Dynamic coupling of mass transfer and chemical reaction for Taylor flow along a serpentine microchannel

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ABSTRACT

The characteristics of multiphase dynamic mass transfer play a big role in gaining understanding and controllability of gas-liquid two-phase chemical absorption processes. In the present work, dynamic coupling of mass transfer and chemical reaction for Taylor flow in a serpentine microchannel was investigated by an online method. The dynamic changes of gas bubbles, key mass transfer parameters (i.e., $k_L$, $k_a$, and $a$) and the enhancement factor were measured and calculated, where three changing stages were found under different operating conditions of $Q_G$ (2-5.5 mL), $Q_L$ (1-2.5 mL), $W_{MEA}$ (1-3 wt%). Based on the experimental results, a modified van Baten’s model was proposed to predict mass transfer performance for both CO$_2$-H$_2$O and CO$_2$-MEA systems, and the correlation coefficients were 0.95 and 0.96, respectively. The correlations also suggested the net physical mass transfer performance in CO$_2$-MEA system could be reasonably separated from the chemical absorption process.

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1. INTRODUCTION

Among the current technologies for the removal of CO₂, chemical absorption with monoethanolamine (MEA) solution has been the most widely used one. Although great efforts have been made to develop gas-liquid contactors for higher process efficiency, the amount of absorbent used, the energy consumption and the equipment investment are still far from satisfactory. Generally, the efficiency of gas-liquid contactors is determined by gas-liquid mass transfer performance, where the dynamic change (e.g., formation, growth, shrinkage, collapse and coalescence) of bubbles plays an important role. Due to the complex hydrodynamics of gas bubble populations, there still remains challenges to measure the instantaneous volumetric mass transfer coefficient with high spatiotemporal precision.

In recent years, microchannel contactors have been developed into powerful tools for measuring the volumetric mass transfer coefficient, owing to its small characteristic dimension, large surface-to-volume ratio and controllable gas-liquid flow patterns. Berčič and Pintar put forward an empirical correlation based on a unit cell model for predicting the liquid side volumetric mass transfer coefficient ($k_La$) in methane-water system under Taylor flow; they found that ($k_La$) was mostly determined by the liquid slug length and velocity. van Baten and Krishna investigated mass transfer from Taylor bubbles to the liquid phase by computational fluid dynamics, and found that $k_La$ was determined by the sum of the contributions of the two bubble caps and the film surrounding the bubble. Yue et al. examined CO₂ absorption in water and NaHCO₃/Na₂CO₃ buffer solution, and proposed an improved
empirical correlation for determining $k_La$.\textsuperscript{17,18} Li et al. studied the effects of flow rates on $k_La$ in Taylor flow with CO\textsubscript{2}-MEA system.\textsuperscript{19} Recently, Chu et al. investigated experimentally the mass transfer characteristics of CO\textsubscript{2} absorption into aqueous solution of ionic liquid in microchannel and proposed a dimensionless correlation to predicting $k_La$.\textsuperscript{20} In these studies, $k_La$ was generally assumed to be constant across the entire reactor under given operating conditions. However, the dynamic shrinkage of Taylor bubble along the microchannel was disregarded.

To address this issue, Cubaud et al. measured the evolution of CO\textsubscript{2} bubble size with physical solvents in long serpentine microchannels by tracking individual bubbles along the flow direction to establish the functional relationship between bubble size and velocity.\textsuperscript{21,22} Abolhasani et al. developed an automated microfluidic platform to determine CO\textsubscript{2} solubility, $k_La$, as well as the reducing rate of Taylor bubbles, which could be translated into mass transfer performance.\textsuperscript{23} Similar effort was made by Tan et al. to measure the changes of gas bubble volume versus flowing distance, which was beneficial to further estimation of $k_La$.\textsuperscript{24} By developing a unit cell model, Yao et al. built a relation between $k_La$ and the reducing rate of CO\textsubscript{2} bubbles, realizing direct measurement of $k_La$ through monitoring CO\textsubscript{2} bubbles size.\textsuperscript{25} Zhang et al. used a similar model to characterize gas-liquid mass transfer for CO\textsubscript{2}-H\textsubscript{2}O system.\textsuperscript{14} Although these investigations shed considerable light on the dynamic characteristics of the gas-liquid mass transfer process, these findings were limited to physical absorption and differed from chemical absorption processes.

There have been efforts made in examining the mass transfer of CO\textsubscript{2} chemical
absorption into aqueous MEA solutions for Taylor flow along microchannels.\textsuperscript{19,26,27} Zhu et al. found the bubble length decreased exponentially along the microchannel due to CO\textsubscript{2} reactive absorption, and a turning point existed in the flowrate ratio of gas and liquid phases for different MEA concentrations\textsuperscript{26}. A correlation for predicting $k_La$ was also proposed by integrating the enhancement factor of chemical reaction\textsuperscript{27}.

However, these studies were still limited to treating $k_La$ as constant. Moreover, the dynamic change of mass transfer enhancement due to chemical reaction was not considered.

Based on the above analysis, the aim of the present work was to quantitatively characterize the dynamic coupling of mass transfer and chemical reaction under Taylor flow of the CO\textsubscript{2}-MEA system along a microchannel. An online imaging technique with a high-speed camera and a stereo microscope was employed to measure the evolution of the geometry and movement of gas bubbles. The dynamic changes were quantified in terms of $k_La$, liquid phase mass transfer coefficient ($k_L$), specific interfacial area ($a$) and enhancement factor ($E$). A theoretical model was established for determining $E$. In order to predict the local mass transfer performance, a model was proposed for both CO\textsubscript{2}-H\textsubscript{2}O and CO\textsubscript{2}-MEA systems. These findings would provide strong foundation for designing and optimizing gas-liquid microchannel contactors towards high process performance.

2. EXPERIMENTAL SECTION

The experimental setup is shown schematically in Figure 1(a). Two high pressure syringe pumps (LSP01-1BH, Longer) and two stainless steel syringes (50 mL, Longer)
were used to deliver gas and liquid, respectively. The gas and liquid flow rates were calibrated by the weighting method and an automated electronic soap film flowmeter (GL-101B, 0.1-100 mL/min, accuracy 1%, Beijing Beifen Sanpu, China), respectively. CO₂ (99.999 vol%) and aqueous MEA solution (analytical grade, Sinopharm Chemical Reagent Co., Ltd.) were used as the working fluids. The operating temperature was set at 25 °C. The pressure drop across the flow channel was measured by a pressure transducer (DMP305X, 0~50kPa, accuracy 0.075%, LEEG, China). Across the whole length (200 mm) of the microchannel, the overall pressure drop was found to be minimal (< 6.6 kPa) suggesting that the evolution of the local pressure along the microchannel was insignificant. Therefore, a linear correlation was used to represent $P_{CO2}$ as a function $X$ in this work. The concentrations of the aqueous MEA solution were set at four levels, i.e., 1.0, 1.5, 2.0 and 3.0 wt%. The main reason for choosing the MEA concentration range (1.0-3.0 wt%) in this study was to enable practically measuring the gas-liquid mass transfer performance, although it was relatively lower than that in industrial applications. The dynamic changes of CO₂ bubbles were recorded by a high-speed camera (Phantom R311, 3200 fps) mounted on a stereo microscope (Olympus SZX 16). The resolution and exposure time were 1280 × 800 pixels and 490 μs, respectively.
**Figure 1.** T-junction microchannel and gas-liquid mass transfer system (a) Schematic of the experimental setup; (b) T-junction microchannel; (c) Typical captured image.

