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Carbon dioxide absorption in glucosamine aqueous solutions

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Abstract

The present paper includes studies related to gas/liquid mass transfer process of carbon dioxide in aqueous solutions of glucose and glucosamine. These studies have been carried out employing a cylindrical bubble column, where the gas phase was fed through an only orifice. The absorption process has been characterized taken into account the influence of gas flow-rate and the liquid phase composition. The interfacial area was determined using a photographic method to calculate the value of the liquid side mass transfer coefficient. Liquid phase composition influences the value of the mass transfer coefficient due to the variation of solution viscosity. Eddy and slip models have been employed to correlate the experimental data of mass transfer coefficient.

Keywords: Absorption; Amine; Bubble column

1. Introduction

Industrial operations where one or more components of a gas phase are absorbed into a liquid phase are common in the chemical process industries. In many cases, this contact serves to achieve desired reactions among components in the two phases. Bubble reactors (columns and stirred vessels) have a wide range of applications in the chemical, biochemical and pharmaceutical industries [1], due to the effective contact of a gas with a liquid in order to carry out chemical or biochemical reactions. The gas transfer into a liquid is often the rate-limiting step [2].

These operation units are also used to prepare gaseous streams, to remove certain substances or to prevent the emission of dust, particles or acid gases. Other processes try to increase the concentration of several elements in the gas stream, like ozone or oxygen, for example. The volumetric mass transfer coefficient, $k_L \times a$, is one of the most important parameters that govern the performance of gas/liquid bubbling contactors. Therefore, extensive efforts have been directed to the estimation of the volumetric mass transfer coefficient. However, most of them are based on empirical analyses. Also, it is important to determine the influence of the operational variables (physical properties, stirring rate, gas flow-rate, etc.) on $k_L \times a$ to optimise the absorption process.

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 present research work has employed a gas that is an important environmental problem, due to the fact that it causes the acid deposition. Great quantities of carbon dioxide are emitted continuously by an important number of industrial processes. International resolutions have imposed a reduction in the emissions of several contaminant gases, such as carbon dioxide. For this reason, it is necessary to design units and processes to achieve this aim [6]. Two solutes' aqueous solutions have been employed as liquid phases in the present paper. One of them is the glucosamine, a molecule with an amino group with certain possibilities to react in the liquid phase with absorbed carbon dioxide. Glucose aqueous solutions have been employed to compare the results obtained and to compare them with the bibliographic data [7]. Glucose has been employed as a catal-

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Nomenclature

a	specific interfacial area [$\text{cm}^2\,\text{cm}^{-3}$]
C	CO_2 concentration [$\text{mol}\,\text{dm}^{-3}$]
C^*	CO_2 solubility [$\text{mol}\,\text{dm}^{-3}$]
d	equivalent sphere diameter [mm]
D	gas diffusivity [$\text{m}^2\,\text{s}^{-1}$]
d_{32}	Sauter mean diameter [mm]
e	minor axis of the projected ellipsoid [mm]
E	major axis of the projected ellipsoid [mm]
g	gravitational constant [$\text{m}\,\text{s}^{-2}$]
k_L	liquid mass transfer coefficient [$\text{cm}\,\text{s}^{-1}$]
n	number of bubbles
P	power dissipated [$\text{W}\,\text{m}^{-3}$]
Q_g	gas flow-rate [$\text{m}^3\,\text{s}^{-1}$]
t	operation time [s]
v_s	slip velocity [$\text{m}\,\text{s}^{-1}$]
Greek symbols	
ε	gas hold-up [mm]
ν	kinematic viscosity [$\text{mm}^2\,\text{s}^{-1}$]
ρ	liquid phase density [$\text{kg}\,\text{m}^{-3}$]

yser in homogeneous phase in absorption processes of carbon dioxide to aqueous solutions of buffers [8].

2. Theoretical fundamentals

Two different groups of models have been employed by different authors (eddy and slip velocity models). These models assume that the mass transfer mechanism corresponding to the liquid phase is due to the renewal of liquid elements in the interface surface.

