}^2$ ). The baffle was located at a distance of 0.1 m from the bottom of the reactor. The riser zone was packed with 25 Polyethylene cylinders (0.115 m in length, 0.04 m in diameter) as shown in Figure 1. The gas-free liquid height in the reactor for each experiment was about 1.23 m. The gas sparger located at the bottom of the riser with 0.02 m in diameter made of the sintered ceramic ball.

A dissolved oxygen electrode was positioned in the riser zone at depth of 0.1 m from the surface of the gas-free liquid (Figure 1). The probe's tip was at an angle of  $30^\circ$  to the horizontal for preventing oxygen bubbles sticking to it. The conductivity electrode was positioned in the down-comer zone at depth of 0.2 m from bottom of the reactor. All experiments were carried out at ambient conditions (atmospheric pressure and  $25(\pm 0.5)^\circ\text{C}$ ).

**Measurement Methods:** Inverted U-tube manometers were used to measure the gas hold-up [1, 2] in the riser and the down-comer zones (Figure 1). The manometer taps were vertically positioned at 0.93 m apart vertically. The liquid circulation velocity was determined from the tracer response curve [1]. For this purpose,  $25 \text{ cm}^3$  of a tracer (3 M aqueous NaCl) was instantaneously injected on the surface of the fluid at the top of the down-comer zone.

Mixing time was estimated from the time interval between the first peak of the tracer signal and the point on  $x$ -axis where the conductivity had 95% of its final stable value.

The overall volumetric gas-liquid mass transfer coefficient was measured by the dynamic gassing-in method [1, 2]. Dissolved oxygen concentration followed the typical pattern during the deaeration-aeration sequence. The overall volumetric oxygen mass transfer coefficient ( $k_L a$ ) was calculated using the following equation which is valid for large response time [16]:

Table 1: Properties of surfactants

$\sigma(\text{mN.m}^{-1})$	CMC( $\text{mM.L}^{-1}$ )	HLB	$M_w(\text{kg.kmol}^{-1})$	Type	Formula	Surfactant
36.955	0.955	-	364.5	Cationic	$\text{C}_{19}\text{H}_{42}\text{BrN}$	HCTBr
37.373	0.08	15.7	1123.5	Nonionic	$\text{C}_{16}\text{H}_{33}(\text{OCH}_2\text{CH}_2)_{20}\text{OH}$	Brij 58
37	0.5	17.6	1968.5	Nonionic	$\text{C}_{94}\text{H}_{182}\text{O}_{41}$	TritonX-405
38.83	0.027	15.5	1283.8	Nonionic	$\text{C}_{62}\text{H}_{122}\text{O}_{26}$	Tween 40