Figure 1(b) illustrates the layout design of the microchannel with a T-junction followed by a long serpentine microchannel. The design parameters (width, depth, length, and radius) for different sections of the microchannel are summarized in Table 1. The microchannels were fabricated on PMMA plates (A grade, 92% of light transmittance, ShenZhen HuiLi Acrylic Products Co., Ltd.) by precision milling and sealed by thermal sealing. Figure 1(c) shows a snapshot captured by a high-speed camera focused on the main area covering all 12 serpentine microchannels. 1000 images were taken from each set of experiment, and the changes in bubble length and velocity along the microchannel were analyzed by Matlab image analysis program.

**Table 1.** Geometries (mm) of microchannel in Figure 1(b)

<table>
<thead>
<tr>
<th>Width/w</th>
<th>Depth/h</th>
<th>L₁</th>
<th>L₂</th>
<th>L₃</th>
<th>R₁</th>
<th>R₂</th>
<th>R₃</th>
<th>R₄</th>
<th>Total length/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.6</td>
<td>0.6</td>
<td>9.43</td>
<td>14.28</td>
<td>13.96</td>
<td>1.0</td>
<td>1.5</td>
<td>0.2</td>
<td>0.8</td>
<td>181.13</td>
</tr>
</tbody>
</table>

### 3. PROCESS ANALYSIS AND MODELING

#### 3.1. Reaction Mechanisms and Mass Transfer Process Analysis

The chemical reaction of CO₂ with aqueous MEA solution can be interpreted by either the zwitterion mechanism\(^{28,29}\) or the termolecular mechanism\(^{30,31}\). The former was more commonly used.\(^{32,33}\)

Zwitterion formation:

\[
\text{CO}_2 + \text{RNH}_2 \xrightarrow{k_1} \text{RNH}_2^+\text{COO}^- \quad (1)
\]

Zwitterion deprotonation:

\[
\text{RNH}_2^+\text{COO}^- + \text{RNH}_2 \xrightarrow{k_2} \text{RNHCOO}^- + \text{RNH}_3^+ \quad (2)
\]
\[
\text{RNH}_2\text{COO}^- + \text{H}_2\text{O} \rightarrow \text{RNHCOO}^- + \text{H}_3\text{O}^+ \quad (3)
\]

\[
\text{RNH}_2\text{COO}^- + \text{OH}^- \rightarrow \text{RNHCOO}^- + \text{H}_2\text{O} \quad (4)
\]

where R denotes \(-\text{CH}_2\text{CH}_2\text{OH}\).

Combining Eqs. (1)-(4), the following expression was obtained based on the assumption of quasi-steady state for the zwitterion concentration.

\[
r_{\text{CO}_2} = \frac{-k_i C_{\text{CO}_2} C_{\text{MEA}}}{1 + \frac{k_{-1}}{k_3 C_{\text{MEA}} + k_3 C_{\text{H}_2\text{O}} + k_4 C_{\text{OH}^-}}} \quad (5)
\]

Generally, the zwitterion deprotonation was much faster than its decomposition, i.e.,

\[
k_{-1} / \left(k_3 C_{\text{MEA}} + k_3 C_{\text{H}_2\text{O}} + k_4 C_{\text{OH}^-}\right) \ll 1,
\]

then the rate of CO\(_2\) reaction with MEA could be expressed by a simple second-order kinetics\(^{29,34}\):

\[
-k_{\text{CO}_2} = k_i C_{\text{CO}_2} C_{\text{MEA}} \quad (6)
\]

where \(k_i\) could be determined by the following empirical correlation:\(^{34}\)

\[
\log_{10} k_i = 10.99 - 2152 / T \quad (7)
\]

The mass balance for CO\(_2\) and MEA in the liquid phase based on two-film model was written in the following,

\[
\begin{align*}
D_{\text{CO}_2} \left( \frac{d^2 C_{\text{CO}_2}}{dz^2} \right) - k_i C_{\text{CO}_2} C_{\text{MEA}} = 0 \\
D_{\text{MEA}} \left( \frac{d^2 C_{\text{MEA}}}{dz^2} \right) - k_i C_{\text{CO}_2} C_{\text{MEA}} = 0
\end{align*}
\quad (8)
\]

Boundary conditions,

\[
z = 0, \ C_{\text{CO}_2} = C_{\text{CO}_2,i}, \quad \frac{dC_{\text{MEA}}}{dz} = 0; \quad z = \delta, \ C_{\text{CO}_2} = 0, \ C_{\text{MEA}} = C_{\text{MEA},L} \quad (9)
\]

where \(C_{\text{CO}_2,i}\) was CO\(_2\) concentration at the gas-liquid interface, \(mol/m^3\); \(C_{\text{MEA},L}\) was
MEA concentration in the bulk liquid phase, mol/m$^3$. $D_{CO}$ and $D_{MEA}$ were the
diffusion coefficient of CO$_2$ and MEA in aqueous MEA solution$^{36}$, respectively, m$^2$/s.

$$\ln D_{MEA} = -13.275 - 2198.3/T - 7.8142 \times 10^{-4} C_{MEA}$$  \hspace{1cm} (10)$$

$$D_{CO} = \left(2.35 + 0.029837 C_{MEA} - 9.7078 \times 10^{-3} C_{MEA}^2\right) \times 10^{-6} \exp\left(-2119 - 20.132 C_{MEA}\right)$$ \hspace{1cm} (11)$$

For determining the enhancement factor, $E$, the analytical solution of van Krevelen
and Hoftijzer was adopted$^{36}$:

$$E = \frac{\sqrt{Ha^2 E_\infty - E}}{\tanh \sqrt{Ha^2 E_\infty - E}} - 1$$ \hspace{1cm} (12)$$

where $Ha$ and $E_\infty$ as Hatta number and the infinite enhancement factor based on
two-film model, respectively, were expressed as,

$$Ha = \sqrt{\frac{D_{CO} k_{L,c} C_{MEA,L}}{k_0^L}}$$ \hspace{1cm} (13)$$

$$E_\infty = 1 + \frac{D_{MEA} C_{MEA,L}}{D_{CO} C_{CO_2,L}}$$ \hspace{1cm} (14)$$

where $k_0^L$ denoted the liquid side physical mass transfer coefficient, m/s.

### 3.2. Modeling of Mass Transfer Coefficient

A unit cell model for Taylor flow was used$^{14,16,25}$ as shown in Figure 2(a). There
were a few assumptions associated with the model: (i) CO$_2$ and aqueous MEA
solution were well-mixed in each phase in a unit cell; (ii) the overall unit cell was
considered as a plug flow; (iii) mass transfer and reaction from CO$_2$ to aqueous MEA
solution only happened within a unit cell; and (iv) each unit cell was independent.
**Figure 2.** Schematic of typical gas-liquid two-phase Taylor flow in a microchannel. (a) Taylor bubble and slug in a unit cell. (b) Cross-section of Taylor bubble (not to scale).