Eddy models assume that the liquid renewal is due to small-scale eddies of the turbulent field [9]. Based in this, Lamont and Scott [10] have explained that the small scales of turbulent motion affect the mass transfer. As a result, the size of the gas bubble is not a very critical parameter to estimate k_L. Eq. (1) shows the expression formulated by Lamont and Scott.

$$k_L = C_1\left(\frac{P\nu}{\rho}\right)^{1/4}\left(\frac{D}{\nu}\right)^{1/2} \tag{1}$$

where P is the total specific power dissipated in the liquid phase, ν and ρ the kinematic viscosity and density of the liquid phase, D the gas diffusivity of gas phase in the liquid, and C_1 is an adjustable parameter.

On the other hand, slip velocity models assume a gross mean flow of fluid relative to the bubble (slip velocity) and a bubble surface mobility control of this renewal rather than the small-scale eddies of the turbulent field [9]. With regard to these kinds of models, the model proposed by Calderbank and Moo-Young [11] and a novel model developed by Alves and co-workers [12] have been employed in the last years. The first one divides the model taking bubbles size into account: small bubbles ($d < 1$ mm), which behave always as a rigid sphere, and large bubbles ($d > 2.5$ mm), which have a completely mobile surface. In our case, the systems studied are included in the large bubbles and the expression for the calculation of k_L for large bubbles is shown in Eq. (2).

$$k_L = 0.42\left(\frac{\Delta\rho\,\nu g}{\rho}\right)^{1/3}\left(\frac{D}{\nu}\right)^{1/2} \tag{2}$$

where $\Delta\rho$ is the difference between liquid and gas densities, ν and ρ the kinematic viscosity and density of the liquid phase, D the diffusivity of gas phase in the liquid, and g is the gravitational constant.

Alves and co-workers [12] have developed recently another variant for the slip velocity models. They have interpreted the mass transport from bubbles in terms of bubble contamination kinetics, where bubbles change from a mobile interface (when they enter the contactor) to a rigid condition due to the presence of surfactant molecules. For bubbles with completely mobile surface (large bubbles), k_L is given by Eq. (3), employing the Higbie's theory [13].

$$k_L = 1.13\sqrt{\frac{v_s}{d}}D^{1/2} \tag{3}$$

where d is the bubble diameter, D the gas diffusivity, and v_s is the bubble–liquid relative velocity (slip velocity). In bubble columns gas–liquid slip velocities are assumed to be close to single bubble terminal velocity, v_t [14]. Then, terminal velocity was determined for the contactor employed at different values solute concentration. An increase in the presence of solutes produces an increment in the viscosity of liquid phase that produces a decrease in the terminal velocity.

The Higbie's equation, employed by Alves et al. includes the slip velocity that is related to the power dissipated in systems without mechanical agitation. Several authors recommend Eq. (4) to determine the value of this power in bubble column reactors. An increase in this operation variable also produces an increase in the mass transfer velocity. Other authors have found a similar behaviour using different liquid phases [15]

$$P = \rho g v_s \tag{4}$$

3. Experimental

The studies of carbon dioxide mass transfer to liquid phases were carried out using the experimental set-up shown in Fig. 1. Our research group has employed this equipment [5] in previous works related to the absorption processes. The gas/liquid contactor used in these studies has been a cylindrical bubble column (internal diameter = 7 cm; height = 100 cm), made in methacrylate with a volume of 2.4 L.

The absorption processes has been carried out at 25 °C. The gas to be absorbed, carbon dioxide, was passed through two "humidifiers" at 25 °C to prepare the gas phase. This procedure removed other resistance to mass transport and allowed only the evaluation of the liquid phase resistance to the gas transfer. Water was placed into the "humidifiers". The gas flow-rate was measured and controlled with two mass flow controllers (5850 Brooks Instruments). The mass flow controllers employed in the