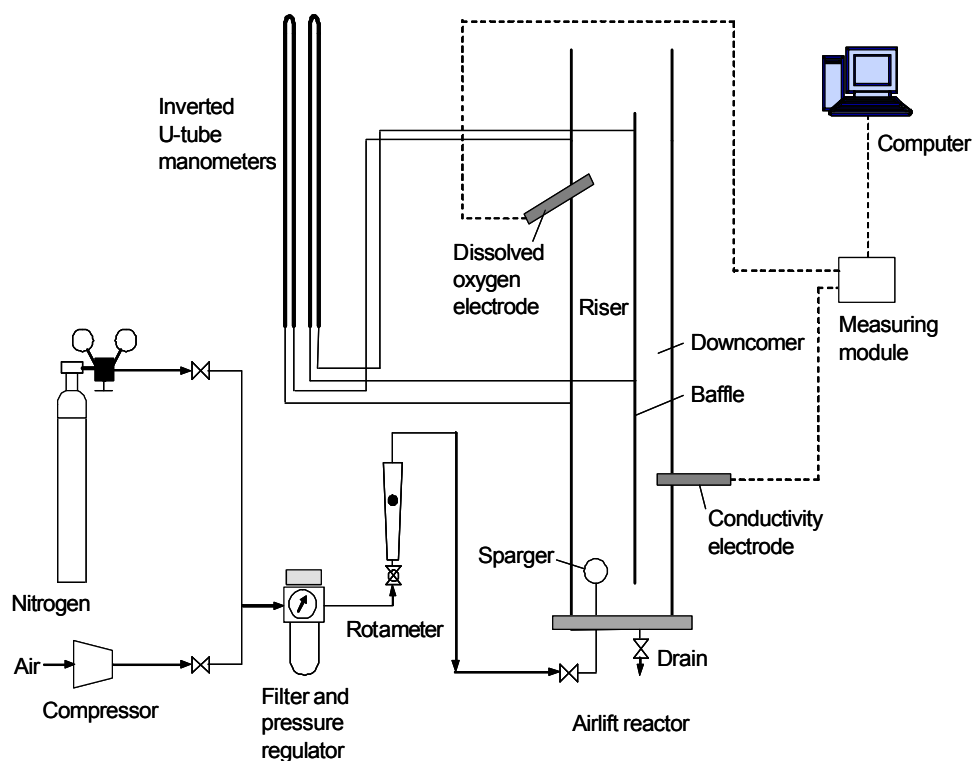


Fig. 1: Schematic diagram of the split-cylinder airlift reactor

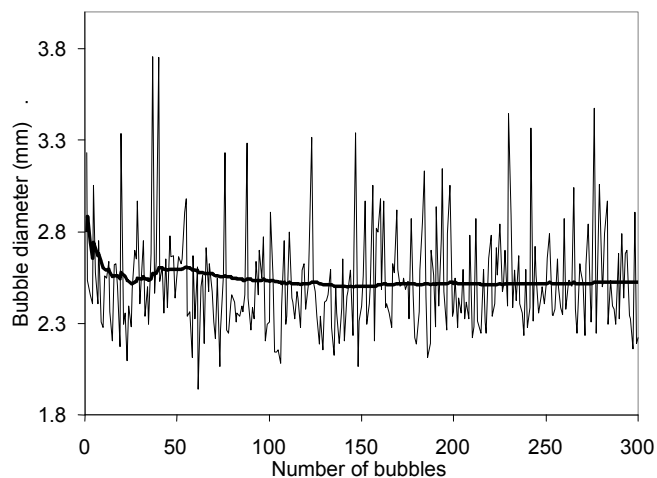


Fig. 2: Bubbles diameter average versus number of bubbles

$$\frac{C^* - C_L}{C^* - C_{L0}} = \left( \frac{e^{-tk_L a}}{t_E} - k_L a e^{-(t/t_E)} \right) \frac{t_E}{1 - t_E k_L a} \quad (1)$$

Where,  $t_E$  is the electrode response time,  $C^*$  is the saturation concentration of oxygen in water at the operating temperature and  $C_L$  is the instantaneous concentration of dissolved oxygen at time  $t$ .

The steady state bubbles diameter size was determined with photographic technique by a digital camera (CANON, model: S51S with resolution of 8 M pixels). The Moving Average method [17] was used to account number of bubbles which were more than 300 bubbles as shown in Figure 2. They were randomly chosen in ten pictures which captured at the middle of reactor (0.6 m above the bottom). Average of bubbles diameter is calculated:

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

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

Zuber and Findlay [18] method was used to find the flow regimes. This method gives acceptable results in air-water and the system which the liquid phase density roughly is equal to the pure water density.

## RESULTS AND DISCUSSION

**Bubble Diameter and Flow Regime:** Effects of various surfactants concentrations at the superficial gas velocity of 0.6 cm/s on bubbles diameter are shown in Figure 3. Surfactant addition to pure water (with bubble diameter of 3.5 mm) decreased the bubbles diameter about 25 % (at the concentration of 5 ppm). Further, surfactant concentration enhancement decreased average of bubbles diameter by decreasing solution surface tension. Sardeing *et al.* used various surfactants [12]. They investigated that bubbles diameter decreased about 30 % (as an average value).

In this research, Triton X-405 as a nonionic surfactant with the highest HLB (Hydrophile-Lipophile Balance) value made the biggest bubbles. Since HLB values for Tween 40 and Brij 58 were the same so, bubbles diameter were isometric (Table 1). Literature supports this output, properly [19].

As shown in Figure 4, homogenous flow regime for pure water is observed in the aeration velocities less than 0.65cm/s. After it the transition flow regime is significantly observed in the unpacked reactor. In surfactant solutions, bubbles diameter decreased with decreasing buoyancy force. Therefore, bubbles import down-comer easily and transition flow regime is observed in all of the aeration velocities.

