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CO_2 capture by aqueous solutions of glucosamine in a bubble column reactor

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ABSTRACT

The present work analyses the carbon dioxide (CO_2) capture process by means of absorption with chemical reaction with glucosamine aqueous solutions, using a bubble column reactor (BCR). Experimental results indicate that this reagent has a similar behaviour to other common amines (i.e. monoethanolamine, widely used in CO_2 capture), as regarding the capture rate of this acidic gas. The value of the mass transfer coefficient corresponding to the liquid phase has been determined, and the effect of different operation conditions upon the value of that coefficient has also been analysed (amine concentration, pH and gas flow-rate).

1. Introduction

The fossil fuel combustion from power plants or refineries is one of the most important sources of carbon dioxide emission [1]. Several technologies are available to reduce the carbon dioxide emission from industrial gas streams, but the chemical absorption with alkanolamines (i.e. monoethanolamine: MEA, diethanolamine: DEA, di-2-propanolamine: DIPA, triethanolamine: TEA, and methyldiethanolamine: MDEA) is the most widely used method for low partial pressures of carbon dioxide [2,3]. This method is efficient and it usually permits the removal of a high percentage of the carbon dioxide emitted. Previous studies concluded that primary and secondary alkanolamines react, directly and reversibly, to the carbon dioxide by forming a zwitterion intermediate, which is deprotonated by the bases present in the solution to form a stable carbamate [4,5], even though the formation of the carbamate increases the reaction rate but usually limits the loading to 0.5 mol of CO_2/(mol of amine) [6]. By contrast, tertiary amines do not react directly to the carbon dioxide in order to form carbamate. Generally, tertiary amines react more slowly to CO_2 than primary and secondary amines [7].

Recently, our research team have performed interesting studies using 2-amino-2-deoxy-D-glucose, called glucosamine (GA), in order to analyse the absorption of carbon dioxide in a gas–liquid contactor. The absorption of CO_2 in this system is accompanied by a chemical reaction between the glucosamine and the CO_2 previously absorbed in the liquid phase. This is a first-order reaction regarding both compounds (CO_2 and GA), as it was concluded by our research group in previous studies [8]. The mechanism suggested for this reaction is a zwiteronic type, where the global process of absorption with a chemical reaction has a moderately fast regime, since the Hatta number (Ha) takes values between $0.3 < Ha < 3$ for the experimental conditions used in the present work [8]. The Hatta number is a dimensionless parameter that compares the absorption rate of a solute in a reactive system to the absorption rate of the same solute, in the case of physical absorption [8].

This new method of CO_2 capture has great advantages if we compare it to more conventional systems, based on the use of the amines abovementioned. These advantages are fundamentally a minor need of safety measures and a minor chemical risk when aqueous solutions of glucosamine are manipulated. Moreover, this system shows a kinetic capture of CO_2 with a similar rate to other systems based in amines industrially used, such as MEA or DEA [8,9]. These previous studies have analysed the kinetics of the chemical reaction but not the mass transfer process, necessary to evaluate the global absorption process, that is the aim of present study.

Then, in the present work, the behaviour of glucosamine aqueous solutions is analysed in order to capture CO_2 by means of an absorption process with a chemical reaction in a bubble column reactor (BCR), and this process permits us the analysis of the mass transfer process and the interfacial area generated between two phases (gas and liquid). Therefore, in addition to the capacity of CO_2 absorption, we have also analysed the influence of the CO_2 loading, the amine concentration in the liquid phase, as well as the gas flow-rate fed to the contactor upon the global process of CO_2 absorption and the interfacial area gas–liquid.

