

HYDRODYNAMICS AND OXYGEN MASS TRANSFER CHARACTERISTICS OF PETROLEUM BASED MICRO-EMULSIONS IN A PACKED BED SPLIT-CYLINDER AIRLIFT REACTOR

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Abstract - The effects of aeration velocity and liquid properties on the pertinent hydrodynamic and mass transfer parameters in a split-cylinder airlift reactor (with and without packing) were examined. Four different oil-in-water micro-emulsion systems containing kerosene, heavy naphtha, light naphtha and diesel as the oil at the concentration of 7% (v/v) were used in the experiments and the results were compared with pure water. The experimental results showed that the gas (air) hold-up and the volumetric gas-liquid oxygen transfer coefficient values for the micro-emulsion systems were usually greater than those of pure water. The packing installation increased the overall gas-liquid volumetric mass transfer coefficient by increasing the flow turbulence and Reynolds number, compared to the unpacked column. The packing increased the gas hold-up and decreased the bubble size and liquid circulation velocity. Furthermore, two empirical correlations were developed to predict the overall gas hold-up and volumetric oxygen transfer coefficient. A good agreement was observed between the experimental and correlated data.

Keywords: Airlift reactor; Gas hold-up; Micro-emulsion; Volumetric mass transfer coefficient.

INTRODUCTION

Airlift reactors are a modified type of bubble column in which the internal structure is divided into two separate sections by a baffle split or draft tube, which are the riser and down-comer (Carvalho, 2000 *et al.*, 2001; Hekmat *et al.*, 2010).

The gas hold-up (ϵ) difference between the riser and down-comer of an airlift reactor creates a density difference between these two zones as a driving force for liquid circulation (Moraveji *et al.*, 2012a).

Petroleum refineries generate large amounts of wastewater with high concentrations of hazardous contaminants. The effluents of these refineries usually contain 1-10% oil, the bulk consisting of water, emulsifiers and other ingredients (Jonsson and

Tragardh, 1990). Many industries such as petrochemical, petroleum production, oil refinery factories, metal, food industries and cosmetics usually produce a wastewater containing emulsions of oil in water (O/W) (Kong and Li, 1999; Lin and Lan, 1998). Because of the toxic nature and important effects of different kinds of oily wastewater on the surrounding environment (soil, water), it is necessary to treat the wastewaters before discharging to the environment.

There are several methods such as dissolved air flotation (DAF) (Al-Shamrani *et al.*, 2002; Bensadok *et al.*, 2007), adsorption (Ayotamuno *et al.*, 2006), biological treatment (Tellez *et al.*, 2002; Zhao *et al.*, 2006), and sedimentation in a centrifugal field (Cambiella *et al.*, 2006) and in hydro-cyclones (Hashmi *et al.*, 2004) that can be used for the treatment of

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wastewater. Since the upper limit of permissible oil in the effluent is steadily reduced by governing bodies, industries have to improve the treatment plants to meet the new limits.

The oil-in-water emulsion is a colloidal dispersion of oil droplets in an aqueous medium. These droplets have a tendency to coalesce and separate from the aqueous phase.

One of the useful equipments that can treat wastewaters is the airlift reactor. Most of the published works on airlift reactors have been on air-water based systems with properties different from the real conditions of the aerobic bio-desulfurization (BDS) processes. There are some reports on BDS in airlift reactors (Monticello, 2000; Lange and Pacheco, 1999) which mostly are water-in-oil (W/O) micro-emulsions.

Mehrnia *et al.* (2004a & 2004b) used a water-in-kerosene (W/K) micro-emulsion at a water-to-oil phase volume ratio (ϕ) of 20%, as a cold model of the BDS inside a draft-tube airlift bioreactor (DTAB) with different geometries. They also correlated their results using viscosity and aeration velocity (U_G), although their variables were reported for the homogenous flow regime.

