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# Continuous removal of carbon dioxide by absorption employing a bubble column

E. Álvarez[a], D. Gómez-Díaz[b], J.M. Navaza[b,*], B. Sanjurjo[b]

[a] Department of Chemical Engineering, University of Vigo, Galicia, Spain
[b] Department of Chemical Engineering, University of Santiago de Compostela, Galicia, Spain

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**Abstract**

The present work includes studies related to the gas/liquid mass transfer process of carbon dioxide in polluted aqueous solutions with surface active substances. These studies have been carried out employing a thermostated cylindrical bubble column, where the gas phase was fed through different gas spargers based on glass porous plates. The absorption process has been characterized taking into account the influence of the gas and the liquid flow-rate, as well as the surfactant liquid phase composition and bubble size. Two opposite effects have been detected in relation to the influence of the surfactant presence upon the mass transfer rate.

*Keywords:* Carbon dioxide; Bubble column; Absorption

## 1. Introduction

Bubble contactors are frequently used in the chemical industry as absorbers, fermenters and reactors, where heterogeneous gas–liquid and gas–solid–liquid operations and reactions take place, in processes that require high contact areas among phases. In these contactors the gas is dispersed in the liquid and/or solid phases in the shape of bubbles, which provide high contact areas. The mass transfer process also occurs during both the bubble formation and the bubble rise [1].

Determining the mass transfer coefficients governing the absorption of gases by liquids in bubble contactors is a growing concern due to the frequent use of these equipment in biotechnological processes whose rates are often limited by the oxygen absorption rate [2].

The influence of liquid phase physical properties upon the gas mass transfer velocity is one of the most important parameters that must be taken into account in gas/liquid processes and has been detected by numerous studies [3–5]. This influence causes that the physical characterization of the liquid phases involved in gas/liquid mass transfer processes will be a previous and necessary step to interpret the absorption experimental results. Moreover, this characterization has great importance to optimise the process to obtain a better efficiency in the mass transfer velocity.

The presence of surfactants undoubtedly affects the formation and coalescence of bubbles [6], and hence the interfacial area. Therefore, it is possible that the surfactant-induced reduction of $k_L$, due to barrier effects, may be offset by means of increasing the area to afford increased values of $k_La$. For absorption through a flat surface, the reduction of the individual mass transfer coefficient, $k_L$, by means of a surfactant, due to whether hydrodynamic or barrier effects [7], can be offset by stirring, which renews the surface. However, there are few quantitative data relating to the influence of surface tension or surfactant presence upon the volumetric mass transfer coefficient, $k_La$, when the gas to be absorbed is bubbled through the liquid phase.

## 2. Experimental

The surfactant employed in the present study was sodium dodecyl sulphate (SDS) (CAS number 151-21-3) supplied by Fluka with a purity of $\geq 96\%$, and aqueous solutions of SDS were prepared employing an analytic balance with a precision of $\pm 10^{-7}$ kg.

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[*] Corresponding author at: Department of Chemical Engineering, ETSE, University of Santiago de Compostela, Rúa Lope Gómez de Marzoa, Santiago de Compostela E-15782, Spain. Fax: +3481595012.

*E-mail address:* eqnavaza@usc.es (J.M. Navaza).

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### Nomenclature

| | |
|---|---|
| $a$ | specific interfacial area |
| $C$ | $CO_2$ concentration |
| $C^*$ | $CO_2$ solubility |
| $k_L$ | liquid mass transfer coefficient |
| $k_La$ | volumetric liquid mass transfer coefficient |
| $n$ | number of bubbles |
| $N$ | absorption intensity |
| $Q_g$ | gas phase flow-rate |
| $Q_L$ | liquid phase flow-rate |
| SDS | sodium dodecyl sulphate |
| $t$ | operation time |
| $U_g$ | superficial gas velocity |
| *Greek symbols* | |
| $\varepsilon$ | gas hold-up |
| $\nu$ | kinematic viscosity |
| $\rho$ | liquid phase density |
| $\sigma$ | surface tension |

The surface tension (at 25 °C) was determined employing a Krüss K-11 tensiometer using the Wilhelmy plate method. This method has been employed by our team in different studies and a clear description of methodology is described in previous papers [8]. The plate employed was a commercial platinum one supplied by Krüss. The platinum plate was cleaned and flame dried before each measurement. The surface tension of pure water was determined and compared with the literature to confirm that this method provides suitable results [9].