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Nomenclature

A	interfacial area [m^2]
a	specific interfacial area [m^2 m^{-3}]
C_A	carbon dioxide concentration [mol dm^{-3}]
$C_A{}^*$	carbon dioxide solubility [mol dm^{-3}]
C_B	glucosamine concentration [mol dm^{-3}]
D_A	carbon dioxide diffusivity [m^2 s^{-1}]
d	equivalent sphere diameter [m]
d_{32}	Sauter mean diameter [m]
E	major axis of the projected ellipsoid [m]
e	minor axis of the projected ellipsoid [m]
F_{abs}	carbon dioxide absorbed [mol]
Ha	Hatta number
k_2	rate constant [m^3 mol^{-1} s^{-1}]
k_L	liquid mass transfer coefficient [m s^{-1}]
N	absorption rate [mol m^{-3} s^{-1}]
n	number of bubbles
Q_g	gas flow-rate [L h^{-1}]
t	operation time [s]
Greek symbols	
ε_G	gas hold-up [m]
η	liquid phase viscosity [mPa s]

2. Experimental

Different quantities of 2-amino-2-deoxy-D-glucose hydrochloride or glucosamine hydrochloride (GA), supplied by Fluka (CAS number 66-84-2) with a purity >99%, have been employed to produce aqueous solutions to use in a bubble column reactor. The solutions were prepared by mass, using a balance with a precision of $\pm 10^{-7}$ kg. Bi-distilled water has been employed to prepare the absorbent phases (in the range 0–0.4 mol dm^{-3}). Commercial grade carbon dioxide of 99.998% purity, supplied by Carburos Metálicos, was also used in this work.

The dynamic viscosity was obtained from the result product of the kinematic viscosity and the corresponding density of the mixture. The kinematic viscosity was determined from the transit time of the liquid meniscus through a capillary viscosimeter (Schott-Geräte AVS 350 Ubbelohde viscometer) with an electronic stopwatch with an accuracy of ±0.01 s to measure efflux times. The density of liquid phases was measured with an Anton Paar DSA 5000 vibrating tube densimeter.

All the absorption experiments were performed at room temperature, operating in batches with regard to the liquid phase. The bubble column is made of methacrylate, 1.03 m high, and it has a square cross-section (side length of 6 cm). The volume of the liquid phase employed in the bubble contactor was 3 L. The gas sparger has been a glass capillary with only one orifice, so the production of a small number of bubbles lets us analyse carefully the influence of the operation conditions on the bubbles size.

The gas to be absorbed was passed through two bubbling flasks to prepare the gas phase. This procedure removed other resistances to the mass transport and it only allowed us the evaluation of the liquid phase resistance to the gas transfer. Pure water was placed into the bubbling flasks. The gas flow-rate was measured and controlled with two mass flow controllers (5850 Brooks Instruments). The mass flow controllers employed in the present study for the gas flow-rate and pressures were calibrated by the supplier. The gas flow-rates employed have been included into 18–40 L h^{-1}. The pressure drop was measured between the column's inlet and outlet, using a Testo 512 digital manometer.

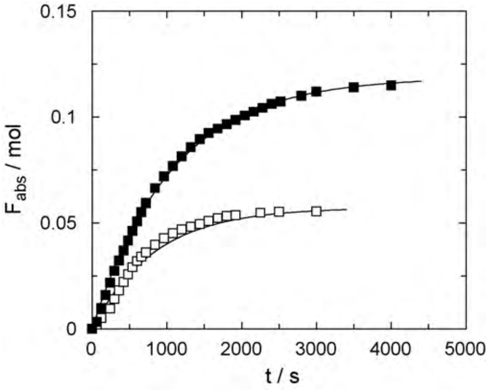

Fig. 1. Absorption kinetics of CO_2-glucosamine aqueous solution system. C_B = 0.1 mol L^{-1}, (□) pH 4.5 (without NaOH addition) and (■) pH 7.