In another research, the viscosity effect on the overall volumetric oxygen transfer coefficient ($k_L a$) using water-in-diesel (W/D) micro-emulsions with a water to diesel volume ratio (ϕ) in the range 2.5–40% inside the same reactor was investigated (Shariati *et al.*, 2007). They found that an increase of the viscosity of the micro-emulsion resulted in a further reduction of the values of $k_L a$ at all air rates for which measurements were done.

However, several techniques were applied to enhance the oxygen transfer rates in columns at low aeration rates (Fraser and Hill, 1993; Godo *et al.* 1999; Su and Heindel, 2004), but the oxygen transfer coefficient was significantly increased by adding packing into the column (Moraveji *et al.*, 2011a).

Nikakhtari and Hill (2005) used stainless steel meshes as a packing with 99.0% porosity in the riser

section of an external loop airlift bioreactor (ELAB). According to this research, the overall volumetric oxygen transfer coefficient increased (by an average factor of 2.45) in a packed bed ELAB compared to the same ELAB without packing. The overall volumetric oxygen transfer coefficient reached 0.021 s^{-1} at a gas superficial velocity of 0.0157 m/s in a packed reactor.

The aim of this research is to investigate the effect of aeration velocity and liquid properties on the hydrodynamic parameters and volumetric mass transfer coefficient in a packed split-cylinder airlift reactor. The packing was installed in the riser section of the airlift reactor.

Four different oil-in-water micro-emulsions containing kerosene, heavy naphtha, light naphtha, and diesel as the oil at a concentration of 7 % (v/v) were prepared and their behavior was carefully studied. The surface tension of the bulk liquid decreases and smaller bubbles were produced in micro-emulsions in comparison with pure water. Therefore, the gas hold-up and mass transfer coefficient increased in micro-emulsions; however, adversely liquid circulation velocity decreased.

EXPERIMENTAL

Materials and Methods

Different petroleum fractions containing kerosene, heavy naphtha, light naphtha and diesel were purchased from Shazand Oil refinery Company (Arak, Iran) and their various solutions at a concentration of 7% (v/v) were prepared locally. Micro-emulsions were prepared from tap water and petroleum fractions. Nonyl Phenol [(NF-60) purity: 99.5% purchased from Isfahan Copolymer Company] was added as the emulsifier. The emulsifier, petroleum fractions and liquid properties are respectively summarized in Tables 1-3.

Table 1: Physical properties of the emulsifier NF-60.

HLB	pH	Cloud point (°C)	Density at 20 °C (g/cm ³)	Water wt%	Avg M.W.	Appearance	Avg EO mole	Trade name
10.9	5-7	60±4	1.045±0.01	0.5 Max	484	Oily liquid	6	NF-60

Table 2: Physical properties of the oil fractions at 20 °C.

RSH (ppm)	H ₂ S	Pour point	Flash point	FBP (°C)	IBP	Sp gravity 15.5/15.5 °C	Trade name
<10	FREE	—	—	89	47	0.668	L. Naphtha
<10	FREE	—	—	157	95	0.7495	H. Naphtha
<10	FREE	—	49	257	161	0.8035	Kerosene
<10	FREE	3	111	380	239	0.8265	Diesel

Table 3: Physical properties of liquid.

Liquid	Oil-in-water volume	Density (kg/m ³)	Kinematic viscosity 10 ⁻⁶ (m ² /s)	Surface tension (mN/m)
Water	—	998.2	0.902	72.8
Water/L. Naphtha	7%	973.829	1.42	24.293
Water/H. Naphtha	7%	979.517	2.11	25.116
Water/Kerosene	7%	983.286	3.70	26.045
Water/Diesel	7%	984.891	27.19	27.532

Apparatus Set Up and Measurement Methods

The split-cylinder airlift reactor used in this research was the same as the apparatus applied by Moraveji *et al.* (2011b), as shown in Figure 1. The main difference between our case and the literature is ceramic Pall Ring packing (as shown in Figure 2), which was inserted in the riser section of the airlift reactor. The packing density, dry factor and specific surface area were 540 kg/m³, 356 m⁻¹ and 150 m²/m³, respectively. The maximum possible porosity of the packing is equal to 78.0%. As is known, packing normally increases the mass transfer and gas hold-up. Therefore, packing was applied in this work to

compare the unpacked results (Moraveji *et al.*, 2011a) with the packed ones.

