Effect of temperature and flow rate on parallel flow CO\textsubscript{2} absorption by distilled water in co-current microchannel device

Efeito da temperatura e taxa de fluxo na absorção paralela de co2 por água destilada em dispositivo de microcanal co-atual

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ABSTRACT

Mass transfer is a transport phenomenon that has great importance in chemical processes, especially in the processes of separation and absorption. The study of this phenomenon between phases carried out in channels has been developed in smaller scales, seeking to intensify the processes, increasing the mass transfer coefficients. To prove that these studies could be performed at low costs, a pure carbon dioxide absorption by distilled water was performed in a PMMA microchannel designed with 0.008 m of hydraulic diameter. A parallel and stratified flow was achieved in the channel and all experiments had this flow characteristics throughout the changes in operating conditions. In the study, the Reynolds numbers were quantified, corresponding to the interval 212-563. These parameters were acquired by varying the gas surface velocity in the range of 0.46-1.84 m/s and the liquid surface velocity in the range of 0.034-0.071 m/s. The mass transfer quantification was also performed by calculating the volumetric mass transfer coefficient for different operating conditions, which included the surface velocities of the phases and the operating temperature of the system. The results of this work allow the analysis of the effects of temperature and flow characteristics between the liquid and gaseous phases on the mass transfer efficiency in channels. This paper shows that a higher mass transfer coefficient can be achieved in microchannels and that the results show the superiority of these systems compared to conventional mass transfer equipment such as bubble tower, spray tower and static mixers.

Keywords: Microchannel; Parallel flow; Mass transfer; CO\textsubscript{2} absorption.

RESUMO

A transferência de massa é um fenômeno de transporte de grande importância em processos químicos, principalmente nos processos de separação e absorção. O estudo desse fenômeno entre as fases realizadas nos canais foi desenvolvido em escalas menores, buscando intensificar os processos, aumentando os coeficientes de transferência de massa. Para provar que esses estudos poderiam ser
realizados a baixo custo, uma absorção pura de dióxido de carbono pela água destilada foi realizada em um microcanal PMMA projetado com 0,008 m de diâmetro hidráulico. Um fluxo paralelo e estratificado foi alcançado no canal e todos os experimentos tiveram essas características de fluxo ao longo das mudanças nas condições de operação. No estudo, os números de Reynolds foram quantificados, correspondendo ao intervalo 212-563. Esses parâmetros foram adquiridos variando a velocidade da superfície do gás na faixa de 0,46-1,84 m / s e a velocidade da superfície do líquido na faixa de 0,034-0,071 m / s. A quantificação da transferência de massa também foi realizada através do cálculo do coeficiente volumétrico de transferência de massa para diferentes condições operacionais, incluindo as velocidades superficiais das fases e a temperatura operacional do sistema. Os resultados deste trabalho permitem analisar os efeitos das características de temperatura e vazão entre as fases líquida e gasosa na eficiência da transferência de massa em canais. Este artigo mostra que um coeficiente de transferência de massa mais alto pode ser alcançado em microcanais e que os resultados mostram a superioridade desses sistemas em comparação com equipamentos de transferência de massa convencionais, como torre de bolhas, torre de pulverização e misturadores estáticos.

Palavras-chave: Microcanal; Fluxo paralelo; Transferência de massa; Absorção de CO2.

1. INTRODUCTION

Transport phenomena are the basis for any chemical and physical process. This field of engineering covers momentum, heat and mass transfer. These three phenomena together can explain and characterize processes that are present in the scientific area and in the industry. Mass transport, for example, is directly related to the main processes in the industry, such as adsorption, gas-liquid mass transfer systems, new technologies have been studied to improve these processes. The use of microchannels has been emphasized in the scientific researches due to the improvement of mass transfer and heat transfer. The contact between the phases is much greater than in conventional equipment due to the increase of the interfacial area between the gas and the liquid.

In microchannels, the size of the channel creates multiple flow patterns that are caused by the predominance of interfacial forces. The study of the different patterns is important due to the variety of applications of each specific type of flow, depending on the fluid nature, size of channel and application of the microchannel.

The system of absorption of carbon dioxide using water is very important for the industry. Carbon dioxide is one of the major gases responsible for global warming and its capture has both environmental and economic importance. In addition, CO₂ is a very common product in many chemical processes, thus being present in many types of industries. Also, this gas can be used as gas in beverages, extinguishers and reactants.

Due to the low efficiency of gas-liquid mass transfer systems, new technologies have been studied to improve these processes. The use of microchannels has been emphasized in the scientific researches due to the improvement of mass transfer and heat transfer. The contact between the phases
is much greater than in conventional equipment due to the increase of the interfacial area between the gas and the liquid.