The use of a BCR, in order to analyse carefully the experimental results, implies the knowledge of the interface area employed in the gas/liquid mass transfer. For this reason, the present paper includes the gas–liquid interfacial area determination under the operation conditions for the mass transfer experiments. The bubble diameter was measured using a photographic method based on taking images of the bubbles along the height of the column, from bottom to top. A Sony (DCR-PC330E) video camera was used to obtain the images. A minimum number of 100 well-defined bubbles along the bubble column were used to evaluate the size distribution of the bubbles in the different liquid phases employed (different concentrations of glucosamine and sodium hydroxide), as well as for each gas flow-rate that has been used. Image Tool v2.0 software package has been used to carry out the necessary measurements of the geometric characteristics of the bubbles. Photographs of different bubble column zones were analysed, taking into account the possible influence of the mass transfer, accompanied of chemical reaction, upon the bubble size along the liquid phase height.

The gas hold-up is necessary to determine the gas–liquid interfacial area, and this one has been calculated using the expansion method that uses the ungassed and gassed liquid height [10].

3. Results and discussion

The behaviour of glucosamine aqueous solutions in the absorption process of CO_2 has been analysed, at the beginning, in a global way. For that reason, as it was previously commented in the experimental section, several experimental conditions were used (liquid phase composition and gas flow-rate). Fig. 1 shows an example of the experimental data obtained of CO_2 capture experiments with glucosamine aqueous solutions. In that figure, the absorption kinetics of two liquid phases, consisting of an aqueous solution of glucosamine with the same concentration (0.1 mol L^{-1}), are compared. One solution has not been modified. However, a controlled quantity of NaOH has been added to another one in order to compensate partially the presence of HCl, present in the commercial product. When a part of the glucosamine molecules are free (for compensation with NaOH), the quantity of the CO_2 absorbed by the liquid phase increases significantly, which indicates that a process of CO_2 absorption is being taken place by means of a chemical reaction. This capture process of transferred gas to the liquid phase is carried out by a chemical reaction between the glucosamine, present in liquid phase, and the absorbed gas. This behaviour was detected in a previous study since, when the glucosamine from the supplier was used directly, a chemical reaction was not observed, due to the presence of HCl.

In relation to the influence of the gas flow-rate fed to the BCR, Fig. 2 shows the influence of this operation variable upon the CO_2

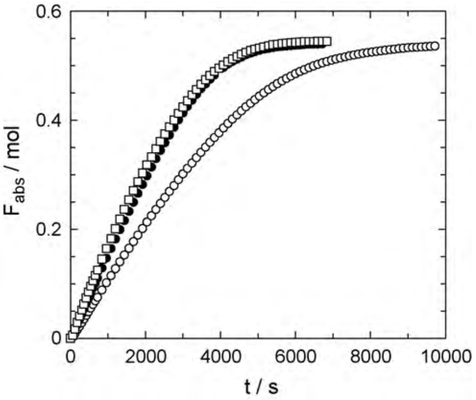

Fig. 2. Influence of gas flow-rate upon CO_2 absorption kinetic. $C_B = 0.3\ mol\ L^{-1}$, pH 9, (○) $Q_g = 18\ L\ h^{-1}$, (●) $Q_g = 30\ L\ h^{-1}$ and (□) $Q_g = 40\ L\ h^{-1}$.

capture kinetic when the other conditions remain constant. On the basis of the experimental data we can observe that, when a high value of gas flow-rate is fed to the gas–liquid contactor, the glucosamine present in the liquid phase is consumed at a high rate and then, the maximum CO_2 loading is reached in a shorter operation time. This behaviour is due to two effects produced by the increase of the gas flow-rate: (i) one of them is an increase in the gas–liquid interfacial area, since the increment in the gas volume fed to BCR implies the formation of a higher number of bubbles. An increase in the number of bubbles commonly produces an increase in the interfacial area and then, upon the gas mass transfer rate, and (ii) on the other hand, the increase in the number of bubbles increases the power supplied by the gas to the liquid phase when the bubbles ascend along the BCR (increasing the turbulence). This phenomenon produces an increase of the turbulence near the gas–liquid interface that produces an increase in the mass transfer rate.