All experiments were carried out in ambient conditions [atmospheric pressure and 25(± 0.5) °C]. A dissolved oxygen electrode (WTWCellox325) for oxygen concentration measurement in the liquid bulk was set in the riser zone at a depth of 0.1 m from the surface of the gas free liquid. The probe's tip was at an angle of 30° to the horizon to prevent oxygen bubbles from sticking to it. The conductivity electrode (Model 740i, WTW, Germany) for liquid circulation velocity and mixing time measurements was positioned in the down-comer zone at a depth of 0.2 m from the bottom of the reactor.

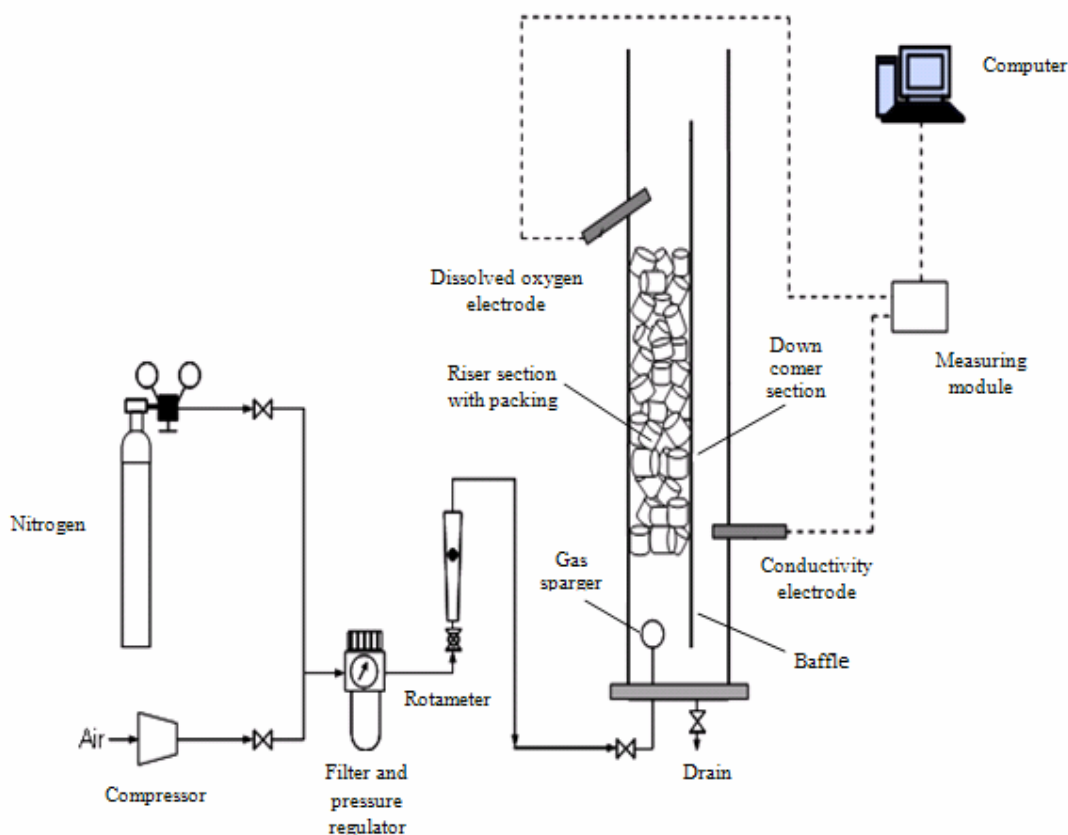


Figure 1: Schematic diagram of the split-cylinder airlift reactor.



Figure 2: Ceramic Pall Ring packing used in this research.