The experimental set-up for absorption experiments has been described in detail in previous papers [5], except for the contact device (bubble column). A schematic diagram of the contact device and experimental set-up is shown in Fig. 1. A bubble column (10 L of volume) was used as contact device in this work, made of two methacrylate concentric cylindrical tubes, 108.6 cm high and 4 mm thick, welded to two flat bases made of the same material. The internal and external diameters of the column are 11.3 and 14.8 cm, respectively. On the external tube there are two side orifices for the inward and outward flow of the thermostated liquid which circulates throughout the empty space between the two tubes. All absorption experiments were carried out at 25 °C using a thermostat shown at experimental set-up present in Fig. 1. The gas to be absorbed was passed through of two humidifiers at 25 °C and entered the contact device at a constant flow-rate. The gas absorption rate was calculated as the difference between inflow and outflow rates. The operation regime was continuous for both phases involved in the absorption process.

The upper cover of the column that is easily removable, is a methacrylate sheet with three openings: the centre orifice is for a thermometer, and the two off-centre orifices are for the outflow of gas and the inflow of liquid phase. The lower cover has also three openings: the middle one is for a sparger plate of 4 cm in diameter, and one of the others is employed for the outflow of

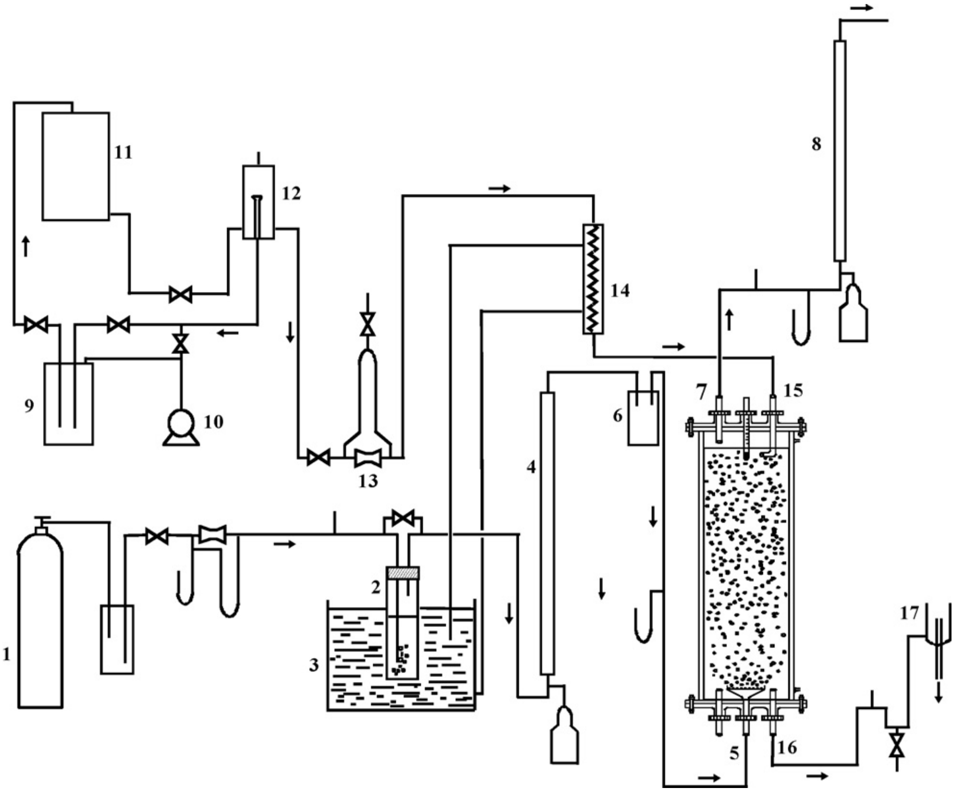

Fig. 1. Experimental set-up employed in continuous absorption experiments. (1) Gas cylinder; (2) humidifier; (3) thermostatic bath; (4) soap flowmeter; (5) gas inlet; (6) security vessel; (7) gas outlet; (8) soap flowmeter; (9) liquid reservoir; (10) compressor; (11) liquid reservoir; (12) constant level reservoir; (13) liquid flowmeter; (14) heat exchanger; (15) liquid inlet; (16) liquid outlet; (17) bubble liquid height regulator.