The mass balance of CO\(_2\) in a unit cell was described as the following.

\[
\frac{P_{\text{CO}_2}}{RT} \frac{dV_B}{dX} U_B = -k_L a \left( C_{\text{CO}_2}^L - C_{\text{CO}_2}^* \right) (V_B + V_L) \tag{15}
\]

where \(P_{\text{CO}_2}\) was gas phase pressure which varied along the microchannel as a result of local pressure, Pa; \(R\) was gas constant, 8.3145 J/(mol·K); \(T\) was operating temperature, K; \(V_B\) was volume of CO\(_2\) bubble, m\(^3\); \(U_B\) was velocity of CO\(_2\) bubble, m/s, varying along the microchannel as a result of mass transfer and reaction; \(X\) was position along the serpentine microchannel, m. \(k_L\) was liquid phase mass transfer coefficient, m/s. \(a\) was specific interfacial area \((a = A_S / (V_{\text{CO}_2} + V_L))\) , m\(^{-1}\). \(A_S\) was effective mass transfer area, m\(^2\). \(V_L\) was liquid phase volume including liquid slug and liquid film, m\(^3\). \(C_{\text{CO}_2}^L\) was CO\(_2\) concentration in bulk liquid phase solution along the serpentine microchannel, mol/m\(^3\). \(C_{\text{CO}_2}^*\) was saturated solubility of CO\(_2\) in aqueous MEA solution, mol/m\(^3\), which was calculated based on Henry’s Law,

\[
C_{\text{CO}_2}^* = \frac{P_{\text{CO}_2}}{1000 H_{\text{CO}_2}^{\text{loaded}}} \tag{16}
\]

where \(H_{\text{CO}_2}^{\text{loaded}}\) is Henry’s constant of CO\(_2\) in the loaded aqueous MEA solution with CO\(_2\), kPa·m\(^3\)·mol\(^{-1}\), which can be estimated by N\(_2\)O approximation method,\(^{37-41}\)
\[
\frac{H_{\text{loaded}}^{\text{CO}_2}}{H_{\text{loaded}}^{\text{N}_2\text{O}}} = \frac{H_{\text{H}_2\text{O}}^{\text{CO}_2}}{H_{\text{H}_2\text{O}}^{\text{N}_2\text{O}}}
\]

(17)

\[
\ln\left(\frac{H_{\text{H}_2\text{O}}^{\text{N}_2\text{O}}}{H_{\text{N}_2\text{O}}}\right) = -700.6501 + 14905.5017/T + 126.3505 \ln T - 0.2267T
\]

(18)

Henry’s constant of CO\(_2\) in water was proposed by Versteeg and van Swaaij.\(^{42}\)

\[
H_{\text{H}_2\text{O}}^{\text{CO}_2} = 2.8249 \times 10^3 \exp(-2044/T)
\]

(19)

where \(H_{\text{CO}_2}^{\text{loaded}}\) is Henry’s constant of CO\(_2\) and N\(_2\)O in the loaded aqueous MEA solution with CO\(_2\), kPa\(\cdot\)m\(^3\)\(\cdot\)mol\(^{-1}\); \(H_{\text{H}_2\text{O}}^{\text{CO}_2}\) and \(H_{\text{N}_2\text{O}}^{\text{CO}_2}\) are Henry’s constants of CO\(_2\) and N\(_2\)O in water, respectively, kPa\(\cdot\)m\(^3\)\(\cdot\)mol\(^{-1}\). For the loaded aqueous MEA solution system, the correlation of Henry’s constant was proposed by Hartono et al.\(^{41}\)

\[
\ln\left(H_{\text{N}_2\text{O}}^{\text{loaded}}\right) = \ln\left(\Delta H_{\text{N}_2\text{O}}^{\text{loaded}}\right) + \ln\left(H_{\text{N}_2\text{O}}^{\text{unloaded}}\right)
\]

(20)

\[
\ln\left(\Delta H_{\text{N}_2\text{O}}^{\text{loaded}}\right) = 0.77m_{\text{MEA}} \cdot \alpha T + 0.033m_{\text{MEA}} \cdot \alpha T
\]

(21)

\[
\ln\left(H_{\text{N}_2\text{O}}^{\text{unloaded}}\right) = \ln\left(\Delta H_{\text{N}_2\text{O}}^{\text{unloaded}}\right) + \varphi_{\text{MEA}} \ln\left(H_{\text{MEA}}^{\text{MEA}}\right) + \varphi_{\text{H}_2\text{O}} \ln\left(H_{\text{H}_2\text{O}}^{\text{N}_2\text{O}}\right)
\]

(22)

\[
\ln\left(\Delta H_{\text{N}_2\text{O}}^{\text{unloaded}}\right) = (1.9210 - 1.6792 \times 10^{-3}T + 3.0951m_{\text{MEA}}\rho_{\text{MEA}} + m_{\text{MEA}} \varphi_{\text{MEA}})\rho_{\text{H}_2\text{O}} + (1 - m_{\text{MEA}})\sqrt{1 - \varphi_{\text{MEA}}^2}
\]

(23)

\[
\varphi_{\text{MEA}} = \frac{m_{\text{MEA}} \cdot \rho_{\text{MEA}}}{m_{\text{MEA}} \cdot \rho_{\text{MEA}} + (1 - m_{\text{H}_2\text{O}}) \cdot \rho_{\text{H}_2\text{O}}}
\]

(24)

\[
\ln\left(H_{\text{MEA}}^{\text{MEA}}\right) = 3.9451 - \frac{589.9365}{T - 99.0406}
\]

(25)

where \(\Delta H_{\text{N}_2\text{O}}^{\text{loaded}}\) and \(\Delta H_{\text{N}_2\text{O}}^{\text{unloaded}}\) were the apparent Henry’s constant deviation in aqueous MEA solution with and without CO\(_2\), kPa\(\cdot\)m\(^3\)\(\cdot\)mol\(^{-1}\). \(H_{\text{N}_2\text{O}}^{\text{unloaded}}\) was Henry’s constant of N\(_2\)O in aqueous MEA solution without CO\(_2\), kPa\(\cdot\)m\(^3\)\(\cdot\)mol\(^{-1}\). \(m_{\text{MEA}}\) and \(m_{\text{H}_2\text{O}}\) were the mole fraction of MEA and H\(_2\)O in aqueous MEA solution, respectively. \(\alpha\) was CO\(_2\) loading in aqueous MEA solution, \(\alpha\text{mol}_{\text{CO}_2}/\text{mol}_{\text{MEA}}\). \(\varphi_{\text{MEA}}\) and \(\varphi_{\text{H}_2\text{O}}\) were the volume fraction of MEA and H\(_2\)O, respectively. \(H_{\text{MEA}}^{\text{MEA}}\) was Henry’s constant of N\(_2\)O in MEA, kPa\(\cdot\)m\(^3\)\(\cdot\)mol\(^{-1}\). \(M_{\text{MEA}}\) and \(M_{\text{H}_2\text{O}}\) were the molecular mass of MEA and H\(_2\)O.
H₂O, respectively, kg·mol⁻¹. ρ_MEA and ρ_H₂O were the density of MEA and H₂O, respectively, kg·m⁻³. The reaction between MEA and CO₂ was considered as a fast irreversible process. In this case, CO₂ concentration in the bulk liquid phase was assumed zero, given that the liquid side mass transfer coefficient was significantly lower than the apparent reaction rate as analyzed by Kasturi and Stepanek.⁴³