Using the experimental data shown in Figs. 1 and 2 for CO_2 absorbed flow-rate along the operation time, under different operation conditions, the CO_2 loading has been analysed, taken into account the deprotonated amine concentration (free amine concentration). An example of the data obtained for this parameter (CO_2 loading) is shown in Fig. 3, and these data indicate that an increase in the amine concentration produces a decrease in the CO_2 loading. This behaviour is in agreement with previous studies using different amines as a reagent to capture acid gases [11–13]. This behaviour has been assigned to different causes in previous works: (i) an increase in the liquid phase viscosity due to an increase in the amine concentration, that produces a decrease in the diffusion coefficients, and (ii) aggregation processes between amine molecules when the concentration increases, being an inconvenient to the access of CO_2 molecules into the amino group in order to produce the reaction. The first cause abovementioned does not produce the decrease in the CO_2 loading, since a reduction in the gas diffusion must influence upon the mass transfer rate, but the CO_2 loading remains constant. Taking into account this hypothesis, the second cause seems to be the behaviour that produces the reduction in the CO_2 loading when the amine concentration increases in the liquid phase.

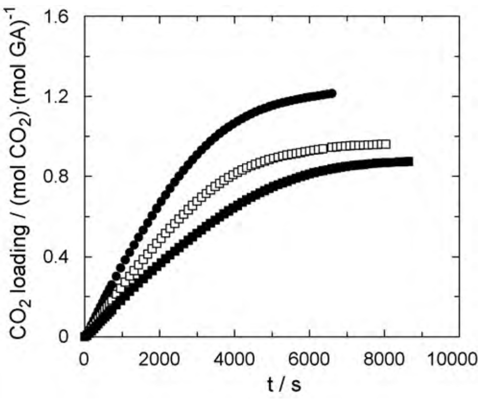

Fig. 3. CO_2 loading obtained for different glucosamine concentration solutions. $Q_g = 18\ L\ h^{-1}$, pH 8, (●) $C_B = 0.2\ mol\ L^{-1}$, (□) $C_B = 0.3\ mol\ L^{-1}$ and (■) $C_B = 0.4\ mol\ L^{-1}$.

Fig. 3 also shows that, in certain experiments, the CO_2 loading takes higher values than the unity. This behaviour is observed when the free amine concentration in the liquid phase is very low. Then, the CO_2 physically absorbed takes an important role in the ratio among the absorbed moles of CO_2 and the corresponding ones of free amines in the liquid phase.

With the aim of analysing carefully the influence of the interfacial area upon the previously commented behaviours, our research group has performed different studies about the influence of the operation conditions upon the gas–liquid interfacial area generated in the BCR. A photographic method has been employed (previously commented in the experimental section) because it has been recommended in different studies to determine this hydrodynamic parameter [14].

The bubble images we obtained show an ellipsoid shape and, for this reason, major (E) and minor (e) axes of the projected ellipsoid (in two dimensions) were determined. The diameter of the equivalent sphere (Eq. (1)) was taken as the representative bubble dimension.

$$d = \sqrt[3]{E^2 e} \qquad (1)$$

Different authors recommend using the Sauter mean diameter (d_{32}) [15], which can be calculated using Eq. (2).

$$d_{32} = \frac{\sum_i (n_i d_i^3)}{\sum_i (n_i d_i^2)} \qquad (2)$$

where n_i is the number of bubbles which have an equivalent diameter (d_i) [15].

The Sauter mean diameter (d_{32}) and the gas hold-up (ε) values allowed us the calculation of the specific interfacial area using Eq. (3) [16].

$$a = \frac{6\varepsilon}{d_{32}(1-\varepsilon)} \qquad (3)$$

In relation to the influence caused by the gas flow-rate upon the interfacial area, an increase in the bubble diameter and the gas hold-up in the bubble column are observed when gas flow-rate is increased. These two parameters have an opposite influence upon the interfacial area, but in Fig. 4 we can observe that the gas hold-up has a more important weight upon the gas–liquid interfacial area and then, an increase in this value is observed when gas flow-rate is increased.