Table 1
Characteristics of gas spargers

| Sparger | Medium pore diameter (μm) |
|---|---|
| N° 0 | 175 |
| N° 1 | 120 |
| N° 2 | 65 |

the liquid phase. Three glass porous plates with different pore diameters have been used as gas spargers. The characteristics of these spargers are listed in Table 1.

## 3. Results and discussion

The evaluation of mass transfer rate in the absorption process under the operation regime employed could be expressed by Eq. (1).

$$N = k_{\mathrm{L}}a\Delta C_{\mathrm{m\,log}} \quad (1)$$

where $N$ is the absorption intensity, $k_La$ is the volumetric mass transfer coefficient and $\Delta C_{\mathrm{mlog}}$ is the driving force. Rochelle et al. have recommended [10] Eq. (2) for calculation of driving force for the experimental procedure employed in the present study.

$$\Delta C_{\mathrm{m\,log}} = \frac{(C^* - C)_2 - (C^* - C)_1}{\ln (C^* - C)_2/(C^* - C)_1} \quad (2)$$

where $C^*$ is the carbon dioxide solubility, and $C_1$ and $C_2$ are the concentration of carbon dioxide absorbed, respectively, at the outlet and inlet of the bubble column in the liquid phase.

Owing to the fact that the concentration of carbon dioxide at the inlet stream of the liquid phase is zero ($C_2 = 0$) and the gas solubility values at the inlet and outlet of the bubble column are the same ($C_1^* = C_2^* = C^*$), the expression to calculate the volumetric mass transfer coefficient is reduced to Eq. (3).

$$k_{\mathrm{L}}a = \frac{N}{C_1} \ln \frac{C^*}{C^* - C_1} \quad (3)$$

The experimental methodology employed in the present work implies the analysis of the gas and liquid flow-rates influence upon the absorption process. The influences generated by both variables upon the mass transfer rate, and more specifically upon the volumetric mass transfer coefficient value, are summarised in Fig. 2. The experimental results included in Fig. 2 indicate that an increase in both operation variables (gas and liquid flow-rates) produce an increase in the value of the volumetric mass transfer coefficient. High values of gas flow-rate or superficial gas velocity in the experimental range employed in this work have low influence, and then the volumetric mass transfer coefficient remains almost constant. This behaviour is due to the fact that an increase in the gas flow-rate fed to the bubble column produces an increase in the quantity of gas phase dispersed in the liquid phase. Nevertheless, the bubble coalescence phenomenon produces, at high gas flow-rate values, an inhibition of the interfacial area increment when the gas phase fed is increased.

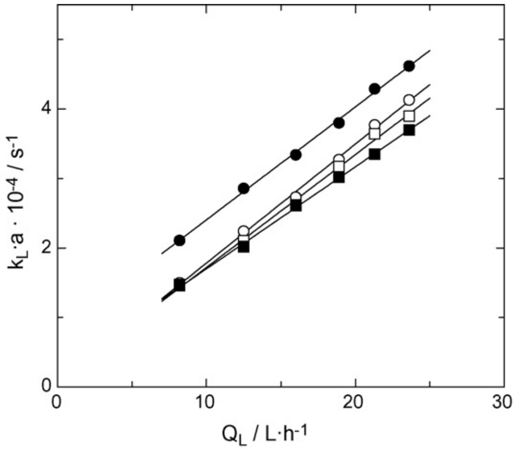

Fig. 2. Influence of superficial gas velocity, liquid phase flow-rate and SDS concentration in the liquid phase upon the volumetric mass transfer coefficient. Sparger N° 0. [SDS] = 0 M: (○) $U_g = 1.8 \times 10^{-3}\ \mathrm{m\,s^{-1}}$ and (□) $U_g = 1.18 \times 10^{-3}\ \mathrm{m\,s^{-1}}$; [SDS] = $10^{-3}$ M: (●) $U_g = 1.8 \times 10^{-3}\ \mathrm{m\,s^{-1}}$ and (■) $U_g = 1.18 \times 10^{-3}\ \mathrm{m\,s^{-1}}$.

In relation to the effect caused by the liquid flow-rate fed to the bubble column upon the mass transfer rate, a continuous increase can be observed. This is due to the fact that this variable increases the value of the driving force, and this behaviour produces an increase in the volumetric mass transfer coefficient.