### 3.3. Determination of Bubble Volume

Figure 2(b) shows the bubble cross-section model. Its volume and surface area were expressed by the following:¹⁴,⁴⁴

\[
V_{CO₂} = \left[ (\pi - 4) I^2 + (d_H - 2\delta)^2 \right] \left[ L_B - d_H \right] + \frac{4}{3} \pi (0.5d_H - \delta)^3
\]

\[ (26) \]

\[
A_s = \left[ 2\pi I + 4(d_H - 2\delta - 2I) \right] (L_H - d_H) + 4\pi (0.5d_H - \delta)^2
\]

\[ (27) \]

where \( I \) was the radius of the quadrant at the corner and obtained from the captured images, m. \( d_H \) and \( L_B \) were the hydraulic diameter and the length of Taylor bubble, respectively, m. \( \delta \) denoted the liquid film thickness near the channel wall center and estimated by the correlation from Aussillous and Quéré.⁴⁵

\[
\frac{\delta}{D_H} = \frac{0.67Ca^{2/3}}{1 + 3.35Ca^{2/3}}
\]

\[ (28) \]

\[
Ca = \frac{\mu_L U_B}{\gamma_{TP}}
\]

\[ (29) \]

where \( Ca \) was capillary number, \( \mu_L \) was the liquid phase viscosity, Pa·s. \( \gamma_{TP} \) was the gas-liquid two-phase interfacial tension and expressed by the correlation proposed by Vázquez et al.⁴⁶

\[
\gamma_{TP} = \gamma_{H₂O} - \left( \gamma_{H₂O} - \gamma_{MEA} \right) \left( 1 + \frac{0.6272m_{H₂O}}{1 - 0.9465m_{H₂O}} \right) m_{MEA}
\]

\[ (30) \]
where $\gamma_{H_2O}$ and $\gamma_{MEA}$ were the surface tension of pure water and MEA, respectively, N/m ($25^\circ$C, $\gamma_{H_2O} = 0.07201, \gamma_{MEA} = 0.04895$). The solution viscosity of the aqueous MEA solution without CO$_2$ was calculated by the correlation by Hartono et al.$^{41}$

$$\ln\left(\mu_{\text{unloaded}}\right) = m_{MEA}\left(8.36 - 4.664 \times 10^{-2} t + 1.6 \times 10^{-4} t^2 - 4.14 m_{MEA}\right) m_{H_2O} + m_{H_2O} \ln\left(\mu_{H_2O}\right) + m_{MEA} \ln\left(\mu_{MEA}\right)$$

(31)

where $\mu_{\text{unloaded}}$ was the viscosity of aqueous MEA solution without CO$_2$ absorption, Pa·s. $\mu_{MEA}$ and $\mu_{H_2O}$ were the viscosity of pure MEA and water, Pa·s ($25^\circ$C, $\mu_{H_2O} = 8.937 \times 10^{-4}; \mu_{MEA} = 1.79 \times 10^{-2}$). $t$ was the solution temperature, $^\circ$C. The concentration of MEA was very low, and the solution viscosity with CO$_2$ was replaced by $\mu_{\text{unloaded}}$. So, the film thickness was calculated from Eqs. (28)-(31).

In this work, $L_{CO_2}$ and $U_B$ were calculated along the microchannel by 1000 continuously captured images. The bubble volume was calculated by Eq. (26) along the channel position, and then $dV_B/dX$. The liquid phase volume within the unit cell was determined from the formation frequency of the liquid slug. The gas phase pressure was measured by the pressure sensor. The equilibrium solubility along the channel position was calculated by Eqs. (16)-(25). Finally, $E$ and $k_{L,a}$ were calculated by Eqs. (12) & (15), respectively.

4. RESULTS AND DISCUSSION

4.1. Length and Velocity of Gas Bubbles

The stable gas-liquid Taylor flow was formed at the T-junction and tracked along the serpentine microchannel. Figure 3 illustrates the evolution of CO$_2$ bubbles with and without chemical reaction along the microchannel. More experimental data are...
shown in Figure S1 of Supporting Information (SI). As bubbles moved downstream, their length gradually decreased and finally leveled off, indicating CO$_2$ absorption reached equilibrium, which was similar to CO$_2$-H$_2$O system.$^{14}$

**Figure 3.** The evolution of Taylor bubble length along the serpentine microchannel at 25 °C, 1.0 atm and $W_{\text{MEA}}$=1 wt% for (a) $Q_G = 5.5$ mL/min, and (b) $Q_L = 1$ mL/min.

Initial bubble length at the T-junction (X=0) decreased with an increase in the liquid flow rate (1.0-2.5 mL/min) (Figure 3(a) & Figure S1(a)), or with reducing gas flow rates (Figure 3(b) & Figure S1(b)). Compared to CO$_2$-H$_2$O system, the initial bubble length appeared reduced considerably, where the reducing extent decreased with increasing liquid flow rate for a given gas flow rate. These phenomena were the results of the combination of the physical properties of the solution and chemical reaction. The increase in MEA content (0-1 wt%) in solution led to the increasing of the liquid viscosity and the decreasing of gas-liquid interfacial tension, as shown in Table 2. Because the variation of the interfacial tension was very small (72.01-71.18 N·m), the effects of the gas-liquid interfacial tension on the initial bubble length were insignificant.$^{47}$ Previous studies have shown the initial bubble length decreased with the increasing of the liquid phase viscosity.$^{10,48}$ In addition, the mass transfer with chemical reaction during the formation of Taylor bubble further decreased the initial
During Taylor bubble flowing along the microchannel, the shrinkage of CO$_2$ bubble was accelerated by chemical reaction in comparison to CO$_2$-H$_2$O system (Figure 3 & Figure S1). In the first 60 mm ($X \leq 60$ mm) section, which was named as the rapid shrinkage stage, Taylor bubble shrank rapidly where approximate 70% of total length reduction ($\Delta L_B$) was observed. In the second 60 mm ($60 < X \leq 120$ mm) section, the reduction of $\Delta L_B$ approached 25%, which was regarded as the moderate shrinkage stage. In the third 60 mm ($120 < X \leq 180$ mm) section, the bubble length kept almost constant where only 5% of $\Delta L_B$ was completed, suggesting an equilibrium shrinkage stage. This originated from the interplay of three competitive factors during the evolution of the bubbles, i.e., CO$_2$ dissolution, chemical reaction and gas-liquid two-phase pressure drop.

As found in our previous work, the evolution of the bubbles along the microchannel was determined by CO$_2$ dissolution and pressure drop. The existence of chemical reaction in the present work could intensify the shrinkage of Taylor bubble, which led to shorter bubble lengths and 70% of $\Delta L_B$ at the rapid shrinkage stage, as shown in Figure 4. With the bubble flowing downstream, MEA was gradually consumed, the reaction rate was weakened, and the decrease in bubble length reduced.
and finally leveled off.

