Effect of Solid Loading on Carbon Dioxide Absorption in Bubble Column

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Abstract

In the present work experiments were conducted to study the effect of solid loading (1.5 and 9 vol.%) on the enhancement of carbon dioxide absorption in bubble column at various volumetric gas flow rate (0.75, 1 and 1.5 m³/h) and absorbent concentration (caustic soda) (0.1, 0.5 and 1 M). Activated carbon and alumina oxide (Al₂O₃) are used as solid particles. The Danckwerts method was used to calculate interfacial area and individual mass transfer coefficients during absorption of carbon dioxide in a bubble column. The results show that the absorption rate was increased with increasing volumetric gas flow rate, caustic soda concentration and solid loading. Mass transfer coefficient and interfacial area were increased with increasing volumetric gas flow rate, and solid loading.

Keywords: Carbon dioxide absorption, bubble column, Danckwerts method, mass transfer coefficients and interfacial area.

1. Introduction

Slurry bubble columns are intensively used as a multiphase contactors in the chemical, biochemical and petrochemical industries where heterogeneous gas-liquid or gas-solid reactions take place, particularly, in which the liquid phase controls the mass transfer process due to the relation with solubility of gases[1, 2]. Important applications of three phase bubble column are in hydrogenation, oxidation, and waste water treatment and in biochemical applications [2].

The rate of acid gas absorption such as carbon dioxide absorption in a gas-liquid or gas-liquid-solid contactor may be enhanced considerably by the presence of particles in the liquid phase. To be effective, the particles have to be considerably smaller than the gas-liquid film thickness and need to have a high affinity for the component to be transferred. Enhancement of the gas absorption rates due to the presence of small particles is explained by the so-called grazing or shuttle mechanism [3-6].

Sharma and Mashelkar (1968) [7] were the first to report an increase in the mass transfer in a bubble column by small particles. Similar effects were found later by Wimmers and Fortuin (1988) [8], Beennackers and van swaaij (1993) [6] and Marius et al (2007) [9].


Vandu and Krishnal (2004) [12] observed that addition of solids and high solid concentrations caused reduced values of mass transfer coefficients due to increased large bubble size.

Sumin et al (2007) [13], studied the absorption of carbon dioxide in carbonate solution (K₂CO₃) in the presence of activated carbon particles and found that the absorption rate enhanced significantly, and the maximum enhancement factor was 3.7.

The present work aimed to study the absorption rate of carbon dioxide in caustic soda (NaOH) solution 0.1, 0.5 and 1M at different volumetric gas flow rate 0.75, 1 and 1.5 m³/h and carbon dioxide concentration of 10% by volume.
with and without solid particles (activated carbon and alumina oxide (Al$_2$O$_3$) (1, 5 and 9 vol.%), keeping other variables constant as temperature of (20±2)$^\circ$C, and atmospheric pressure.

2. Experimental Work

Experiments of absorption of carbon dioxide from gaseous mixture (10% carbon dioxide - air) has been carried out by using aqueous solution of (0.1, 0.5 and 1M) NaOH. This has been performed in a conventional slurry glass cylindrical bubble column of 7.5 cm inside diameter, 100 cm height over a wide range of gas flow rate of 0.75, 1 and 1.5 m$^3$/h (as velocity 0.0117-0.0235 m/sec), solid loading (1, 5 and 9vol.%) and different types of solid particles (Activated carbon, Alumina oxide) to study the effect of these parameters on the fractional conversion, absorption rate and mass transfer coefficient of carbon dioxide. Schematic diagram of experimental set up is shown in Figure (1). Table (1) shows the characteristics properties of Activated carbon, Alumina oxide.

Perforated plate sparger was used as a gas distributor 104 hole of 1mm diameter and placed between the column and distributor chamber which has a drain at the bottom and gas inlet at the side.

![Experimental Setup Diagram](image-url)
Table 1, The Characteristics Properties of Activated Carbon, Alumina Oxide.