In this study, the volume expansion method was applied to measure the overall gas hold-up (ϵ) during the steady state condition in the airlift bioreactor. Liquid circulation velocity (U_L) and oxygen mass transfer coefficient measurement methods have been proposed and reviewed elsewhere (Moraveji *et al.*, 2011b).

RESULTS AND DISCUSSION

Overall Gas Hold-Up

Figure 3 shows the overall gas hold-up for four different micro-emulsions versus superficial gas velocity. Three independent parameters involving surface tension (σ), density (ρ) and viscosity (ν) affect the micro-emulsions properties in an unpacked airlift reactor (Moraveji *et al.*, 2012b).

According to the literature, the order of the surface tension, density and viscosity for the four different micro-emulsions (at the same concentration) is the following (Moraveji *et al.*, 2012c):

light naphtha < heavy naphtha < kerosene < diesel

As shown in Figure 3, the gas hold-up originally increased with an increase of the superficial gas velocity. Furthermore, the gas hold-up for oil-in-water micro-emulsion systems is significantly higher than that of the pure water system. This is due to the reduction of surface tension and the increased hindering of coalescence characteristic of the micro-emulsion systems (compared to pure water). Therefore,

in the micro-emulsion systems small bubbles are created and gas hold-up increases.

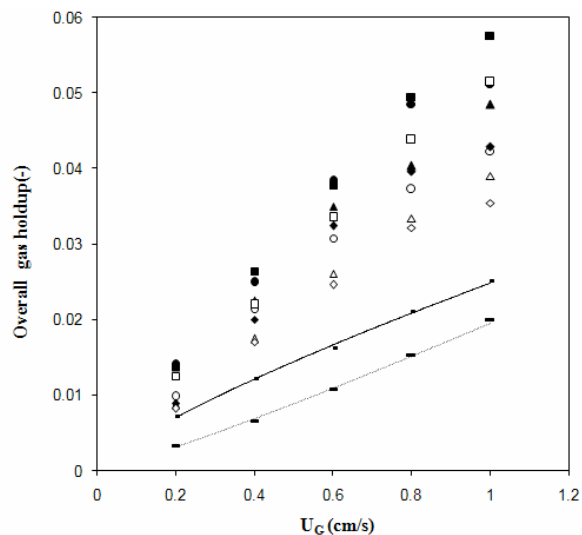


Figure 3: Overall gas hold-up versus superficial air velocity (U_G).

(♦ Diesel 7%; ■ Light Naphtha 7%; ▲ Kerosene 7%; ● Heavy Naphtha 7%; - pure water) Packed.
(◇ Diesel 7%; □ Light Naphtha 7%; △ Kerosene 7%; ○ Heavy Naphtha 7%; - pure water) Unpacked.

In the solutions with higher densities, buoyancy force enhancement helps the bubbles to rise, so the gas hold-up decreases. Although viscosity enhancement slightly increases the bubble residence time and gas hold-up, it creates bigger bubbles and increases the buoyancy force. Therefore, the gas hold-up decreases.

Further, packing decreases the gas bubble size. Some bubbles are captured inside the packing, so the gas hold-up increases.

According to our research, the maximum gas hold-up in a packed bed reactor was obtained for a light naphtha-in-water micro-emulsion. It was 60% more than that of pure water at the highest aeration velocity ($U_G=1\text{cm/s}$). Therefore, the overall gas hold-up increased as follows:

water < diesel < kerosene < heavy naphtha < light naphtha

According to our conditions in this research, the gas hold-up was correlated by applying dimensionless numbers as:

$$\frac{\varepsilon}{(1-\varepsilon)^4} = 0.42\text{Bo}^{0.26}\text{Ga}^{0.08}\text{Fr} \quad (1)$$

where, Bo, Ga and Fr are the Bond number [the ratio of body forces (which often is equal to the gravity forces) to the surface tension forces], the Galilei number (the ratio of gravity forces to viscous forces) and the Froude number (the ratio of inertial forces to the gravity forces), respectively.