**Figure 4.** The effects of MEA content on the bubble length of along the serpentine microchannel for $Q_G=5$ mL/min and $Q_L=1$ mL/min.

Figure 5 & Figure S2 depict the evolution of Taylor bubble velocity accompanied with and without chemical reaction along the serpentine microchannel by varying gas (or liquid) flow rate. It was also observed that the bubble velocity decreased drastically for aqueous MEA solution, compared to the CO$_2$-H$_2$O system. The reduction in Taylor bubble velocity at three stages (the rapid, moderate and equilibrium shrinkage stages) were found to be 75%, 19% and 6% for a given gas flow rate, respectively. Figure 5(a) & Figure S2(a) also present the existence of a critical point during the evolution of the bubble velocity for CO$_2$-MEA system. It was attributed to the competition of two kinds of modes when increasing in liquid flow rate. One was the increase in the bubble velocity (the increasing mode), while the other one was the increase in the absorption rate that reduced the bubble velocity (the decreasing mode). As could be seen from Figure 5(a) & Figure S2(a), the bubble velocity increased with the liquid flow rate before the critical point, indicating that the increasing mode was dominant. After the critical point, the decreasing mode began to dominate. Figure 5(b) & Figure S2(b) shows Taylor bubble velocity increased with
increasing gas flow rate for a given liquid flow rate. Obviously, the gas flow rate dominated over the increase in bubble velocity. The reductions in bubble velocity at three stages were 72%, 22% and 6% for a given liquid flow rate, respectively. The effects of MEA content on bubble velocity are shown in Figure 6. The reduction in bubble velocity in three stages was found to be 83%, 14% and 3%, respectively. The changes of in bubble velocity were similar to that of CO$_2$ bubble length as displayed in Figure 4. The results allowed us to identify key parameters such as flow rate and absorption rate for understanding and quantifying the dynamic evolution of mass transfer.

Figure 5. The evolution of Taylor bubble velocity along the serpentine microchannel at 25 °C, 1.0 atm and W$_{\text{MEA}}$=1 wt% for (a) Q$_G$=5.5 mL/min, and (b) Q$_L$=1 mL/min.

Figure 6. The effects of MEA content on the velocity of Taylor bubble along the serpentine microchannel for Q$_G$=5 mL/min and Q$_L$=1 mL/min.
4.2. Gas-Liquid Mass Transfer Coupled with Chemical Reaction

$k_{La}$, $k_L$ and $a$ are generally regarded as key parameters to characterize the gas-liquid mass transfer. Figure 7 & Figure S3 show the comparison of measured $k_{La}$ along the serpentine microchannel for CO$_2$-H$_2$O and CO$_2$-MEA systems. Compared to CO$_2$-H$_2$O system, $k_{La}$ increased considerably due to the chemical reaction. For a given gas (or liquid) flow rate, $k_{La}$ increased with the increasing of liquid (or gas) flow rate. The effects of the liquid flow rate were more significant, which could be also attributed to the role of chemical reaction. Moreover, $k_{La}$ in a unit cell decreased along the microchannel for both CO$_2$-H$_2$O and CO$_2$-MEA system, where the latter exhibited more dramatic decrease.

**Figure 7.** The effects of the gas or liquid flow rate on $k_{La}$ in a unit cell along the serpentine microchannel at 25 °C, 1.0 atm and $W_{MEA}=1$ wt% for (a) $Q_G=5.5$ mL/min, and (b) $Q_L=1$ mL/min.

In the rapid shrinkage stage (X≤60 mm), approximate 78% reduction in $k_{La}$ ($\Delta(k_{La})$) was obtained, where $\Delta(k_{La})$ denoted the difference between the initial (X=0 mm) and the end (X=180 mm) of the volumetric liquid phase mass transfer coefficient in a unit cell. In the moderate shrinkage stage (60<X≤120 mm), $\Delta(k_{La})$ approached 17%. In the equilibrium shrinkage stage (120<X≤180 mm), the value of $k_{La}$ decreased until
becoming almost constant, where only 5% occurred. The effects of MEA content on $k_{L,a}$ are depicted in Figure 8. $k_{L,a}$ increased with increasing MEA content due to the reaction enhancement. About 78% and 17% reduction in $k_{L,a}$ were obtained in the first and second stages, respectively. The significantly higher reduction in the first stage demonstrated the kinetic effect of the fast reaction.

**Figure 8.** The effects of MEA content on $k_{L,a}$ in a unit cell along the serpentine microchannel at 25 °C and 1.0 atm for $Q_G = 5$ mL/min and $Q_L = 1$ mL/min.

In order to further elucidate the gas-liquid mass transfer performance for CO$_2$-MEA system, $k_L$ and $a$ were separately analyzed. Figure 9 & Figure S4 compares the evolution of $k_L$ along the microchannel for both CO$_2$-MEA and CO$_2$-H$_2$O systems. The decrease of $k_L$ in CO$_2$-MEA system was more dramatic than that in CO$_2$-H$_2$O system due to the intensification of chemical reaction, which was found to be 68%, 24% and 8% in three stages, respectively.

For a given gas flow rate, $k_L$ increased with the increasing liquid phase flow rate, but decreased sharply as Taylor bubbles moved downstream along the microchannel for CO$_2$-MEA system (Figure 9(a) & Figure S4(a)), which was determined synergistically by the leakage flow in the liquid film and the circulation in the liquid slug. Taylor bubble velocity decreased sharply for CO$_2$-MEA system (Figure 5(a) &
Figure S2(a)), which led to the significant decrease of the circulation and then $k_L$ and such effects were weakened gradually along the microchannel. In addition, increasing the viscosity of aqueous MEA solution (Table 2) could also decrease $k_L$ by reducing the diffusion coefficient and the circulation. However, the substantial decrease of the bubble length (Figure 3(a) & Figure S1(a)) caused the reduction of the liquid film length between the bubble and the channel wall, where the leakage flow in the film was enhanced and in favor of increasing $k_L$. With the consumption of MEA in liquid phase, the effect of chemical reaction was gradually weakened, where $k_L$ approached to CO$_2$-H$_2$O system. Finally, both positive and negative effects on $k_L$ determined the results of Figure 9(a) & Figure S4(a).

Figure 9. The evolution of $k_L$ in a unit cell along the serpentine microchannel at 25 °C, 1.0 atm and W$_{MEA}$=1 wt% for (a) $Q_G$ = 5.5 mL/min, and (b) $Q_L$ = 1 mL/min. For a given liquid flow rate, there was also a critical position (X ≈ 25 mm) for $k_L$ along the microchannel (Figure 9(b) & Figure S4(b)), where the reductions in $k_L$ at three stages were 67%, 24% and 9%, respectively. When X < 25 mm, $k_L$ increased with the increase in the gas phase flow rate, and decreased as X > 25 mm. This was similar to the conclusion of our previous work. However, the point moved forward compared to CO$_2$-H$_2$O system. As we all known, CO$_2$ absorption capacity was...
constant for a given initial MEA content, the larger the bubble length or the gas flow rate was, the more decreasing extent of free MEA molecule and CO$_2$ absorption, which weakened dramatically the intensification of chemical reaction on $k_L$ as absorption proceeded. Simultaneously, the liquid slug velocity decreased due to CO$_2$ absorption, which also reduced the circulation in the liquid slug. On the other hand, the increasing gas flow rate and the decreasing bubble length were in favor of $k_L$ by enhancing the circulation and the leakage flow, respectively. Compared to CO$_2$-H$_2$O system, both the negative and positive effects on $k_L$ were enlarged due to chemical reaction, and finally the critical point occurred in advance for CO$_2$-MEA system.