<table>
<thead>
<tr>
<th></th>
<th>Activated Carbon (A.C)</th>
<th>Alumina oxide (Al₂O₃)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface area (m²/g)</td>
<td>1122</td>
<td>300</td>
</tr>
<tr>
<td>Bulk density (g/m³)</td>
<td>0.44</td>
<td>0.56</td>
</tr>
<tr>
<td>Porosity (-)</td>
<td>0.46</td>
<td>0.51</td>
</tr>
<tr>
<td>Particle diameter (m)</td>
<td>1.1*10⁻³</td>
<td>1.5*10⁻³</td>
</tr>
<tr>
<td>Min. fluidizing velocity (m/s)</td>
<td>0.000329</td>
<td>0.00537</td>
</tr>
<tr>
<td>Particle terminal velocity (m/s)</td>
<td>0.000309</td>
<td>0.0521</td>
</tr>
</tbody>
</table>

3. Procedure of Experiment

In all experiments the volume of liquid has been set constant and equal to 1.5 liter, 30 cm height above the sparger. The gas flow of carbon dioxide and air were measured by two calibrated rotameters separately, then entered the bottom of the bubble column. The samples were taken from the side of the bubble column every 3 min. The temperature was measured periodically by a thermocouple until the end of the experiment. All experiments were performed at constant temperature of (20±2)°C and atmospheric pressure.

3.1. Chemical Reactions

When carbon dioxide is absorbed into aqueous sodium hydroxide solutions, the following two reactions should be considered [14, 15]:

\[
\text{CO}_2 + 2\text{OH}^- \rightarrow \text{HCO}_3^- + \text{H}_2\text{O} \quad \ldots(3)
\]

\[
\text{CO}_2 + \text{OH}^- \rightarrow \text{HCO}_3^- \quad \ldots(1)
\]

\[
\text{HCO}_3^- + \text{OH}^- \rightarrow \text{CO}_3^{2-} + \text{H}_2\text{O} \quad \ldots(2)
\]

At 30°C and at infinite dilution reaction (1) is practically considered irreversible and second order, i.e. first order with respect to both carbon dioxide and OH ions. Reaction (2) is a proton transfer reaction and has a very much higher rate constant than reaction (1), thus this reaction can be regarded as an instantaneous reversible reaction[14, 15].

In strong hydroxide solutions, the equilibrium concentration of HCO₃⁻ ions can be neglected and the overall reaction is:

\[
\text{CO}_2 + 2\text{OH}^- = \text{CO}_3^{2-} + \text{H}_2\text{O} \quad \ldots(3)
\]

3.2. Physico - Chemical Properties

1. Diffusivity \( (D_{\text{CO}_2,\text{NaOH}}) \) of carbon dioxide in aqueous caustic soda solution was estimated as follows[16 ]:-

\[
D_A = 1.833*10^{-6}T - 4.717*10^{-4} - 1.042*10^{-5}N \quad \ldots(4)
\]

2. Reaction rate constant \( (k_2) \) was estimated as follows[9, 17]:-

\[
\log k_2 = 11.985 - \frac{2895}{T} + 0.221I_c - 0.016I_c^2 \quad \ldots(5)
\]

Where \( I_c \) is the ionic strength and can be estimated as follows:

\[
I_c = \frac{1}{2} \sum Z_i^2 C_i \quad \ldots(6a)
\]

\[
I_c = \frac{1}{2} \left[ \text{OH}^- + 2 \times \text{CO}_3^- \right] \quad \ldots(6b)
\]

Let \( \text{OH}^- = b_1 \) and \( \text{CO}_3^- = b_2 \)

Then

\[
m = b_1 + 2b_2
\]

\[
my = b_2
\]

\[
\therefore I_c = \frac{1}{2} m \quad \ldots(6c)
\]

3. Solubility of carbon dioxide in the liquid phase was calculated using the Henry's law. Henry's law constant for \( \text{CO}_2-\text{Na}_2\text{CO}_3 \) system was determined as follows[10]:-

\[
\log H^0 = \sum I_c h_i \quad \ldots(7)
\]

Where \( h_i = h_- + h_+ + h_g \)

\( h_- \) for \( \text{OH}^- = 0.061 \text{ liter/g ion} \)

\( h_+ \) for \( \text{Na}^+ = 0.094 \text{ liter/g ion} \)

\( h_g \) for \( \text{CO}_2 = 0.017 \text{ liter/g ion} \)

Where \( H^0 \) is Henry's law constant for \( \text{CO}_2 \) in water and can be obtained as follows[13]:-

\[
\log H^0 = 9.1229 - 5.9044*10^{-2}T + 7.8857*10^{-5}T^2 
\]

\[
\ldots(8)
\]

4. Causticsod conversion % = \[\frac{\text{[NaOH] reacted}}{\text{[NaOH] input}}\]... (9)
3.3. Interfacial Area A and Individual Mass Transfer Coefficient \( k_L \)