A comparison between the experimental data and the correlated results for the gas hold-up is illustrated in Figure 4. As shown in this figure, Eq. (1) can predict the data with 16% error. Sada *et al.*'s equation showed a good agreement with the experimental data while Akita and Yoshida's equation showed some errors. This is due to the Bo number effect.

Table 4 compares two correlations obtained from the literature (Sada *et al.*, 1984; Akita and Yoshida, 1974) and our correlation.

Liquid Circulation Velocity

It was experimentally found that the liquid circulation velocity (U_L) strongly depends on the gas hold-up. The difference of gas hold-up between the riser and the down-comer zones of the airlift reactor provides the driving force for the liquid circulation.

Figure 5 shows the experimental data for the velocity of bulk liquid circulation in the bioreactor versus the superficial gas velocity in the riser for water and various micro-emulsions employed in this study. The gas hold-up sharply increases in the down-comer with a decrease of surface tension and bubble diameter. Therefore, the difference of gas hold-up between the riser and down-comer decreases. On the other hand, the driving force for the liquid circulation decreases. So, the velocity of the micro-emulsions liquid circulation velocity is less than that of pure water. The liquid circulation velocity increases as:

light naphtha < heavy naphtha < kerosene < diesel < water

At the highest aeration velocity ($U_G=1\text{cm/s}$), the minimum liquid circulation velocity was observed for light naphtha (30% less than that of pure water) and the maximum liquid circulation velocity was observed for diesel (8.5% less than that of pure water).

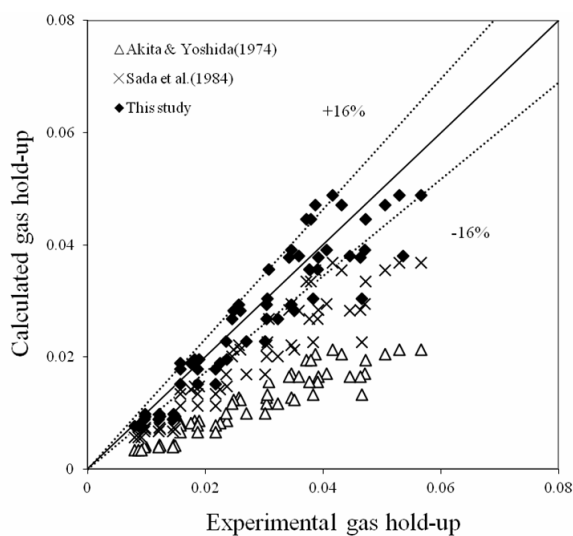
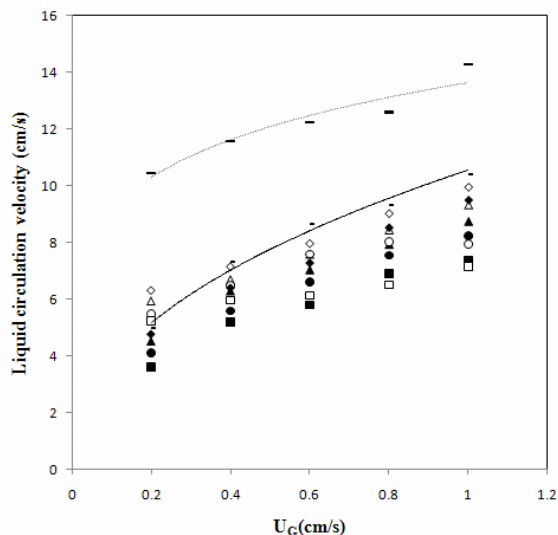


Figure 4: The correlated data versus experimental gas hold-up.

Table 4: Gas hold-up correlations based on Bo number, Ga number and Fr number.