In packed bed reactor, packings perform like a baffle which wastes the kinetic energy of liquid bulk and bubbles. Therefore, some dead zones are created under the packings and some bubbles are captured inside them. They join together and create bigger bubbles. So, bubbles leave the reactor without importing down-comer. In this situation the homogenous flow regime is highly observed (in both pure water and surfactant solutions).

**Hydrodynamic:** The effects of packing installation and surfactants addition on gas hold-up in the riser and down-comer zones are shown in Figures 5 and 6, respectively.

In both zones, the surfactant addition increased the gas hold-up about 26% (at the highest aeration and surfactant concentration) compared with pure water. The gas hold-up effect in the surfactants solutions depended on the HLB value, surface tension and molecular weight. These factors decreased the bubbles diameter and gas hold-up. So in both riser and down-comer zones, the gas hold-up increased as following:

$$\text{Water} < \text{Tween 40} = \text{Triton X-405} = \text{Brij58} < \text{HCTBr}$$

The effect of packings installation in riser and down-comer is different. In the riser zone, they increased the bubbles residence time. So, gas hold-up increased while in the down-comer zone homogenous flow regime was highly observed and gas hold-up decreased.

The gas hold-up difference between the riser and down-comer created density difference between these zones which was a driving force for liquid movement and its circulation. Surfactant addition changed the homogenous flow regime to transition one and decrease the driving force. As shown in Figure 7, packings installation decreased the liquid bulk and gas bubbles kinetic energies. So, liquid circulation decreased by packings installation and surfactant addition. The circulation velocity reduced as following:

$$\text{Water} > \text{HCTBr} > \text{Brij 58} = \text{Tween 40} = \text{Triton X-405}$$

Any reason which reduces the velocity of circulation adversely increases the mixing time because in an airlift reactor the mixing time is primarily controlled by the liquid velocity [1]. So, surfactant addition and packings installation affect directly on the magnitude of the induced liquid circulation velocity and increase the mixing time. As shown in Figure 8, the mixing time increased as following for any specified aeration velocity:

$$\text{Water} < \text{HCTBr} < \text{Brij 58} < \text{Triton X-405} < \text{Tween 40}$$

**Mass Transfer:** The measurements of the overall volumetric mass transfer coefficient,  $k_L a$ , are shown in Figure 9. Surfactants existence always decreased the  $k_L a$  in comparison with pure water.

As shown in Figure 10,  $k_L a$  decreased by increasing the surfactant concentration. According to the surfactant type, Brij 58 decreased  $k_L a$  (around 44 %) at concentration of 5 ppm and at highest aeration rate in comparison with pure water. In fact, the effect of surfactants on  $k_L a$  was due to affecting active surface ( $a$ ) and mass transfer

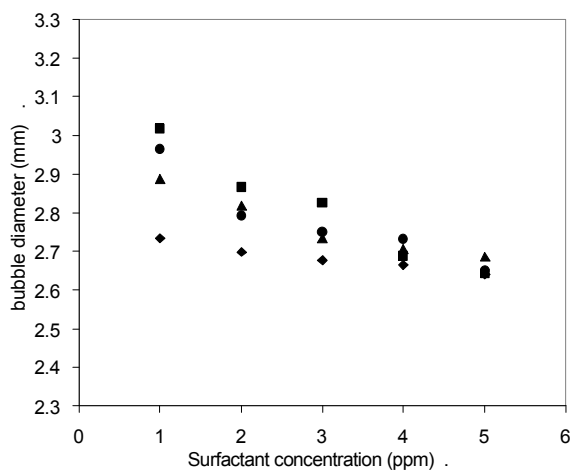


Fig. 3: Bubbles diameter versus surfactant concentration at  $U_g = 0.6$  cm/s  
 □: HCTBr, □: TritonX-450, □: Tween 40, □: Brij58

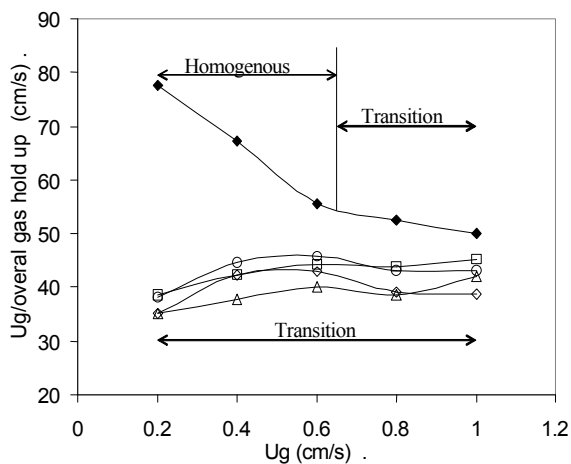


Fig. 4: Experimental characterization of flow regimes in unpacked reactor containing pure water and surfactant (5ppm).  
 ◊: pure water, □: HCTBr, ◊: TritonX-450, ◊: Tween40, ◊: Brij58,