**Figure 10.** The evolution of $a$ in a unit cell along the serpentine microchannel at 25°C, 1.0 atm and $W_{MEA}=1$ wt% for (a) $Q_G = 5.5$ mL/min, and (b) $Q_L = 1$ mL/min.

Figure 10 & Figure S5 shows the evolution of $a$ in a unit cell along the serpentine microchannel. The reduction of $a$ in three stages were 57%, 30% and 13%, respectively, which were smaller than the variation of $k_L$. Compared to CO$_2$-H$_2$O system under the identical conditions, the decrease of $a$ was accelerated as Taylor bubbles moved downstream along the microchannel, especially for the initial shorter bubble length which approached to spherical (i.e., bubble sphericity $\Phi$), as shown in Figure 11 & Figure S6. The effects of MEA content on gas-liquid mass transfer
performance, in terms of $k_L$, $a$ and $\Phi$, are depicted in Figure 12. The reductions in $a$ and $k_L$ in three stages were 60%, 30% and 10%, respectively. It was observed $k_L$ and its reducing degree increased with increasing MEA content. On the one hand, the decrease in the length of the bubble and the liquid film could lead to the increase of $k_L$. On the other hand, both the decrease of the bubble velocity and the increase of the liquid phase viscosity resulted in the decrease of $k_L$ by weakening the circulation. Finally, the positive and negative factors from MEA content together determined the dynamic changes of $k_L$ (Figure 12(a)). As generally known, the increase of MEA content resulted in increasing CO$_2$ absorption rate and capacity in aqueous MEA solution, which further accelerated the decrease of bubble length and the increase of its $\Phi$, and led to the decrease of $a$ (Figure 12(b) & (c)).

**Figure 11.** The evolution of the sphericity ($\Phi$) along the serpentine microchannel, at 25 °C, 1.0 atm and $W_{MEA}$=1 wt% for (a) $Q_G = 5.5$ mL/min, and (b) $Q_L = 1$ mL/min.
Figure 12. The effects of the MEA content on $a$ (a), $k_L$ (b) and $\Phi$ (c) along the serpentine microchannel at 25 °C, 1.0 atm for $Q_G = 5$ mL/min and $Q_L = 1$ mL/min.

4.3. Dynamic Changes of the Enhancement Factor

It was reasonably expected that gas-liquid two-phase mass transfer performance could be enhanced by chemical reaction for CO$_2$-MEA system. To quantitatively characterize the process, $E$ was adopted to represent the ratio of mass transfer between with and without chemical reaction. However, the local dynamic conditions are generally difficult to measure directly in any reactor because the mass transfer never stops before reaching the absorption equilibrium, representing the dynamic process of gas-liquid two-phase flow. Figure 13(a) demonstrates the dynamic changes of $E_{exp}$ along the microchannel at four MEA contents, where the experimental enhancement factor ($E_{exp}$) of CO$_2$-MEA system was calculated from the following equation by taking into account of both $k_L^0$ and $k_L$.

$$E_{exp} = \frac{k_L^0 (C_{\text{in}} - C^{\text{eq}}_{\text{CO}_2})}{k_L C^{\text{eq}}_{\text{CO}_2}}$$  \hspace{1cm} (32)$$

where $k_L^0$ denotes the physical mass transfer coefficient for identical operating conditions at the entrance of microchannel for CO$_2$-H$_2$O and CO$_2$-MEA system. That is, the maximum physical mass transfer coefficient is chosen for a given operating
For a given gas and liquid flow rate, $E$ increased with the increasing MEA content and also decreased gradually as Taylor bubbles moved downstream along the microchannel. The chemical reaction rate was proportional to MEA content. Furthermore, the viscosity of the solution increased with the increase of MEA content, which could weaken the influence of chemical reaction. As Taylor bubbles moved beyond the critical value, $E$ tended to be stable and close to one, indicating that the intensification of the chemical reaction was weakened until to be ignored. This suggested the microchannel length could be optimized based on the above critical position for the design of gas-liquid two-phase low pressure drop microchannel reactor.

The enhancement factor ($E_{exp}$) of CO$_2$-MEA system agreed well with the calculated enhancement factor ($E_{cal}$) from Eq. 12, which indicated the validity of the model, as shown in Figure 13(b). Hikita et al.\textsuperscript{50} investigated the kinetics of the reaction of CO$_2$ with MEA for MEA concentration in the range of 0.0152-0.177 mol/L at temperatures ranging from 5.6 °C to 35.4 °C, and found the reaction to be an irreversible second order reaction. In the present work, the initial MEA concentration was varied in the range of 0.163-0.490 mol/L, so the reaction rate can be expressed reasonably by the second-order model. Furthermore, Santiago and Farina\textsuperscript{51} demonstrated that the accuracy of the prediction of this model was 3% for fast second order and irreversible reactions with $Ha > 3$. In the present work, $Ha$ values (Figure 13(c)) fit well in the range stated by Santiago and Farina, further supporting suitability of the expression.
used in the study.

Figure 13. The effects of MEA content on $E$ in a unit cell along the serpentine microchannel (a), prediction of $E$ (b) and $Ha$ (c) for CO$_2$-MEA system at 25 °C, 1.0 atm $Q_G = 5$ mL/min and $Q_L = 1$ mL/min.

4.4. Empirical Correlations for Mass Transfer Performance

Figure 14. Schematic overview of unit cell, bubble, slug and film.

In order to further investigate and predict the mass transfer of the unit cells detailedly, the improved mass transfer model proposed by van Baten and Krishna$^{16}$ was introduced to verify experimental data. The core principle of the model was that
the contributions from caps \((k_L a)_{caps}\) and film \((k_L a)_f\) to mass transfer were evaluated based on Higbie’s penetration theory. The improved mass transfer model was shown in the following.

\[
k_L a = \frac{2\sqrt{2}}{\pi} \sqrt{\frac{D_{CO_2} U_{B-sat}}{d_f}} + \frac{2}{\sqrt{\pi}} \sqrt{\frac{D_{CO_2} U_{B-sat}}{\varepsilon_v L_{UC}}} \frac{4\varepsilon_v}{d_f}
\]  

(33)

where \(U_{B-sat}\), \(\varepsilon_v\) and \(L_{UC}\) were the difference of the transient bubble velocity and the saturated bubble velocity for absorption equilibrium state, the gas void fraction based on the unit cell volume and the unit cell length, respectively.