About the influence of the glucosamine and sodium hydroxide concentration upon the interfacial area, previous studies indicate that an increase in the liquid phase viscosity [17] and the presence of electrolytes [18] could have certain influence upon the interfacial area, causing a decrease in this parameter. In this work, the variation of glucosamine concentration does not have any influence upon the gas–liquid interfacial area, with similar values to the determined ones for pure water. An increase in the glucosamine concentration in the liquid phase increases the viscosity [19], and this change in this physical property produces a negative effect upon THE interfacial area (previously commented). In this system, the change in viscosity with a glucosamine concentration is low and then, the influence of the glucosamine presence could be con-

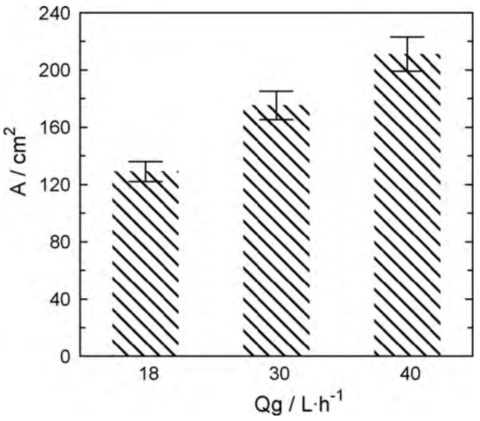

Fig. 4. Influence of CO_2 gas flow-rate upon gas–liquid interfacial area produced in the bubble column reactor.

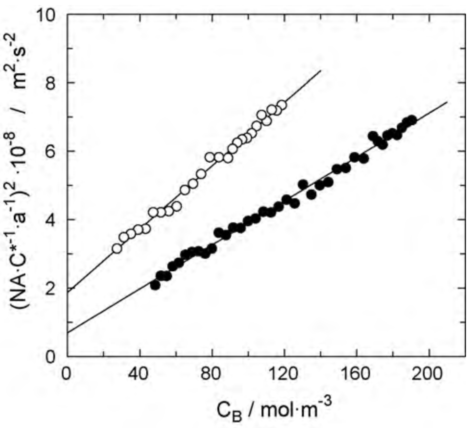

Fig. 5. Fits corresponding to Eq. (5) to determine the mass transfer coefficient data. $Q_g = 18\,\mathrm{L\,h^{-1}}$, $C_B = 0.2\,\mathrm{M}$, (○) pH 8 and (●) pH 10.

sidered negligible. The same behaviour has been obtained in the analysis of the influence of sodium hydroxide concentration upon the gas–liquid interfacial area. Taking also into account the influence of the chemical reaction upon the interfacial area, no influence was detected since the reaction regime is not instantaneous.

The reaction between the CO_2 and the glucosamine in an aqueous solution is moderately fast (*vide supra*), and this characteristic implies that the mass transfer process is the rate limiting reaction stage of the global process. Under certain conditions, the amine concentration at the gas/liquid interface could be the same as in the liquid bulk, and the reaction could be carried out completely at the interface. Eq. (4) must be used under these conditions.

$$N = C_A^* a \sqrt{D_A k_2 C_B^{\text{bulk}}} \tag{4}$$

where N, is the absorption rate of CO_2, C_A^* and D_A, the solubility and diffusivity of CO_2 in the aqueous phase, a, is the interface area, k_2, the rate constant for the reaction between CO_2 and hydroxyl ions and C_B^{bulk}, the glucosamine concentration in the bulk of the aqueous phase.