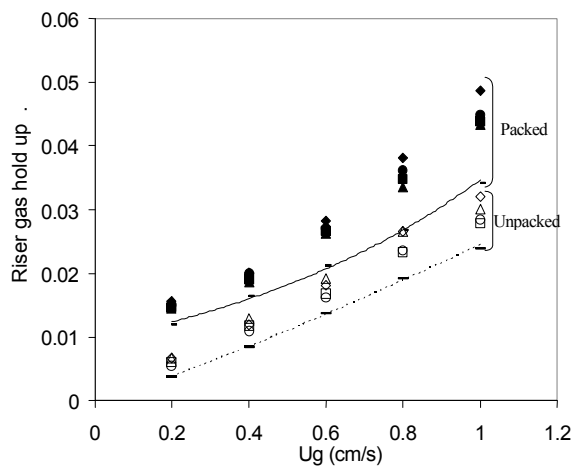


Fig. 5: Gas hold-up in riser versus superficial air velocity  $U_g$  in riser.

□: HCTBr, □: TritonX-450, □: Tween40, □: Brij58. —: water (packed)  
 □: HCTBr, □: TritonX-450, □: Tween40, □: Brij58. - - : water (unpacked)

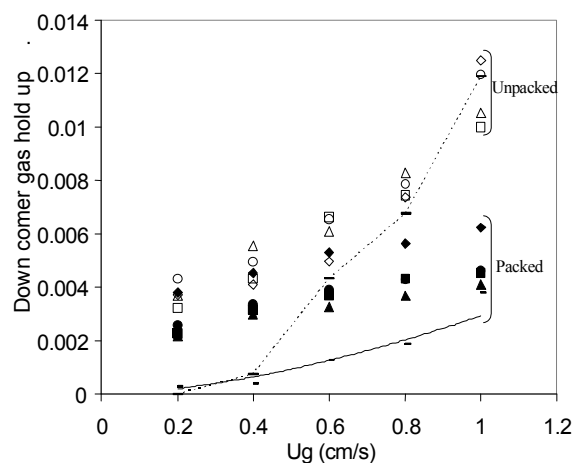


Fig. 6: Gas hold-up in down-comer versus superficial air velocity ( $U_g$ ) in riser  
 $\circ$ : HCTBr,  $\square$ : TritonX-405,  $\triangle$ : Tween40,  $\diamond$ : Brij58. —: water (packed)  
 $\circ$ : HCTBr,  $\square$ : TritonX-405,  $\triangle$ : Tween40,  $\diamond$ : Brij58. - - : water (unpacked)

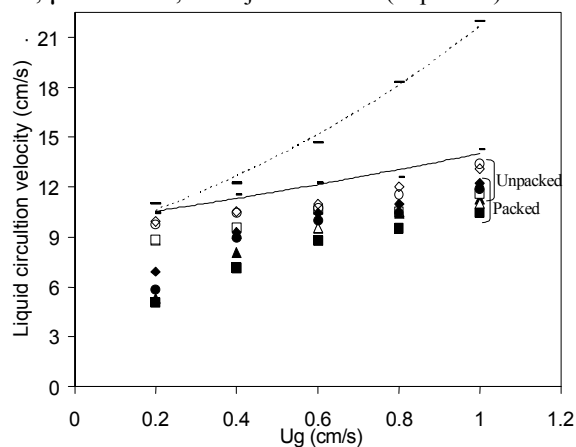


Fig. 7: Liquid circulation velocity versus aeration velocity ( $U_g$ )  
 $\circ$ : HCTBr,  $\square$ : TritonX-405,  $\triangle$ : Tween40,  $\diamond$ : Brij58. —: water (packed)  
 $\circ$ : HCTBr,  $\square$ : TritonX-405,  $\triangle$ : Tween40,  $\diamond$ : Brij58. - - : water (unpacked)