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The operating safety and the ability to easily scale up laboratory prototypes are some of the advantages of this type of equipment. On the other hand, the production of these channels is still expensive and sophisticated. It means that less defective channels leads to more expensive systems, because the manufacturing technique tends to be more sophisticated to meet the requirements (Kreutzer, 2003; Sattari-Najafabadi, 2018).

The flow profile studied in this experimental study was the parallel flow or stratified. This flow pattern does not have the formation of bubbles, which is a very important factor, because the formation of bubbles could be a problem in a microchannel system. Due to the fact those channels are difficult to produce in a level of perfection, a bubble can lodge in a curve or channel imperfection, interfering directly in the functionality of the system.

There are still few studies relating the use of microchannels for gas-liquid mass transfer. Most of these studies use bubble formation or flow patterns that form slugs. Few studies report the study of mass transfer without bubble formation. Therefore, the object of the present investigation was the transfer of gas-liquid mass between carbon dioxide and water in a jacketed micro channel produced with polymethyl methacrylate plates. The relationship of velocity variation of both fluids and Reynolds number with mass transfer was also studied. Also, an analysis was performed to discuss the effect of the temperature on the gas-liquid mass transfer system. Finally, a volumetric mass transfer coefficient was calculated for each operating condition. Using this mass transfer coefficient, the developed microchannel device was compared to several types of equipment that perform gas absorption operations to show its superiority.

Fig. 1 – The microchannel used to perform the experiments with corresponding channel’s height and width of 0.8 cm, length of the straight section equals to 17.3 cm and junction angle of 50 degrees.
2. EXPERIMENTAL PROCEDURE

2.1 GAS-LIQUID MICROCHANNEL SPECIFICATION

The mass transfer system was composed of 8 polymethyl methacrylate plates that were assembled, as shown in Fig. 1. The plates were machined to form a system of two channels that combined an entrance and exit in the shape of the letter ‘Y’ one on a different level of the other. The angle of encounter of the channels was 50 degrees. Those two channels joined after the entrance, forming one channel 17.3 cm long and with 0.8 cm of both height and width. The hydraulic diameter of the channel is 0.008 m (Ong, 2008, p. 2757-2773). After that, the channel separated equal to the shape of entrance. In addition, plates were cut to form an envelope around the plates that formed the mass transfer microchannel. This envelope was responsible to the passage of refrigerant fluid. All system connections were made with pneumatic quick-connect fittings to facilitate hose adjustment and channel cleaning. The hoses used to connect the system were pneumatic with 8 mm of diameter and 2 mm thick.

2.2 EXPERIMENTAL SETUP

A water recirculatory equipment was used to control the temperature of the system. Water and gas flow metering systems were based on sensor technologies for Arduino and coupled flowmeter, respectively. For the water flow measurement, an Arduino UNO microcontroller and a flow sensor YF-S401 model was used. This sensor has an operating range from 300 ml/min to 6000 ml/min. The gas flow meter used is coupled to the manometer on the CO$_2$ cylinder available in the laboratory. This manometer is a Harris 351 model, normally used for welding. A schematic diagram of the entire assembled system is shown in Fig. 2.

As shown in Fig. 2, the gas exits the CO$_2$ tank and passes through the coupled flow meter. The CO$_2$ flow is controlled by a valve in this equipment. For distilled water, a pump was placed inside

Fig. 2 – Diagram of the complete system used in laboratory to perform the experiments.
the tank and an external valve to the tank was installed to control that flow. Just after the valve, a water flow sensor was used to perform the readings and ensure a steady flow going to the microchannel system. After the fluids came into contact within the microchannel system, the phases were separated by gravity promoted by the ‘Y’ outlet cut in the PMMA plates. A small piece of piping was placed on the side where the distilled water exited for sample collection. The sample was collected in a vessel with total volume accurately calibrated.

The solution leaving the system that was collected in the vessel was poured immediately into Erlenmeyer containing an excess of a 0.095 M NaOH solution. As a result, the outgoing solution was neutralized and stabilized, allowing no gaseous exchanges with the atmosphere or loss of CO₂ by the solution to occur during the period corresponding to the titration of the other samples. The titrations were performed with a 0.081 M HCl solution, which consisted of titration of excess NaOH and thus determination by difference of the amount of CO₂ absorbed by the water (Benedetti-Pichler, 1939, p. 327-332). The indicators used were phenolphthalein for the first end-point and methyl orange for the second end-point. Finally, titration results after passage into the system were compared to the blank titrations made with the inlet distilled water. All glassware and tanks containing distilled water or the outlet solution of the system were covered as long as possible to avoid contact with the atmosphere.