The use of this expression (Eq. (4)) needs the concentration of amine to remain practically constant throughout time [20], but if this condition is not satisfied, then, a part of the chemical reaction between the CO_2 and the glucosamine is carried out at the interface, and the other part in the liquid bulk. The surface renewal theory developed by Dankwerts contributed to the expression shown in Eq. (5).

$$N = C_A^* a \sqrt{D_A k_2 C_B^{\text{bulk}} + k_L^2} \tag{5}$$

The use of Eq. (5) to fit the experimental data to the mass transfer coefficient calculation implies the knowledge of the specific interfacial area value under the different operation conditions. The specific area determination could be set employing Eq. (4), but the application conditions are not satisfied under the present operation conditions. For this reason, another methodology must be applied for specific area determination (i.e. photographic method used in present work). Fig. 5 shows an example of how the experimental data fit employing Eq. (5) for different operation conditions. The linear fits shown in Fig. 5 let us calculate the value of the liquid side mass transfer coefficient by means of the intercept value.

Fig. 6 has been built to analyse the influence of the glucosamine initial concentration and for a different initial pH, using the values determined for the mass transfer coefficient for systems with different glucosamine concentration in the liquid phase. The obtained behaviour indicates that an increase in glucosamine concentration in the liquid phase produces a decrease in the mass transfer coefficient. The experiments shown in Fig. 6 have been performed at different initial pH and it implies that, taking into account that, when the glucosamine concentration increases, the free amine (deprotonated) increases as well. Though the free glucosamine increases, the mass transfer coefficient decreases. This behaviour is assigned to two causes with negative influence upon mass transfer rate: (i) an increase in liquid phase viscosity when the glucosamine concentration increases [19]. This physical property increases too by the addition of different quantities of sodium hydroxide to adjust the pH (see Fig. 7). The effect of viscosity upon the mass transfer in gas–liquid systems has been deeply studied observing a negative

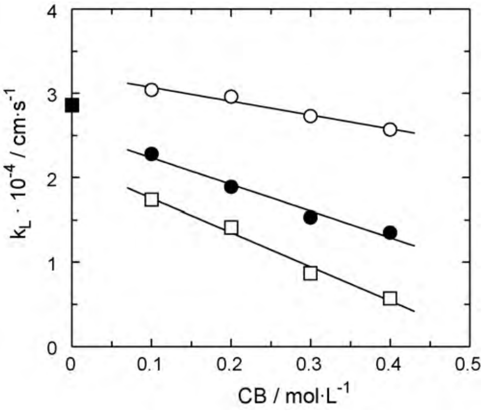

Fig. 6. Influence of glucosamine concentration and pH upon mass transfer coefficient data. $Q_g = 18\,\mathrm{L\,h^{-1}}$. (○) pH 7, (●) pH 8, (□) pH 9 and (■) water.

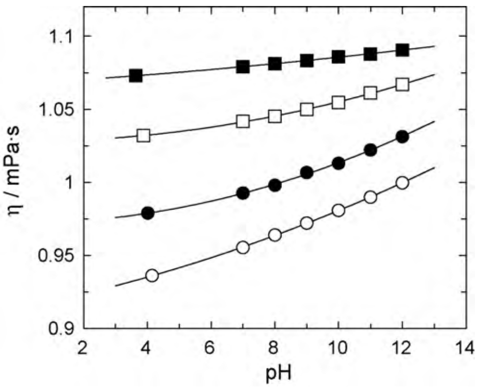

Fig. 7. Effect of glucosamine concentration and the addition of sodium hydroxide upon the dynamic viscosity value. (○) $C_B = 0.1\,\mathrm{mol\,L^{-1}}$, (●) $C_B = 0.2\,\mathrm{mol\,L^{-1}}$, (□) $C_B = 0.3\,\mathrm{mol\,L^{-1}}$ and (■) $C_B = 0.4\,\mathrm{mol\,L^{-1}}$.