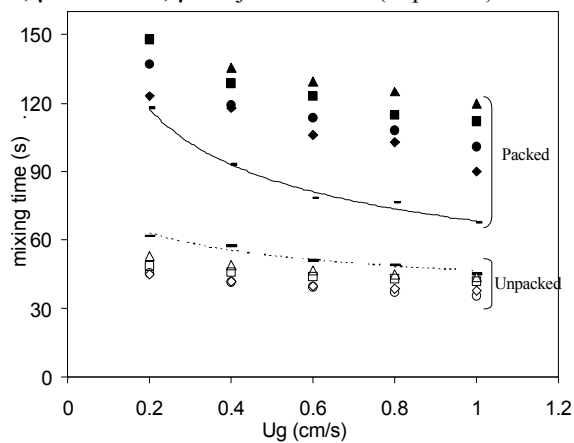


Fig. 8: Mixing time versus superficial air velocity ( $U_g$ ) in riser  
 $\circ$ : HCTBr,  $\square$ : TritonX-405,  $\triangle$ : Tween40,  $\diamond$ : Brij58. —: water (packed)  
 $\circ$ : HCTBr,  $\square$ : TritonX-405,  $\triangle$ : Tween40,  $\diamond$ : Brij58. - - : water (unpacked)

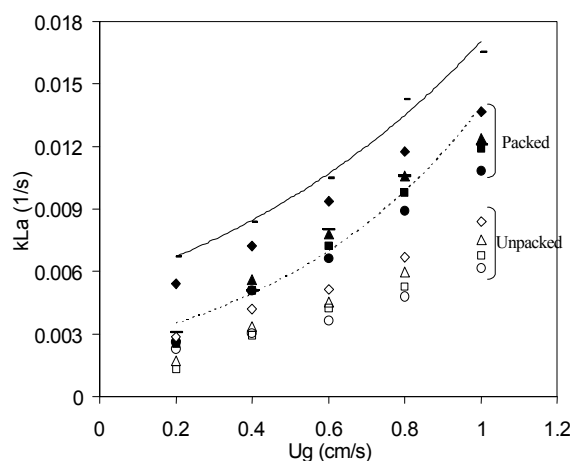


Fig. 9: Overall volumetric oxygen mass transfer coefficient ( $k_La$ ) versus superficial air velocity ( $U_g$ ) in riser.  
 $\diamond$ : HCTBr,  $\square$ : TritonX-405,  $\triangle$ : Tween40,  $\circ$ : Brij58. —: water (packed)  
 $\square$ : HCTBr,  $\diamond$ : TritonX-405,  $\triangle$ : Tween40,  $\circ$ : Brij58. - - : water (unpacked)

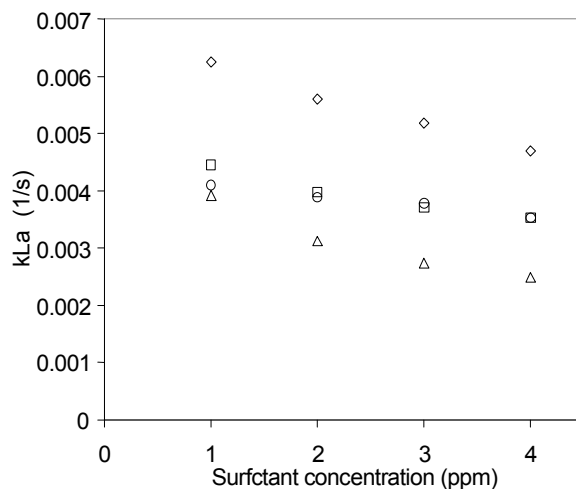


Fig. 10: Overall volumetric oxygen mass transfer coefficient ( $k_La$ ) versus surfactant concentration at  $U_g=0.6$  cm/s.  
 $\square$ : HCTBr,  $\diamond$ : TritonX-405,  $\triangle$ : Tween40,  $\circ$ : Brij58.