A first example of the influence of a surface contaminant (SDS) upon the absorption process is shown also in Fig. 2. This figure shows an effect of the presence of SDS upon the volumetric mass transfer process, although no clear influence was observed. At high values of gas flow-rates, the experimental results show a positive effect of SDS presence enhancing the mass transfer rate, but a decrease in the gas phase fed to the contactor shows a negative influence of SDS upon mass transfer. This experimental behaviour implies the necessity of improving the experiments to analyse the influence of SDS concentrations.

Absorption experiments with different SDS concentration liquid phases have been carried out under the experimental conditions of gas and liquid flow-rates previously mentioned. Fig. 3

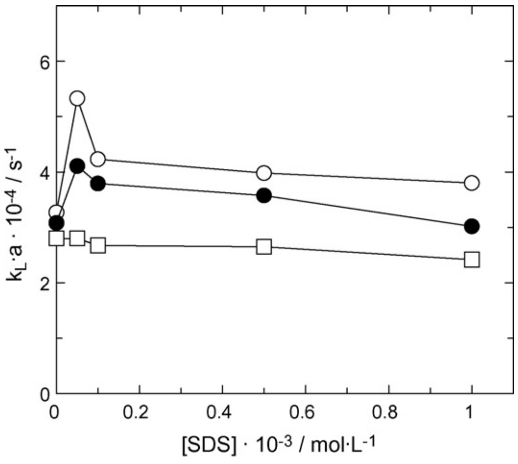

Fig. 3. Influence of SDS concentration of liquid phase upon the volumetric mass transfer coefficient. Sparger N° 0. $Q_L = 19\ \mathrm{L\,h^{-1}}$. (○) $U_g = 1.8 \times 10^{-3}\ \mathrm{m\,s^{-1}}$; (●) $U_g = 1.34 \times 10^{-3}\ \mathrm{m\,s^{-1}}$; (□) $U_g = 0.85 \times 10^{-3}\ \mathrm{m\,s^{-1}}$.

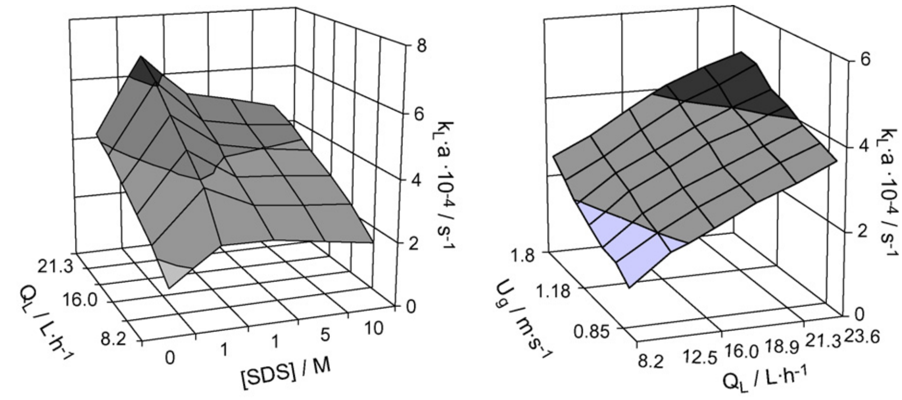

Fig. 4. Influence of operation conditions upon volumetric mass transfer coefficient. Left: Sparger N° 0. $U_g = 1.8 \times 10^{-3}\ \mathrm{m\,s^{-1}}$; Right: Sparger N° 0. $[SDS] = 10^{-3}\ \mathrm{mol\,L^{-1}}$.

shows a summary of the experimental results obtained, which indicate that a low surfactant concentration in the liquid phase enhances the value of the volumetric mass transfer coefficient, but a continuous increase in the SDS concentration produces a decrease in the value of this coefficient. This behaviour is emphasized at high values of the gas flow-rate in the studied range.

A similar behaviour was observed in other studies [11] in the absorption process of ammonia in aqueous solutions contaminated with surfactants. The explanation for this behaviour is related with the simultaneous action of two opposite effects: (i) a positive influence due to an increase in the interfacial turbulence, due to the existence of surface tension gradients [12] and (ii) a negative effect due to the barrier effect [13].