Danckwerts method was used to calculate the interfacial area (a) and the individual mass transfer coefficient (\( k_L \)) as follows [18, 19]:

\[
N = C_{CO_2}^o a \sqrt{D_A k_2 C_B^\text{bulk} + k_L^2} \tag{10}
\]

\[
\left[ \frac{N}{C_{CO_2}^o} \right]^2 = a^2 D_A k_2 C_B^\text{bulk} + k_L^2 \tag{11}
\]

Interfacial area \( a \) and individual mass transfer coefficient \( k_L \) are obtained by plotting \( \left[ \frac{N}{C_{CO_2}^o} \right]^2 \) vs. \( D_A k_2 C_B^\text{bulk} \), the relation is straight line of slope \( a^2 \) and intercept \( [k_L, a]^2 \).

Figure (2) shows Danckwerts plot for estimated interfacial area (a) and mass transfer coefficient (\( k_L \)). This method is used when the concentration of absorbent is not constant with time.

Fig.2. Danckwerts Plot: a) Free Solid Concentration; b) 1% vol. \( \text{Al}_2\text{O}_3 \) Solid Concentration at Different Volumetric Gas Flow Rate.

Fig.3. Caustic Soda Concentration vs. Time at 0.75 m\(^3\)/h Volumetric Gas Flow Rate.
This implies that a part of the chemical reaction between carbon dioxide and hydroxyl ions is carried out at interface and the other part in the bulk of the liquid. Therefore; the surface renewal theory developed by Danckwerts is satisfied for estimating interfacial area $a$ and mass transfer coefficient $k_{La}$[19].

Figure (3) shows the caustic soda concentration during carbon dioxide absorption at a given condition. It can be seen that the caustic soda decreases with time.

### 4. Results and Discussion

#### 4.1. Influence of Superficial Gas Velocity

Figures (4 to 6) show the effect of volumetric gas flow rate on absorption rate, mass transfer coefficient and interfacial area. It can be seen that at a given solid loading the absorption rate increased with increasing volumetric gas flow rate. This is attributed to the fact that the rate of breakup of bubble increased. In addition, higher superficial gas velocity gives smaller bubbles. The smaller bubble of lower rising velocity leads to form large residence time and consequently higher gas – liquid interfacial, mass transfer coefficient and absorption rate. These results are in agreement with previous work [20, 21].

**Fig.4.** Absorption Rate vs. Total Volumetric Gas Flow Rate ($CO_2$+air): a) A.C Solid Loading , b) $AL_2O_3$ Solid Loading.

**Fig.5.** Interfacial Area vs. Total Volumetric Gas Flow Rate ($CO_2$+air).
Fig. 6. Mass Transfer Coefficients vs. Total Volumetric Gas Flow Rate (CO$_2$+air).

Fig. 7. Absorption Rate vs. Total Volumetric Gas Flow Rate (CO$_2$+air) As a Function of Absorbent Concentration and Solid Loading: a) A.C Solid Loading; b) AL$_2$O$_3$ Solid Loading.
4.2. Influence of Absorbent Concentration

Figure (7) shows the effect of absorbent concentration on absorption rate at a given volumetric gas flow rate and solid loading. The results show that the absorption rate increases with increasing absorbent concentration. Increasing of absorbent concentration will cause the zone of reaction approach the gas-liquid interface rapidly which leads to increase the driving force (ΔC) and decreases the thickness of the liquid film through the solute due to increase OH\(^-\) concentration in solution that reacts with carbon dioxide which leads to increase the reaction controlling step (equation 2). These results are in agreement with previous work [22, 23, 24].

4.3. Influence of Solid Loading

Figures (4 to 7) show the effect of solid loading on absorption rate, mass transfer coefficient and interfacial area. It can be seen that the absorption rate, mass transfer coefficient and interfacial area increase with increasing solid loading. Enhancement of the gas absorption rates due to the presence of small particles is explained by the so-called grazing or shuttle mechanism. It is assumed that the particles travel between the stagnant liquid mass transfer layer and the bulk of the liquid. Near the interface, the adsorptive particles are loaded with solute and the solute concentration in the liquid mass transfer layer decreases. The concentration gradient of the solute in the mass transfer layer increases leading to enhance gas absorption. After a certain time in the liquid side mass transfer layer, the particles returns to the bulk of the liquid where the gas-phase component is desorbed and the particle regenerated [3-6, 13, 25]. Due to the hydrophobic properties of active carbon, the concentration of particles in the mass transfer layer is much higher than in the bulk of the suspension, leading to higher absorption rates and mass transfer coefficients [25]. Also, the surface area of activated carbon was higher than the alumina. This leads to that the absorption rate, mass transfer coefficient and interfacial area with activated carbon particle is higher than that when alumina particle was loaded.