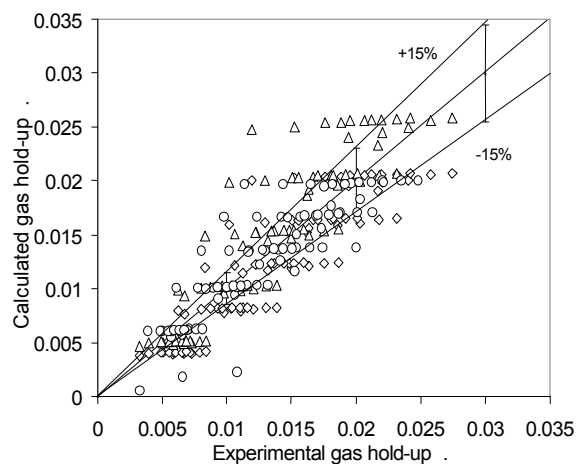


Fig. 11: The predicted gas hold up data versus experimental gas hold-up  
 $\square$ : Akita and Yoshida (1974),  $\triangle$ : Sada *et al* (1984),  $\circ$ : Mouza *et al* (2005).

Table 2: Reynolds number in pure water

Ug (cm/s)	1	0.8	0.6	0.4	0.2
Re number (unpacked)	803	777	687	627	566
Re number (packed)	1015	975	838	810	687

Table 3: Gas hold-up correlation based on *Bo* number, *Ga* number and *Fr* number

Author	Remarks	Correlations
Akita <i>et al</i> (1974)	$d_b > 2.5\text{mm}$	$\frac{\varepsilon_g}{(1-\varepsilon_g)^4} = 0.2Bo^{1/8}Ga^{1/12}Fr$
Sada <i>et al</i> (1984)	$0.3 < u < 30\text{cm/s}$	$\frac{\varepsilon_g}{(1-\varepsilon_g)^4} = 0.32Bo^{0.21}Ga^{0.086}Fr$
Mouza <i>et al</i> (2005)	—	$\varepsilon_g = 0.001 \left( Bo^{2.2} Ga^{0.1} Fr \frac{d_{sp}}{d_b} \right)^{2/3}$

coefficient ( $k_L$ ). Surfactants increase the active surfaces by bubbles diameter reduction. It is clear that surfactants have a strong negative effect on the true mass transfer coefficient ( $k_L$ ) and this is more effective than the interfacial area enhancement effect. Surfactants coat gas-liquid interfaces and create a rigid layer which it can impede the mass transfer by various mechanisms such as interfacial turbulence reduction [20, 21] and slowed diffusion [22, 23].

**Therefore, the  $K_L a$  Decrease for Any Aeration Velocity, as Following:**

Water > HCTBr > Tween 40 > Triton X-405 > Brij 58

As shown in Table 2, in a packed bed ALR, Reynolds number which is a turbulence symbol increased from 803 to 1015 in the riser zone (at  $U_g=1$  cm/s). Further, gas hold-up increased and liquid velocity decreases. The residence time (delay time) which contributes to the mass transfer increased at lower velocities. These factors assist to provide higher mass transfer rate in a packed bed ALR (more than 43 %) compared to an unpacked one. Nikakhtari *et al.* obtained  $k_L a$  at range of 0.007-0.016 in an airlift reactor equipped with stainless steel wire meshes [5]. Chisti *et al.* obtained  $k_L a$  of  $0.016 \text{ s}^{-1}$  (as an average value) in a concentric draft-tube bioreactor which was agitated by two identical down ward hydrofoil impellers (with 260 rpm) [14].

**Correlation:** Empirical correlation based on dimensionless groups is one of the best methods for reactors scale up. The effects of surfactants on interfacial properties of operational phase such as surface active, mass diffusion coefficient and body forces and also liquid bulk properties such as surface tension, density and viscosity were carefully considered.

*Ga* number (the ratio of gravity forces to viscous forces), *Fr* number (the ratio of inertia forces to the gravity forces) and *Bo* number [the ratio of body forces (which often is equal to the gravity forces)] were used for gas hold-up correlation [24-26]. The used equations are illustrated in Table 3. Figure 11 shows the predicted data obtained from the equations versus experimental data. Mouza *et al.*'s equation [26] showed the best agreement with the experimental data. In surfactant solutions, surface tension which is an effective parameter decreases in comparison with pure water. The surface tension has the most effect on *Bo* number. So, *Bo* number effect on Mouza *et al.*'s equation is more than the other equations.

For mass transfer correlation purpose, *Re* number (the ratio of inertial forces to viscous forces), *Sc* number [the ratio of momentum diffusivity (viscosity) and mass diffusivity], *Sh* number (the ratio of convective to diffusive mass transport) and *Bo* number were used by Akita and Yoshida [24], Asgharpour *et al.* [27] and Bird *et al.* [28]. The used equations are illustrated in Table 4.