Correlations	Remarks	References
$\frac{\varepsilon}{(1-\varepsilon)^4} = 0.2Bo^{1/8}Ga^{1/12}Fr$	$d_b > 2.5\text{mm}$	Akita & Yoshida (1974)
$\frac{\varepsilon}{(1-\varepsilon)^4} = 0.32Bo^{0.21}Ga^{0.086}Fr$	$0.3 < U_G < 30\text{cm/s}$	Sada <i>et al.</i> (1984)
$\frac{\varepsilon}{(1-\varepsilon)^4} = 0.42Bo^{0.26}Ga^{0.08}Fr$	$0.2 < U_G < 1\text{cm/s}$	This study

**Figure 5:** Liquid circulation velocity in the riser–down–comer loop versus aeration velocity (U_G).

(♦ Diesel 7%; ■ Light Naphtha 7%; ▲ Kerosene 7%; ● Heavy Naphtha 7%; - pure water) Packed.
 (◇ Diesel 7%; □ Light Naphtha 7%; △ Kerosene 7%; ○ Heavy Naphtha 7%; - pure water) Unpacked.

Mixing time

Figure 6 shows the mixing time (t_m) versus superficial gas velocity for micro-emulsions and pure water. The mixing time decreased by increasing the aeration velocity in the riser. As shown in this figure, the mixing time increased as follows:

water < diesel < kerosene < heavy naphtha < light naphtha

The mixing time enhancement is due to the decrease of surface tension and bubble diameter in micro-emulsions. Circulation of bubbles in the reactor assists the mixing process. Packing installation has a negative impact on the magnitude of the induced liquid circulation velocity and increases the mixing time.

Mass Transfer

Figure 7 shows the volumetric mass transfer coefficient ($k_L a$) for pure water and four different micro-emulsions versus the superficial gas velocity. As shown in this figure, $k_L a$ values increased upon increasing the superficial gas velocity. The $k_L a$ values for all of the micro-emulsions were significantly greater than that of pure water, as:

water < diesel < kerosene < heavy naphtha < light naphtha

However, the maximum $k_L a$ was obtained for the light naphtha micro-emulsion (about 31% more than that of pure water), but the minimum $k_L a$ was obtained for the diesel micro-emulsion (about 4.7%

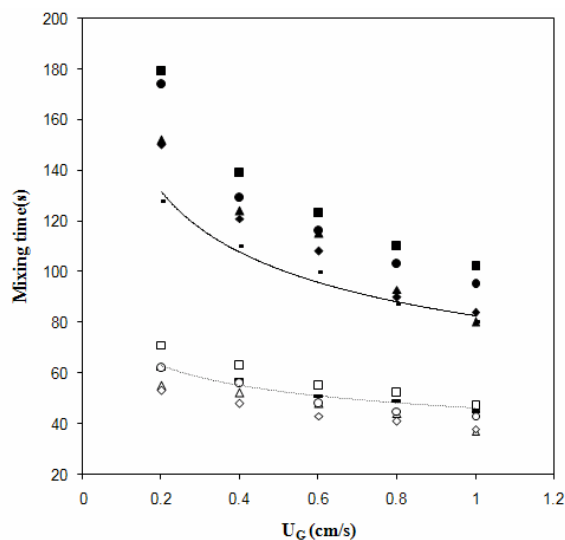


Figure 6: Mixing time versus superficial air velocity U_G in the riser.

(◆ Diesel 7%; ■ Light Naphtha 7%; ▲ Kerosene 7%; ● Heavy Naphtha 7%; - pure water) Packed.
(◇ Diesel 7%; □ Light Naphtha 7%; △ Kerosene 7%; ○ Heavy Naphtha 7%; - pure water) Unpacked.

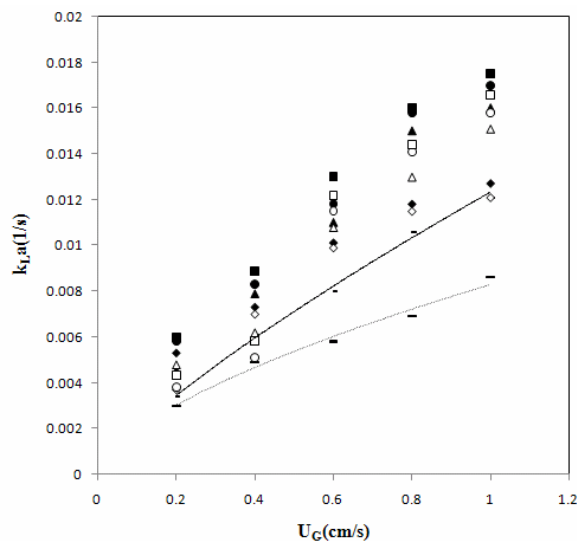


Figure 7: Overall volumetric oxygen mass transfer coefficient ($k_L a$) versus superficial air velocity (U_G) in the riser.