5. Conclusions

The following points are concluded from the present work:-

β The absorption rate increased with increasing volumetric gas flow rate and absorbent concentration within the conditions used.

β The absorption rate, mass transfer coefficient and interfacial area increase with increasing solid loading according to grazing or shuttle mechanism.

β Danckwerts method was used to calculate interfacial area (a) and individual mass transfer coefficient (k_L).

### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>m(^2)/m(^3)</td>
<td>Interfacial area</td>
</tr>
<tr>
<td>C_B</td>
<td>kmol/m(^3)</td>
<td>Concentration of liquid reactant (B) in the bulk</td>
</tr>
<tr>
<td>C_CO(_2)</td>
<td>kmol/m(^3)</td>
<td>Concentration of carbon dioxide at equilibrium</td>
</tr>
<tr>
<td>C_i</td>
<td>kmol/m(^3)</td>
<td>Concentration of ions</td>
</tr>
<tr>
<td>D_A</td>
<td>m(^2)/s</td>
<td>Diffusivity of carbon dioxide in caustic soda solution</td>
</tr>
<tr>
<td>H</td>
<td>atm.m(^3)/kmol</td>
<td>Henry's constant</td>
</tr>
<tr>
<td>H(^o)</td>
<td>atm.m(^3)/kmol</td>
<td>Henry's constant in pure water</td>
</tr>
<tr>
<td>h_i</td>
<td>L/g ion</td>
<td>Parameters of equation 7 of h, h, and h_g respectively, h_f = h + h + h_g</td>
</tr>
<tr>
<td>h_f</td>
<td>L/g ion</td>
<td>Parameter of cation</td>
</tr>
<tr>
<td>h_g</td>
<td>L/g ion</td>
<td>Parameter of anion</td>
</tr>
<tr>
<td>I_c</td>
<td>m(^3)/kmol</td>
<td>Ionic strength</td>
</tr>
<tr>
<td>k_{L,a}</td>
<td>1/ s</td>
<td>mass transfer coefficient</td>
</tr>
<tr>
<td>k_L</td>
<td>m/s</td>
<td>Liquid side mass transfer coefficient</td>
</tr>
<tr>
<td>k_2</td>
<td>m(^3)/kmol.s</td>
<td>Reaction rate constant</td>
</tr>
<tr>
<td>N</td>
<td>kmol/m(^3).min.</td>
<td>Absorption rate</td>
</tr>
<tr>
<td>m</td>
<td>kmol/m(^3)</td>
<td>Molarity</td>
</tr>
<tr>
<td>T</td>
<td>K</td>
<td>Temperature</td>
</tr>
<tr>
<td>y</td>
<td>( - )</td>
<td>Fractional conversion</td>
</tr>
<tr>
<td>Z_i</td>
<td>( - )</td>
<td>Valance of ion</td>
</tr>
</tbody>
</table>

6. References


دراسة تأثير تحميل مادة الصلب على عملية امتصاص غاز ثنائي أوكسيد الكربون في العمود الفقاعي

علياء خضير مجيد
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الخلاصة

هذ البحت تقدم دراسة تأثير تحميل الصلب (1.5% نسبة حممية) على عملية امتصاص غاز ثنائي أوكسيد الكربون في العموم الفقاعي في معدل التشفق الحجمي لغاز (0.015 مماسفة) وتركيز المادة الماصة (أوكسيد الصوديوم) (0.5 ملمول/م3). تم استعمل الكربون المنشط وأوكسيد الالمنيوم كمواد صلبة. طريقة دانوهرزت استعملت لحساب الماصة البيئية ومعامل انتقال الكثافة أثناء عملية امتصاص غاز ثنائي أوكسيد الكربون في العمود الفقاعي.

أظهرت النتائج أن معدل الامتصاص يزداد بزيادة معدل التشفق الحجمي للغاز وتركيز المادة الماصة (أوكسيد الصوديوم) ومقدار تحميل الصلب. كما تبين

أن معامل انتقال الكثافة والماصة البيئية يزدادان بزيادة معدل التشفق الحجمي للغاز ومقدار تحميل الصلب.