Table 4: Sherwood number correlations for liquid-gas mass transfer coefficient based on  $Re$  number,  $Sc$  number and  $Bo$  number

Author	Remarks	Correlations
Asgharpour <i>et al</i> (2010)	$0.118 < u < 2.35 \text{ cm/s}$	$Sh = 0.15 Re^{2/3} Sc^{1/2} Bo^{2/3}$
Bird <i>et al</i> (2002)	Higbie's model	$Sh = (4/\pi)^{1/2} Re^{1/2} Sc^{1/2}$
Akita <i>et al</i> (1974)	Homogeneous flow	$Sh = 0.6 Re^{1/2} Sc^{1/2} Bo^{3/8}$

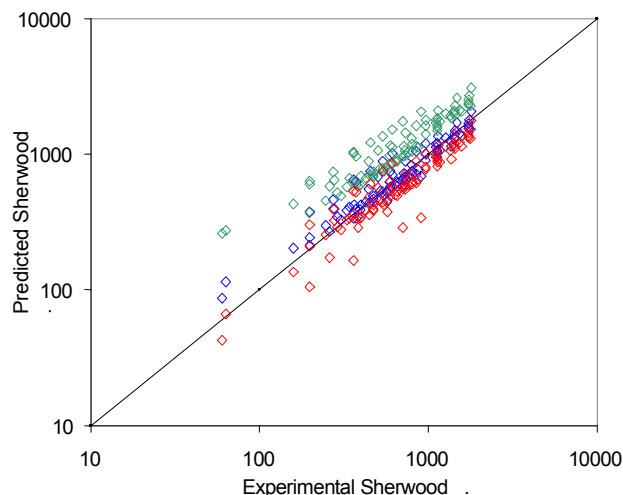


Fig. 12: The predicted Sherwood number versus the experimental Sherwood numbers.

□: Akita and Yoshida (1974), ○: Mehra *et al* (2010), △: Bird *et al* (2002).

Figure 12 shows the predicted data versus experimental data. Akita and Yoshida's equation [24] and Asgharpour *et al.*'s equation showed better agreement with experimental data than Bird's equation which highly depends on the  $Bo$  number.

## CONCLUSIONS

Effects of some surfactants (containing HCTBr, TritonX-405, Tween 40 and Brij 58) and their concentrations (from 1 to 5 ppm) on bubbles diameter, hydrodynamic and mass transfer in a split-cylindrical airlift reactor were studied.

HCTBr which is a cationic surfactant with the minimum molecular weight was the most effect on bubbles diameter reduction. Surfactants addition increased gas hold-up and mixing time and decreased the liquid circulation by changing the flow regime from the homogenous to transition. Furthermore, they decreased the volumetric mass transfer coefficient about 44 %. Turbulency affected on the liquid bulk. So, packings existence increased the mass transfer. They could cover surfactant effect on mass transfer reduction. The packings performed like baffle. They increased the gas hold-up and decreased the liquid circulation.

## Nomenclature

$Bo$ :	[kg/m <sup>3</sup> ]	Bond number [28]
$C_L$ :	[kg/m <sup>3</sup> ]	concentration of dissolved oxygen at any time $t$
$C_0$ :	[kg/m <sup>3</sup> ]	initial concentration of dissolved oxygen
$C^*$ :	[kg/m <sup>3</sup> ]	saturation concentration of dissolved oxygen
$d_{ave}$ :	[mm]	average diameter of bubbles
$Fr$ :		Froude number [28]
$Ga$ :		Galilei number [28]
$k_L a$ :	[s <sup>-1</sup> ]	overall volumetric gas-liquid mass transfer coefficient
$N$ :		Number of bubbles
$Re$ :		Reynolds number [28]
$Sh$ :		Sherwood number [28]
$Sc$ :		Schmit number [28]
$U_G$ :	[m/s]	superficial aeration velocity in the riser zone

## Greek Symbols

$\varepsilon$ :		Gas holdup
$\mu$ :	[k/Pa.s]	Viscosity of phase
$\rho$ :	[kg/m <sup>3</sup> ]	Density of phase
$\sigma$ :	[mN/m]	Surface tension

## Subscript

k Phase, k= G: gas phase, k=L: liquid phase

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