Fig. 4 shows a complete plot to analyse the combining influences of different operational variables employed in the present work. This figure shows the complicated behaviour observed by the influence of SDS concentration upon the value of the volumetric mass transfer coefficient, observing a maximum value. In relation to the influence of gas and liquid flow-rates upon the mass transfer, both variables produce positive effects increasing the value of the volumetric mass transfer coefficient.

In relation to the positive effect commented previously, caused by the interfacial turbulence, Fig. 5 shows the influence of SDS concentration in aqueous solutions upon the value of the surface tension. This behaviour shows that small quantities of contaminant produce important variations in the value of the physical property analysed. The differences between the surface tension in the liquid surface and in the liquid bulk generate the formation of turbulences in the proximity of the gas–liquid interface [11]. As a result, an increase in the frequency of liquid surface elements renovation at the interface is produced. And this increase, taken into account the renewal theory, produces an increase in the mass transfer rate.

On the other hand, mass transfer is reduced at high SDS concentration due to the barrier effect based on the presence of surfactant molecules at the gas–liquid interface. The presence of these molecules produces a reduction on the interfacial area and also certain studies [13] have proposed that the decrease in the mass transfer rate is due to the reduction in the liquid film by the presence of surfactants. The described phenomenon causes a reduction in the volumetric mass transfer coefficient when SDS concentration in the liquid phase increases.

Another interesting study related to the absorption process of gases in aqueous surfactant solutions is the effect of the bubble size upon the mass transfer coefficient. Three types of gas spargers have been employed to develop this study. The characteristics of these spargers in relation to the pore diameter have been included in the Section 2.

The experimental results for absorption process shown that the use of a sparger with a smaller pore diameter produces a higher mass transfer rate. Due to the use of a sparger with small pores, it produces high interfacial areas because the generated bubbles are smaller than the others produced by the other spargers.

In relation with this parameter (interfacial area), the influence of SDS concentration, superficial gas velocity and gas sparger type has been analysed. This experimental data has been obtained from a previous study of our research team [14].

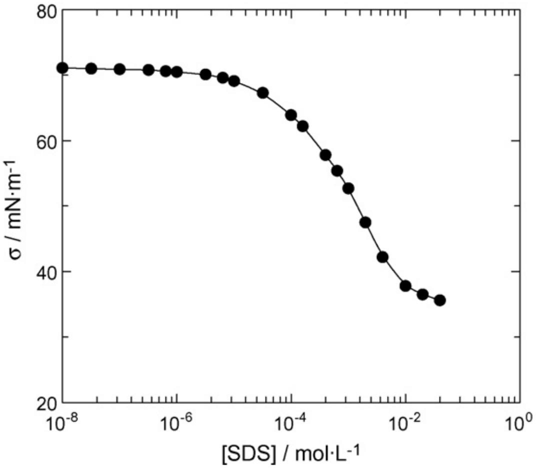

Fig. 5. Surface tension of aqueous solutions of SDS.

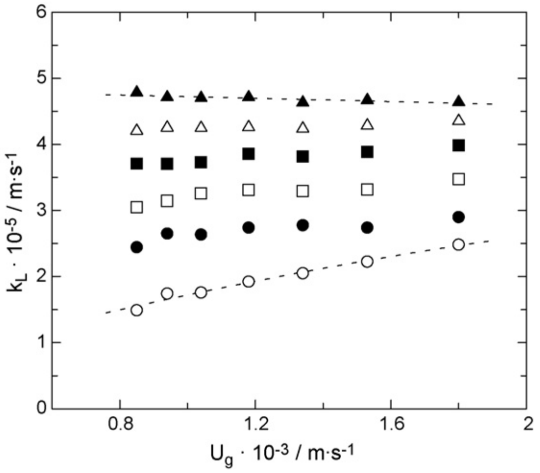

Fig. 6. Influence of superficial gas velocity and liquid flow-rate upon mass transfer coefficient. (○) $Q_L = 8.2\,\mathrm{L\,h^{-1}}$; (●) $Q_L = 12.5\,\mathrm{L\,h^{-1}}$; (□) $Q_L = 16\,\mathrm{L\,h^{-1}}$; (■) $Q_L = 19\,\mathrm{L\,h^{-1}}$; (△) $Q_L = 21.3\,\mathrm{L\,h^{-1}}$; (▲) $Q_L = 23.6\,\mathrm{L\,h^{-1}}$. Sparger N° 2.