It was well known that the basic assumption of van Baten’s model (Eq. (33)) was the ideal mixing in the liquid slug for a unit cell. Actually, there was a big concentration distribution in the liquid slug,\(^{52}\) so van Baten’s model was difficult to accurately predict the experimental data of this work. Considering that the deviation mainly originated from the non-ideal mixing in the liquid slug by mass exchange with the liquid film layer close to the wall, a correction term in the form of Eq. (34) was proposed.

\[
(k_L a)_c = \frac{3.66D_{CO_2} L_s (d_h - 2\delta)}{d_h^2 L_{UC}}
\]  

(34)

This mass exchange between the slug and liquid film layer could be approximated under-developed laminar flow in a circular channel with a constant boundary,\(^{53,54}\) as well as considering the ratio of the slug length to the unit cell length. The above correlation was derived from the classical theory, where the caps were assumed perfectly spherical and the film was annular due to the circular capillary in van Baten’s model. Compared to the non-spherical caps and the non-annular film due to
the square microchannel in this work, it was necessary to modify the parameters in
order to fit the experimental data, as shown in Table 3.

As shown in Eq. (34), \((k_L)C\) is set as 3.66D\(_{CO2}/d_l\) since the film-slug exchange is
initially simplified as flowing in a circular channel with fully developed laminar flow.
Thus, underdeveloped laminar flow in the liquid film and different thickness of the
liquid film in the cross section require the modification of ideal \((k_L)C\). In this case, the
additional constant is added to the semi-empirical correlation according to the
experimental data.

The validity of the models for both \(CO_2-H_2O\) and \(CO_2-MEA\) systems are shown in
Figures 15 & 16, respectively. The predicted values were well agreement with the
experimental results for a deviation of \(\pm 25\%\). Applying the model for \(CO_2-H_2O\)
system to \(CO_2-MEA\) system, the net physical mass transfer coefficient could be
approximately separated from chemical absorption process.

The basic assumption of van Baten’s model was the ideal mixing in the liquid slug
for a unit cell without concentration gradient, where the overall mass transfer
coefficient of the unit cell could be considered as the sum of the overall mass transfer
coefficients from two caps and the film, \(k_La = (k_La)_{caps} + (k_La)_f\). This was inconsistent
with the situation where concentration difference existed between the film and the
bulk liquid slug, and the mass exchange was mainly determined by the internal
circulation in the bulk liquid slug. This is believed to be the main reason for the
inconsistence shown in Figures 15 and 16 for fitting the correlations (Table 3) with
the measured data. More experimental data are shown in Figure S7 & S8 of SI.
Table 3. Separated contributions for predicting \( k_{La} \)

<table>
<thead>
<tr>
<th>System</th>
<th>((k_{La})_{\text{cap}}) (1/s)</th>
<th>((k_{La})_{\text{film}}) (1/s)</th>
<th>((k_{La})_{C}) (1/s)</th>
<th>(R^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{H}_2\text{O}-\text{CO}_2)</td>
<td>(1.4\sqrt{\frac{\pi}{y}} \sqrt{\frac{D_{\text{CO}<em>2}U</em>{\text{H-oil}}}{d_{H}}} \frac{4}{L_{UC}})</td>
<td>(5.4\sqrt{\pi} \sqrt{\frac{D_{\text{CO}<em>2}U</em>{\text{H-oil}}}{d_{H}}} \frac{4e_s}{e_{L-UC}} \frac{1}{d_{H}})</td>
<td>(-13 \frac{3.66D_{\text{CO}<em>2}L</em>{2}(d_{H}-2\delta)}{d_{H}} \frac{1}{d_{H}^2L_{UC}})</td>
<td>0.95</td>
</tr>
<tr>
<td>(\text{MEA}-\text{CO}_2)</td>
<td>(\sqrt{\frac{\pi}{y}} \sqrt{\frac{D_{\text{CO}<em>2}U</em>{\text{H-oil}}}{d_{H}}} \frac{4}{L_{UC}})</td>
<td>(7.4\sqrt{\pi} \sqrt{\frac{D_{\text{CO}<em>2}U</em>{\text{H-oil}}}{d_{H}}} \frac{4e_s}{e_{L-UC}} \frac{1}{d_{H}})</td>
<td>(-35 \frac{3.66D_{\text{CO}<em>2}L</em>{2}(d_{H}-2\delta)}{d_{H}} \frac{1}{d_{H}^2L_{UC}})</td>
<td>0.96</td>
</tr>
</tbody>
</table>

Figure 15. Prediction of overall mass transfer coefficients for \(\text{CO}_2-\text{H}_2\text{O}\) system at 25\(^o\)C, 1.0 atm for (a) \(Q_L = 1\) mL/min, and (b) \(Q_G = 5.5\) mL/min.

Figure 16. Prediction of overall mass transfer coefficients for \(\text{CO}_2\)-\(\text{MEA}\) system at 25\(^o\)C, 1.0 atm and \(W_{\text{MEA}} = 1\) wt% for (a) \(Q_L = 1\) mL/min, and (b) \(Q_G = 5.5\) mL/min.

As analyzed above, the semi-empirical correlations were proposed to predict the physical/chemical mass transfer coefficients in the \(\text{CO}_2-\text{H}_2\text{O}\) system. If the range of the parameters in the empirical correlation is reasonably wide to adapt the situation of
chemical mass transfer coefficient, the enhancement factor can be estimated. It should be noted that the semi-empirical correlations may not be suitable when beyond the scope of the experimental conditions. For the CO$_2$-MEA system, the semi-empirical correlation indeed reflects the effect of only an overall mass transfer coefficient covering both mass transfer and chemical reactions. This is mainly due to the fact that they are impractically measureable separately in the dynamic physical/chemical coupling process, although approximate solutions may be obtained through numerical analysis. The development in this work with in-situ measurements along microchannels provides a practical approach to quantify the parameters in order to estimate the overall apparent mass transfer coefficient in the CO$_2$-MEA system, which is still difficult in the macroscale system. It can also be considered as a benchmark for estimating the enhancement factor of other physical/chemical coupling systems having different kinetics, such as CO$_2$-DEA and CO$_2$-MDEA. Nevertheless, by incorporating a wider variety of experimental data a simpler empirical correlation may be established for determining the enhancement factor. This is under active investigation in our laboratories.

5. CONCLUSIONS

Dynamic coupling of mass transfer and chemical reaction during Taylor flow in serpentine microchannels was experimentally characterized by an online method. The dynamic changes of gas bubbles, and key mass transfer parameters including $k_La$, $k_L$, and $a$ were measured or calculated under the optimized experimental conditions (i.e., $Q_G$, $Q_L$, $W_{MEA}$), respectively. The dynamic enhancement of mass transfer by chemical
reaction was further quantified in terms of $E$. Based on the experimental results, a modified van Baten’s model was proposed to predict mass transfer performance for both CO$_2$-H$_2$O and CO$_2$-MEA systems and the $R^2$ values were 0.95 and 0.96, respectively.

Overall, the dynamic shrinkage of Taylor bubbles flowing along the microchannel was observed, showing more significant changes in CO$_2$-MEA system. Three stages of the shrinkage of Taylor bubbles were observed, where about 70% of the length reduction took place in the first stage, and lower change rates in other stages, indicating the dynamic impact of the fast chemical reaction. Compared to CO$_2$-H$_2$O system, $k_La$ was considerably intensified due to chemical reaction for CO$_2$-MEA system. The dynamic decrease and experiencing three stages of $k_La$ in a unit cell was also found along the microchannel for CO$_2$-MEA system, where $k_La$ in the first stage reached approximately 70%. $k_L$ also decreased sharply as Taylor bubbles moved downstream along the microchannel for CO$_2$-MEA system. For a given liquid flow rate, the decreasing curves of $k_L$ along the microchannel for different gas flow rates crossed at a critical position, indicating the interplay between the effects of chemical reactions and the physical shape of bubbles.