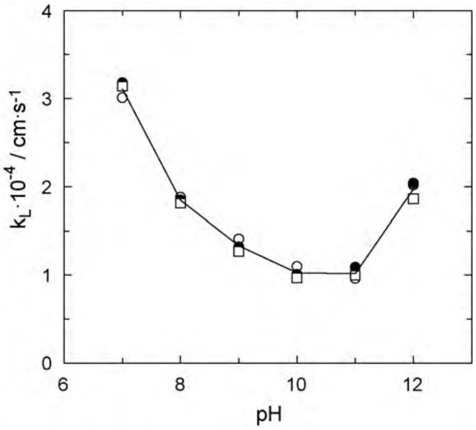

Fig. 8. Influence of pH and gas flow-rate upon mass transfer coefficient. $C_B = 0.2$ mol L^{-1}. (○) $Q_g = 18$ L h^{-1}, (●) $Q_g = 30$ L h^{-1} and (□) $Q_g = 40$ L h^{-1}. Solid line corresponds to the mass transfer coefficient medium value for all gas flow-rate.

effect in all the cases, producing a clear decrease in the mass transfer coefficient value [21,22], and (ii) the presence of electrolytes in the liquid phase, due to the hydrochloride acid (accompanying to glucosamine molecule) and the addition of sodium hydroxide, has a negative effect upon the mass transfer coefficient, as these substances produce an increase in liquid phase viscosity (see Fig. 7).

The influence of the liquid phase pH upon the mass transfer coefficient has also been analysed obtaining two different behaviours (Fig. 8). When the pH is increased, an increase in the free glucosamine concentration is obtained, but a reduction in the value of mass transfer coefficient is observed (until pH 10). And then, when the pH increases up to 10, a clear enhancement in the mass transfer coefficient is produced. The mass transfer coefficient reduction observed in Fig. 8, when the pH increases, is due to the previously commented effects. The addition of higher quantities of sodium hydroxide (and electrolytes) produces an increase in viscosity (Fig. 7) that produces a decrease in the mass transfer coefficient. When the addition of sodium hydroxide reaches the glucosamine concentration, free hydroxyl ions are present in the liquid phase and then, CO_2 molecules could react with both reagents (glucosamine and hydroxyl ions). As different studies have indicated, the reaction between CO_2 and hydroxyl ions is fast [23] and then, the mass transfer coefficient increases significantly with regard to the corresponding ones determined for systems with low pH values, without presence of hydroxyl ions in the aqueous solution. The existence of an instantaneous gas–liquid reaction regime (reaction between CO_2 and hydroxyl ions) increases the driving force in the system and then, the value of the mass transfer coefficient for high pH values increases too. But the existence of two reactions to capture CO_2 for pHs higher than 9 implies that, being the mass transfer coefficient under these operation conditions, it must not be taken into account to be compared to the previous ones, determined for a system where there were only one single reaction between the CO_2 and the glucosamine.

We can also observe in Fig. 8 the influence of the gas flow-rate fed to the bubble column reactor upon the mass transfer coefficient. For a bubbling equipment, this variable (gas flow-rate) could play an important effect in mixtures and stirring processes upon the liquid phase. The experimental results obtained from the system employed in the present work show that, under the used experimental conditions, differences between the mass transfer coefficient values for different gas flow-rates are not observed. This behaviour is in agreement with previous studies developed by our team [24,25] in similar experimental systems to the present work.

4. Conclusions

The present work has analysed the use of glucosamine aqueous solutions to capture CO_2. This capture has been done by means of absorption with a chemical reaction between this gas and the glucosamine present in the liquid phase, with a moderately fast chemical reaction regime.

The analysis of the influence of different operation variables and conditions upon the gas–liquid interfacial area generated in a bubble column reactor has been analysed. The results indicate that there is no influence of liquid phase composition (in the studied range) upon the interfacial area. However, the gas flow-rate fed to the bubble reactor has an important influence, causing an increase in this parameter due to an increase in the gas hold-up, as well.

The mass transfer coefficient determination has been developed under the different operation conditions, and the results for this parameter show that the gas flow-rate does not show any influence upon the mass transfer coefficient, but the liquid phase composition (glucosamine concentration) produces a decrease in the value of the mass transfer coefficient when the free glucosamine concentration increases in the liquid phase.

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