(◆ Diesel 7%; ■ Light Naphtha 7%; ▲ Kerosene 7%; ● Heavy Naphtha 7%; - pure water) Packed.
(◇ Diesel 7%; □ Light Naphtha 7%; △ Kerosene 7%; ○ Heavy Naphtha 7%; - pure water) Unpacked.

more than that of pure water) (at the highest superficial air velocity).

The surface tension of water is higher (more than three times) than that of the oily micro-emulsions. This causes a bubble diameter reduction of micro-emulsions. The gas-liquid interfacial area (a) increases due to bubble diameter reduction. Therefore, the mass transfer increases.

Further, a liquid viscosity enhancement increases the bubble coalescence rate. It means that the interfacial area decreases in size. The liquid viscosity enhancement thickens the liquid boundary layer bubbles (Appasani, 2007) and decreases the solute diffusivity (Wilke and Chang, 1955). Therefore, $k_L a$ values decrease upon increasing the viscosity of the oily micro-emulsions (Kilonzo and Margaritis, 2004; Calderbank, 1967).

Density is another bulk liquid property that plays a major role in the overall mass transfer. By increasing density, the buoyancy force increases. Therefore, bubbles can rise easily and rapidly. Furthermore, the necessary time for mass transfer between the two phases decreases upon increasing the density.

In addition, the packing increases the gas hold-up, and decrease the liquid circulation velocity and gas bubble size in an airlift reactor.

The volumetric mass transfer coefficient for oil-

in-water micro-emulsions was correlated using the Sh number with the following equation:

$$\text{Sh} = 0.14 \text{Re}^{2/3} \text{Sc}^{1/2} \text{Bo}^{0.98} \quad (2)$$

where Sh, Re and Sc are the Sherwood number (the ratio of convective and diffusive mass transfer coefficients), the Reynolds number (the ratio of inertial forces to viscous forces) and the Schmidt number (the ratio of momentum diffusivity (viscosity) to mass diffusivity), respectively. For this equation, the coefficient of determination (R^2) was around 0.96.

However, our correlation was in good agreement with the results obtained by Asgharpour *et al.* (2010) and Akita and Yoshida (1974), but the Bird *et al.* (2002) correlation was not in good agreement with our experimental data. This may be due to the Bond number influence. In such solutions, the liquid density, surface tension and bubble diameter clearly change, so the Bo number affects the results and cannot be neglected. All of the correlations are illustrated in Figure 8 and compared with our correlated data. Table 5 also shows the corrected correlations based on the literature (Akita and Yoshida, 1974; Asgharpour *et al.*, 2010; Bird *et al.*, 2002) and our correlation.

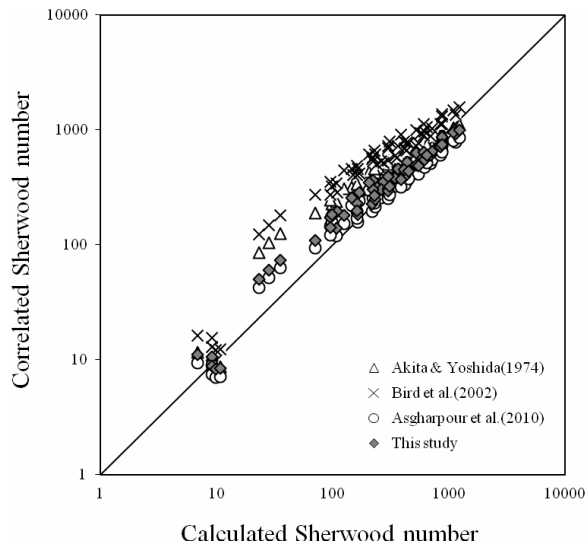


Figure 8: The correlated Sherwood number versus the Sherwood numbers obtained from the experimental work.