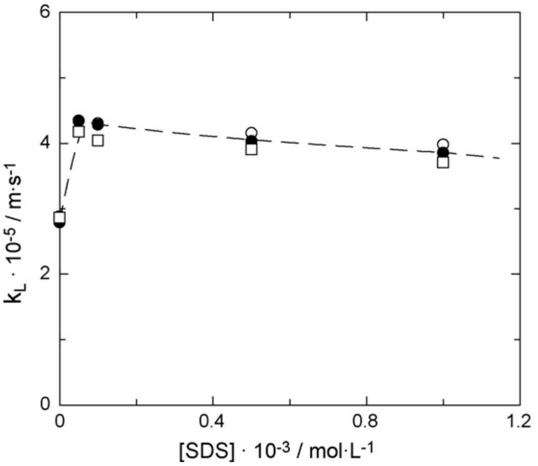

Fig. 7. Effect of SDS concentration upon mass transfer coefficient. $Q_L = 8.2\,\mathrm{L\,h^{-1}}$. (○) $U_g = 1.8 \times 10^{-3}\,\mathrm{m\,s^{-1}}$; (●) $U_g = 1.18 \times 10^{-3}\,\mathrm{m\,s^{-1}}$; (□) $U_g = 0.85 \times 10^{-3}\,\mathrm{m\,s^{-1}}$. Sparger N° 2.

The interfacial area data shows that surfactant presence produces a decrease at low surfactant concentration, but at higher concentrations, in the studied range, only a slight decrease is observed. This behaviour is due to the barrier effect caused at gas–liquid interface by the presence of surfactant molecules that reduce the mass transfer area.

On the other hand, an increase in gas flow-rate (superficial gas velocity) produces a clear increase on interfacial area due to the higher gas volume fed to contactor. Also the use of a gas sparger with smaller pore diameter generates a higher number of bubbles and then an increase in interfacial area.

Specific interfacial area values have been used to calculate liquid side mass transfer coefficient, $k_L$. Fig. 6 resumes the behaviour observed about the influence of operational conditions upon mass transfer coefficient. In this figure is possible observes two situations: (i) at low liquid phase flow-rate, an increase in superficial gas velocity causes an increase in mass transfer coefficient due to a higher turbulence in the liquid phase and then a better surface renewal that increases mass transfer. (ii) at high values of liquid flow-rate the effect of superficial gas velocity is negligible because the influence of liquid flow-rate is higher than the produced by the gas flow-rate. An increase in liquid phase flow-rate produces a clear increase in the value of mass transfer coefficient due to the higher driving force existed in liquid phase.

In relation with the influence of surfactant presence upon mass transfer coefficient, a similar behaviour that the previous one obtained and commented for volumetric mass transfer coefficient, has been observed. Dual behaviour was obtained, an increase in mass transfer coefficient at low concentrations of SDS until a maximum and a slight decrease at high concentration of surfactant, in the studied range. This behaviour agrees with previous studies [11] that employ different systems (i.e. $NH_3$ absorption in water in surfactant presence). This behaviour is explained on the basis of interfacial turbulence that enhance mass transfer due to liquid interface surface renewal, but an increase in surfactant concentration produces a reduction in the renewal process at liquid interface by the presence of a higher number of surfactant molecules at gas–liquid interface (Fig. 7).

## 4. Conclusions

The present paper analyses the gas/liquid mass transfer process of a pure gas phase to liquid phases formed by aqueous solutions of a surface contaminant working in continuous regime for gas and liquid phases. The flow-rate of both phases produces a clear increase in mass transfer rate due to the effect upon the interfacial area (gas flow-rate) or upon driving force (liquid flow-rate).

The presence of surfactant in the liquid phase causes a characteristic behaviour due to the positive effect upon the mass transfer at very low surfactant concentration, but a negative influence in the rest of the concentration range. This reduction is due to the decrease in the mass transfer interfacial area that is caused by the barrier effect (accumulation of surfactant molecules at gas–liquid interface).

The influence of surfactant concentration upon mass transfer coefficient confirms that exists an enhancement of mass transfer at low surfactant concentration due to interfacial turbulence but high concentrations of SDS produce a reduction of this turbulence by surfactant accumulation at interface.

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