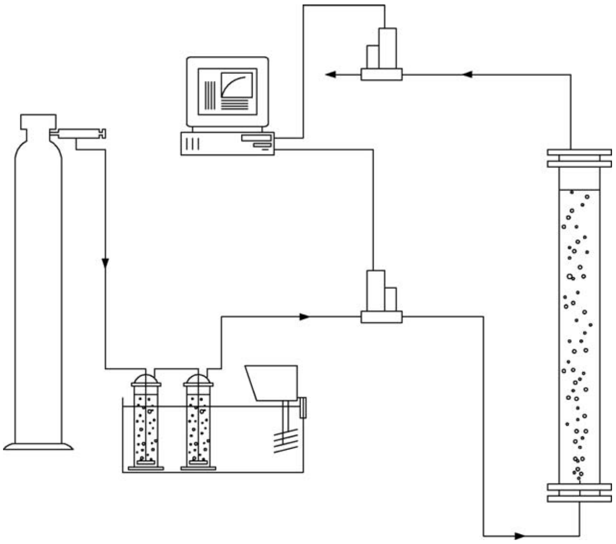

Fig. 1. Mass transfer equipment to carry out absorption experiments of carbon dioxide in a bubble column.

present study for the gas flow-rate and pressures were calibrated by the supplier.

The pressure drop was measured between the column's inlet and outlet, using a Testo 512 digital manometer. The operational regime was continuous in relation to the gas phase and batch as regards the absorbent liquid.

The present paper analyses the effect caused by the typical operational variables used in the present contact device (gas flow-rate and liquid phase composition) upon the kinetics of carbon dioxide mass transfer process to liquid phases. The liquid phases chosen in the present paper were aqueous solutions of glucose and glucosamine, being its mixtures in the range of 0–0.4 M. Three gas flow-rates were employed (18, 30 and 40 $\mathrm{L\,h^{-1}}$) to analyse the influence of this variable upon the gas/liquid absorption process.

The use of a BCR 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 determination of the interfacial area to the operation conditions under which the mass transfer experiments have been carried out. This study has been developed using a bubble column with the same sparger employed in mass transfer experiments, although this time with a square geometry (10 cm × 10 cm × 100 cm) to develop a physical method (photographic method) to determine the interface area and remove the influence of the cylindrical wall upon this measurement.

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-TRV9E) video camera was used to obtain the images. A minimum number of 80 well-defined bubbles along the bubble column were used to evaluate the size distribution of bubbles in the liquid phase employed, and for each gas flow-rate which has been used. We used the Image Tool v2.0 software to carry out the necessary measurements of the geometric characteristics of the bubbles.

Fitting parameters corresponding to the equations employed in the present paper have been obtained using commercial package software (Grafit v5.00), which allows to carry out a multivariable fit.

4. Results and discussion

The present communication summarizes the studies we have carried out in order to characterize the mass transfer process of carbon dioxide to aqueous solutions of glucosamine. We have also compared our results to similar studies which had employed glucose as solute, a system which has already been employed in literature.

Several studies developed by different research groups [5,15] have demonstrated the great importance of the physical properties values corresponding to the liquid phase in gas/liquid systems. For this reason, dealing with mass transfer studies requires the knowledge of certain physical properties which have an influence on mass transfer process. The physical properties used to explain the absorption kinetics in previous papers about gas/liquid absorption have been principally the viscosity (absolute or kinematic) and the surface tension. The last one has been employed in studies that involve the use of mass transfer processes containing hydrophobic molecules such as surfactants [12,16]. Surfactants have not been employed in the present paper as solutes, therefore we have used the value of pure water surface tension (72.1 $\mathrm{mN\,m^{-1}}$) [17].

Viscosity has been considered an important operation variable related to the liquid phase composition (solute concentration) that could affect significantly upon the absorption process. Our research group has published in a previous paper experimental data corresponding to aqueous solutions of glucosamine and glucose viscosity [18]. The values of this physical property for aqueous glucose solutions have also been compared to the previously published data [19]. In relation to the influence of solutes presence, it can be observed that an increase in the solutes concentration also provokes an increase in the viscosity, and the aqueous glucose solutions show bigger values than the solutions formed by glucosamine.

The first step of the study was evaluating the physical properties of interest to understand the mass transfer process that has been carried out. Then, the absorption operation will be analysed, using the experimental set-up described in Section 3.