Table 5: Sherwood number correlations for the liquid-gas mass transfer coefficient based on Re number, Sc number and Bo number.

Correlations	Remarks	References
$Sh = 0.15 Re^{2/3} Sc^{1/2} Bo^{2/3}$	$0.118 < U_G < 2.35 \text{ cm/s}$	Asgharpour <i>et al.</i> (2010)
$Sh = (4/\pi)^{1/2} Re^{1/2} Sc^{1/2}$	Higbie's model	Bird <i>et al.</i> (2002)
$Sh = 0.6 Re^{1/2} Sc^{1/2} Bo^{3/8}$	Homogeneous flow	Akita & Yoshida (1974)
$Sh = 0.14 Re^{2/3} Sc^{1/2} Bo^{0.98}$	$0.2 < U_G < 1 \text{ cm/s}$	This study

CONCLUSIONS

In this study, gas hold-up, liquid circulation velocity, mixing time and the volumetric oxygen transfer coefficient were considered in a packed split-cylinder airlift reactor and compared with the unpacked reactor data. Four different oil-in-water micro-emulsions [containing light naphtha, heavy naphtha, kerosene and diesel at a concentration of 7% (v/v)] were used and compared with the pure water system. The light naphtha based micro-emulsion with the lowest surface tension and viscosity had the maximum mass transfer coefficient and gas hold-up and had the minimum liquid circulation velocity, while diesel with the highest surface tension and viscosity had the minimum mass transfer coefficient and gas hold-up and had the maximum liquid circulation velocity. Since gas bubbles were captured inside the packing, the mass transfer increased compared to an unpacked reactor. Furthermore, these bubbles increased the gas hold-

up. The bubble size was more uniform and they had smaller diameters after passing through the packing. In addition, for the micro-emulsion systems in an airlift reactor, two appropriate correlations based on dimensionless groups (Bo, Ga and Fr for gas hold-up and Bo, Re and Sc for Sh) were developed. A satisfactory agreement between the correlated data and experimental results was observed.

NOMENCLATURE

a	gas-liquid interfacial area per unit volume of the liquid	m^2/m^3
Bo	Bond number $\left(\frac{\rho g d_b^2}{\sigma_L} \right)$	(-)
d_b	bubble diameter	m
D	gas diffusivity	m^2/s
Fr	Froude number $\left(\frac{U_G}{\sqrt{g \cdot d_b}} \right)$	(-)

G	acceleration due to gravity	m/s ²
Gal	Galilei number $\left(d_b^3 \cdot \rho^2 \cdot \frac{g}{\mu^2} \right)$	(-)
k _L	liquid film mass transfer coefficient	m/s
k _L a	overall volumetric gas-liquid mass transfer coefficient	s ⁻¹
O/W micro-emulsion	oil-in-water	(-)
Re	Reynolds number $\left(d_b \cdot U_G \cdot \frac{\rho_L}{\mu} \right)$	(-)
Sh	Sherwood number $\left(k_L \cdot \frac{d_b}{D} \right)$	(-)
Sc	Schmidt number $\left(\frac{\mu}{\rho D} \right)$	(-)
t _m	mixing time	s
U _G	superficial aeration velocity in the riser zone	m/s
U _L	liquid circulation velocity	m/s
W/O	water-in-oil micro-emulsion	(-)

Greek Symbols

ε	overall gas hold-up	(-)
ν	kinematic viscosity	m ² /s
μ	viscosity of phase	k/Pa.s
ρ	density of phase	kg/m ³
σ	surface tension	mN/m
φ	water-to-oil phase volume ratio	(-)

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