Finally, the dimensionless correlations on the mass transfer performance for both CO$_2$-H$_2$O and CO$_2$-MEA systems showed that the calculated values agreed well with the experimental results and the experimental results for a deviation of ± 25%, demonstrating the feasibility of the model for predicting key mass transfer parameters. The correlations also suggested the net physical mass transfer performance in...
CO₂-MEA system could be reasonably separated from the chemical absorption process.

6. SUPPORTING INFORMATION

Experimental data under other operating conditions, the derivation process of Eq. 5, Eq.15 and Eq. 34 are shown in Supporting Information.

7. ACKNOWLEDGMENTS

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NOMENCLATURE

\( a \) = specific surface area (m\(^{-1}\))
\( A_\text{s} \) = gas bubble surface area in a unit cell (m\(^2\))
\( C_{\text{CO}_2} \) = CO₂ concentration in liquid phase (mol·m\(^{-3}\))
\( C_{\text{CO}_2}^{*, \text{L}} \) = saturated solubility of CO₂ in liquid phase (mol·m\(^{-3}\))
\( C_{\text{CO}_2}^l \) = CO₂ concentration in liquid phase (mol·m\(^{-3}\))
\( C_{\text{CO}_2, i} \) = CO₂ concentration at gas-liquid interface (mol·m\(^{-3}\))
\( C_{\text{MEA}} \) = MEA concentration in liquid phase (mol·m\(^{-3}\))
\( C_{\text{MEA}, L} \) = MEA concentration in the bulk liquid phase (mol·m\(^{-3}\))
\( C_{\text{MEA}, 0} \) = initial MEA concentration in liquid phase (mol·m\(^{-3}\))
\( d_H \) = microchannel hydraulic diameter (m)
\( D_{\text{CO}_2} \) = diffusion coefficient of CO₂ in aqueous MEA solution (m\(^2\)·s\(^{-1}\))
\( D_{\text{MEA}} \) = diffusion coefficient of MEA in aqueous MEA solution (m\(^2\)·s\(^{-1}\))
$E$ = enhancement factor

$E_\infty$ = infinite enhancement factor

$H_{CO_2}^{\text{loaded}}$ = Henry’s constant of CO$_2$ in the loaded liquid phase (kPa·m$^3$·mol$^{-1}$)

$H_{N_2O}^{\text{loaded}}$ = Henry’s constant of N$_2$O in the loaded liquid phase (kPa·m$^3$·mol$^{-1}$)

$H_{CO_2}^H$ = Henry’s constant of CO$_2$ in water (kPa·m$^3$·mol$^{-1}$)

$H_{N_2O}^H$ = N$_2$O Henry’s constant in water (kPa·m$^3$·mol$^{-1}$)

$H_{MEA}^N$ = N$_2$O Henry’s constant in MEA (kPa·m$^3$·mol$^{-1}$)

$\Delta H_{N_2O}^{\text{unloaded}}$ = Henry’s constant deviation of N$_2$O in unloaded liquid (kPa·m$^3$·mol$^{-1}$)

$\Delta H_{N_2O}^{\text{loaded}}$ = Henry’s constant deviation of N$_2$O in loaded liquid (kPa·m$^3$·mol$^{-1}$)

$I$ = radius of the quadrant at the corner (m)

$k_1$ = second-order reaction rate constant (m$^3$·mol$^{-1}$·s$^{-1}$)

$k_L$ = liquid side mass transfer coefficient (m·s$^{-1}$)

$k_L^0$ = liquid side physical mass transfer coefficient (m·s$^{-1}$)

$L_B$ = Taylor bubble length (m)

$L_{\text{caps}}$ = length of liquid slug and two caps (m)

$L_f$ = liquid film length (m)

$L_S$ = liquid slug length (m)

$L_{UC}$ = unit cell length (m)

$m_{\text{MEA}}$ = mole fraction of MEA in aqueous MEA solution (mol%)

$m_{H_2O}$ = mole fraction of MEA in aqueous MEA solution (mol%)

$M_{\text{MEA}}$ = molecular mass of MEA (kg·mol$^{-1}$)

$M_{H_2O}$ = molecular mass of H$_2$O (kg·mol$^{-1}$)

$P_{CO_2}$ = gas phase pressure (Pa)

$r_{CO_2}$ = CO$_2$ absorption rate in aqueous MEA solution (mol·m$^{-3}$·s$^{-1}$)

$R$ = gas constant (8.3145 J/(mol·K))

$t$ = solution temperature (°C)

$T$ = operating temperature (K)

$U_B$ = gas bubble velocity (m·s$^{-1}$)

$U_{B,sat}$ = the difference between the transient bubble velocity and the saturated bubble velocity for absorption equilibrium state (m·s$^{-1}$)
\( V_B \) = gas phase volume in a unit cell (m\(^3\))

\( V_L \) = liquid phase volume in a unit cell (m\(^3\))

\( W \) = microchannel width (m)

\( X \) = position of the serpentine channel (m)

1. **Greek Symbols**

\( \alpha \) = loading of CO\(_2\) (mol\(_{\text{CO}_2}\)/mol\(_{\text{MEA}}\))

\( \gamma_{H_2O} \) = surface tension of pure water (N·m\(^{-1}\))

\( \gamma_{\text{MEA}} \) = surface tension of pure MEA (N·m\(^{-1}\))

\( \gamma_{\text{TP}} \) = gas-liquid interfacial tension (N·m\(^{-1}\))

\( \delta \) = liquid film thickness near the channel wall center (m)

\( \varepsilon_V \) = gas void fraction based on the unit cell volume

\( \mu_L \) = aqueous MEA solution viscosity (Pa·s)

\( \mu_{\text{MEA}} \) = aqueous MEA solution viscosity without CO\(_2\) (Pa·s)

\( \mu_{\text{MEA}} \) = viscosity of MEA (Pa·s)

\( \mu_{H_2O} \) = viscosity of water (Pa·s)

\( \rho_{H_2O} \) = density of H\(_2\)O (kg·m\(^{-3}\))

\( \rho_L \) = density of aqueous MEA solution (kg·m\(^{-3}\))

\( \rho_{\text{MEA}} \) = density of MEA (kg·m\(^{-3}\))

\( \phi_{H_2O} \) = volume fraction of H\(_2\)O

\( \phi_{\text{MEA}} \) = volume fraction of MEA

\( \Phi \) = bubble sphericity (the ratio of the surface area of a sphere to the surface area of a gas bubble with the same volume)

2. **Dimensionless Parameters**

\( Ca \) = capillary number (\( Ca = (\mu_L U_B) / \gamma_L \))

\( Ha \) = Hatta number (\( Ha = \sqrt{D_{\text{CO}_2} k_C \tilde{C}_{\text{MEA},L} / k_D^0} \))
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