On the basis of experimental data obtained for the evolution of carbon dioxide concentration in the bulk of the liquid phase, we can conclude that the absorption process in aqueous solutions of glucose and glucosamine shows a physical absorption type. If there is a chemical reaction in this system, the quantity of carbon dioxide absorbed will be higher when the glucosamine concentration in the liquid phase increases.

The non-existence of chemical reaction in the system employed in the present paper is due to the acid solutions formed by the glucosamine. The presence of an acid medium causes the inhibition of the reaction between absorbed carbon dioxide and amino group of glucosamine.

In this kind of absorption processes, working in a semi-continuously regime, Eq. (5) is used to determine the volumetric mass transfer coefficient obtained, using a gas phase mass balance.

$$\frac{\mathrm{d}C}{\mathrm{d}t} = K_{\mathrm{L}}a(C^{*} - C) \quad (5)$$

where $k_{\mathrm{L}} \times a$ is the volumetric mass transfer coefficient, and C^{*} and C are the solubility and carbon dioxide concentration, respectively.

Using the experimental data of gas concentration on the liquid phase, determined by the difference between the inflow and outflow rates, the volumetric mass transfer coefficients ($k_{\mathrm{L}} \times a$) have been calculated by means of linear regressions. The product of k_{L} and a, has been determined because the specific area of mass transfer in the bubbles does not remain constant: while the bubbles ascend through the liquid phase, the volume of the bubbles decreases.

Fig. 2 shows the influence of glucose and glucosamine concentration in the liquid phases upon the absorption process, being the influence based on the values of volumetric mass transfer coefficient. An increase in the solutes concentration causes a decrease in the product of mass transfer coefficient and specific area. Similar conclusions were obtained for glucose in previous studies, when glucose solutions were employed in the absence of chemical reaction in the liquid phase [7].

To obtain more information about the influence of solutes presence upon the global absorption process it is necessary to take into account the value of the specific area, so that we can draw a conclusion about the way the glucose and glucosamine affect on the carbon dioxide absorption in the BCR. The procedure employed to determine the bubble size distribution in the experimental contactor has been based on a photographic method, previously used by other research group [12,20].

The images we obtained of the bubbles in the liquids employed show an ellipsoid shape. 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. (6)) was taken as the representative bubble dimension.

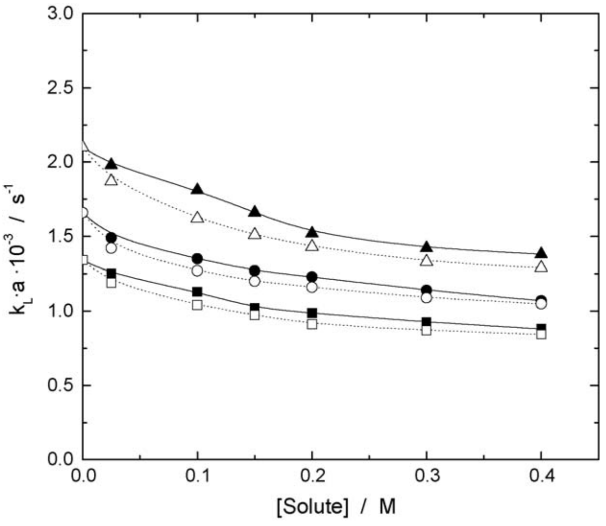

Fig. 2. Influence of glucose and glucosamine concentration upon the volumetric mass transfer coefficient. Glucose solutions: (□) $Q_g = 18\,\mathrm{L\,h^{-1}}$; (○) $Q_g = 30\,\mathrm{L\,h^{-1}}$; (△) $Q_g = 40\,\mathrm{L\,h^{-1}}$. Glucosamine solutions: (■) $Q_g = 18\,\mathrm{L\,h^{-1}}$; (●) $Q_g = 30\,\mathrm{L\,h^{-1}}$; (▲) $Q_g = 40\,\mathrm{L\,h^{-1}}$.

$$d = \sqrt[3]{E^{2}e} \quad (6)$$

Different authors recommend to use the Sauter mean diameter (d_{32}) [21], which is possible to determine using the data calculated for the equivalent diameter.

$$d_{32} = \frac{\sum_i n_i d_i^3}{\sum_i n_i d_i^2} \quad (7)$$

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

The Sauter mean diameter and the gas hold-up values allow the calculation of the specific interfacial area using Eq. (8) [22].

$$a = \frac{6\varepsilon}{d_{32}(1 - \varepsilon)} \quad (8)$$

The changes observed in the physico-chemical properties of the aqueous solutions employed in the present paper are not important. The influence of solute presence in the liquid phase upon the interfacial area has been considered negligible, so the value employed for this important parameter for the gas flow-rates used in this work was the equivalent to pure water.

Fig. 3 shows an example of the bubbles size distribution present in the BCR, based on the diameter of the equivalent sphere. In this figure, we can observe a Gaussian distribution type, and on the basis of these experimental results and the equations previously shown, we have calculated the Sauter diameter and the interfacial area between two phases.

The experimental data plotted in Fig. 3 also indicate that there is not a clear influence of the gas flow-rate fed to BCR upon the bubbles size distribution formed inside the contactor. Similar results have been obtained for all the flow-rates employed in the present study.

In relation to the value of the interfacial area (a basic parameter to analyse the mass transfer coefficient), its determination depends on the Sauter diameter and the gas hold-up caused by

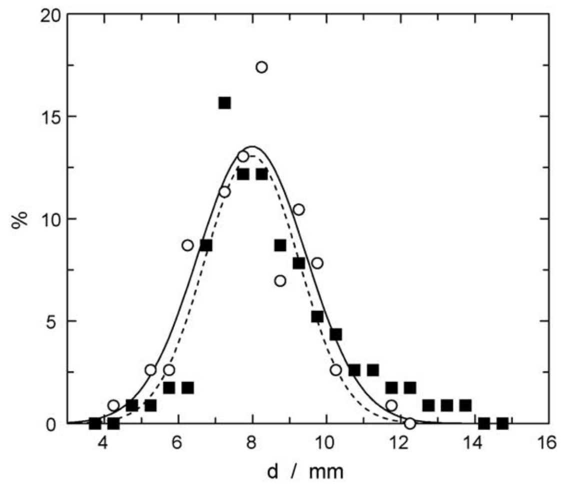

Fig. 3. Bubble size distribution obtained in the experimental contactor used in the present paper. (○) $Q_g = 18\,\mathrm{L\,h^{-1}}$; (■) $Q_g = 30\,\mathrm{L\,h^{-1}}$.

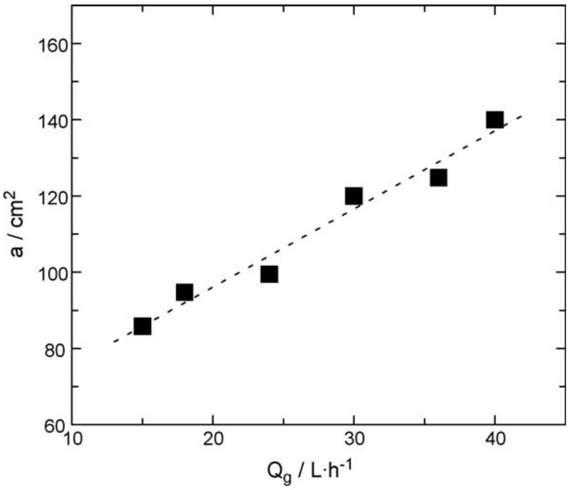

Fig. 4. Influence of gas flow-rate upon interfacial area values calculated using the photographic method.

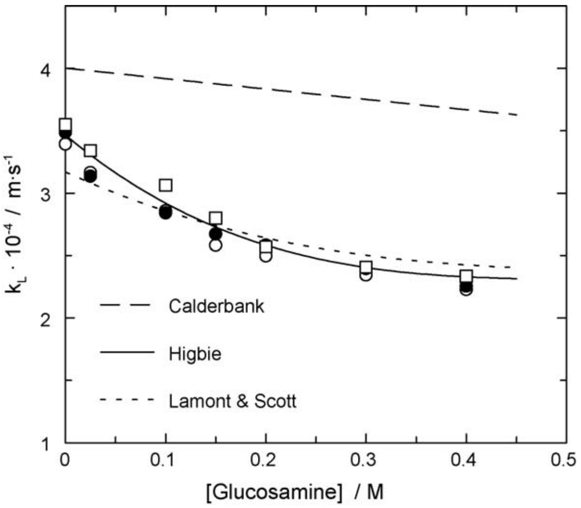

Fig. 5. Fit of different models to experimental data obtained in the present paper.

the gas flow-rate employed in the BCR, as it is shown in Eq. (8). Previously, we had concluded that the bubbles size is not affected by the gas flow-rate, but the other parameter (gas hold-up) shows a clear influence of this operational variable, and as a result of this, upon the value of the interfacial area (see Fig. 4). When the gas flow-rate increases, the value of the gas hold-up increases too, and the bubbles average diameter remains constant. To sum up, an increase in the gas hold-up implies the gas flow-rate to cause an increase in the gas/liquid mass transfer area.

We have analysed the behaviour of the mass transfer for this experimental system on the basis of the mass transfer coefficient, using the values obtained for interfacial area in this contactor and under the operation conditions employed in the present paper. The values obtained for this coefficient corresponding to glucose and glucosamine aqueous solutions indicate that when both solutes increase their concentration in the sorbent phase, the carbon dioxide mass transfer velocity to these liquid phases decreases continuously. This behaviour can be explained employing the physical characterization previously developed, where the viscosity increases when the solutes concentration increases in the liquid phase. This increment in the viscosity implies a higher resistance to the mass transfer along the interface between gas and liquid phases. This conclusion is consistent with the previous results obtained for other researchers and our own group [23,24].

We have analysed the influence of the gas phase flow-rate upon the values of mass transfer coefficient, and the trend observed is shown in Fig. 5. This figure concludes that the effect of the gas flow-rate is negligible, since all data corresponding to mass transfer coefficient at different gas flow-rates agree. This agreement implies that the gas flow-rate only has an influence upon the interfacial area, but not upon the mass transfer coefficient. The physical properties of the liquid phase (in the present case) upon the last parameter (k_L) produce the decrease observed in the value of the mass transfer coefficient, previously explained.

The value for the carbon dioxide diffusivity in the liquid phases employed in the present work has been considered equal to its diffusivity value in aqueous solutions of glucose. This is due to the similar values for viscosity in aqueous solutions of glucose and glucosamine (liquid phase viscosity is an important parameter with an influence on gas diffusivity). Vázquez and Álvarez [8] recommend an equation to calculate the value of this parameter in aqueous solutions of different substrates.

As regards the models shown in a previous section, Linek et al. [9] have commented that these kind of equations (slip and eddy models) supply different results in relation to the effect of dissipated power in liquid phase, because slip models do not take into account the influence of this variable.

If we use a kind of contactor like the previous one described, the dissipated power is due only to the expansion of the entering gas phase, and this produces an increase in the mass transfer coefficient when the gas flow-rate fed increases too.

Fig. 5 also shows an example of the results obtained when comparing experimental values with calculated ones, using the models previously commented. This figure shows that the Higbie's equation supply excellent results and allows calculate liquid side mass transfer coefficients values, while Calderbank and Moo-Young's one allows the calculation of high values in relation with the experimental data. The model developed by Lamont and Scott shows a good behaviour although there is a certain deviation. It is necessary to take into account that this last model includes one fitting parameter, and this characteristic implies certain flexibility for the model, but Higbie's equation is totally predictive. The parameter C_1 has a low value in comparison with the previous ones obtained by other authors [9]. Higbie's equation contributes better results that Lamont and Scott model due that the bubble column employed in the present paper produce low turbulence in the liquid phase because the gas is introduced in the contactor through an only glass capillary.

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