3. RESULTS AND DISCUSSION

3.1 FLOW PATTERN

Under the experimental conditions used in the present investigation, the parallel flow with undulations was observed in the microchannel. This type of flow fits into the predictions made prior to the beginning of the experiments. The flow is co-current and the channel hydraulic diameter is 0.008 m, which is relatively large when compared to the mass transfer systems used in similar research. This implies that since this channel dimension is large, the Reynolds number tends to be high if we compare with microchannel systems, for example, which have Reynolds within the tens and even smaller than one.

| T (°C) | J₁ (m/s) | J₂ (m/s) | Re₁ |
|--------|----------|----------|-----|
| 10,0   | 0,035    | 0,460    |     |
|        |          | 0,920    | 212,892 |
|        |          | 1,381    |       |
|        |          | 1,841    |       |
|        | 0,071    | 0,460    |     |
|        |          | 0,920    | 432,5424 |
|        |          | 1,381    |       |
| Temperature | Liquid Velocity | Gas Velocity | Reynolds Number |
|-------------|----------------|--------------|----------------|
| 0.035       | 1,841          | 0.460, 0.920, 1.381, 1.841 | 244,1465       |
| 0.071       | 1,841          | 0.460, 0.920, 1.381, 1.841 | 496,0436       |
| 0.035       | 277,279        | 0.460, 0.920, 1.381, 1.841 |               |
| 0.071       | 563,3605       | 1,381, 1.841 |               |

Table 1 – Number of Reynolds as result of variation of the liquid and gas phase superficial velocities and also by variation of the temperature.

Table 1 lists the velocities of the phases obtained in the experiments and the calculated Reynolds numbers. We know that the gas velocity does not significantly affect the Reynolds number as compared to the liquid velocity. Thus, we can observe the effect of this water velocity variation on the Reynolds number by Fig. 3. This happens due to the physical characteristics of the fluids. The water is denser and more viscous than carbon dioxide, since it is a liquid. So, that the change in velocity of water in the channel significantly modifies the intrinsic characteristics of the Reynolds number, which is the relation between inertial forces and viscous forces.

![Reynolds Number Diagram](image)

Fig. 3 – Effect of temperature change in Reynolds number at $V_L = 0.035$ m/s and $V_L = 0.071$ m/s.
In addition, we can analyze the effect of temperature changes in the flow characteristics. We may observe that as the temperature increases, the Reynolds number also increases due to the decrease in viscosity, which is inversely proportional to this dimensionless number. Another point of view would be that with increasing temperature, the fluid becomes less dense as its volume increases and its mass remains the same.

Finally, by analyzing the numbers quantitatively, we observe that when the water velocity is 0.035 m/s, the flow regime is laminar with Reynolds in the range of 200-300, but when the velocity doubles, the Reynolds number increases on average 103.5%, causing the flow to be in the range between 430 and 570 of the laminar flow. This confirms observations made about the flow profile found. This means that the observed ripples are a visual feature of the range in which the flow stabilizes in the laminar flow and as the Reynolds increases, the range increases and becomes more evident.

3.2 MASS TRANSFER COEFFICIENT

The mass transfer flow through the interface formed by the gas-liquid contact can be represented by

\[ N_s = k_L \times (C_{s,l,t} - C_{s,l}) \]  (1)

in which \( k_L \) is the mass transfer coefficient in the liquid phase, \( C_{s,l,t} \) is the concentration of solute at the gas-liquid interface, and \( C_{s,l} \) is the concentration of solute in the liquid phase (Cremasco, 2008).

In cases which the solubility of the gas in the liquid is very low, as is the case with carbon dioxide in the water, the resistance of the gas phase is very low and so the resistance of the liquid phase is predominant when the overall resistance is analyzed. In this way, we can say that the liquid phase controls the mass transfer (Haslam, 1924).

By performing a mass balance in the liquid phase, we can describe the flow of solute that will be transferred from the gas phase to the liquid phase:

\[ (S_{in}) - (S_{out}) + (S_{gen}) = (S_{accu}) \]  (2)

in which \( S_{in} \) is the solute entering the system, \( S_{out} \) is the solute leaving the system, \( S_{gen} \) is the solute generated in the system and \( S_{accu} \) is the accumulation of solute in the system. The term that represents the flow from the liquid phase to the gas phase is zero, since the phase of higher solute concentration is the gas phase. Also, the generation term is zero, since no solute is generated in the liquid phase for the case of carbon dioxide absorption. With this, we have:
Taking the volume and area as intrinsic characteristics of the transfer system, defining \( \frac{A}{V} = a \), with the area being the interfacial area with units of \([m^{-1}]\), we have:

\[
N_s \times A = \frac{d(V \times C_{CO2})}{dt}
\]  

(4)

Substituting Eq. (5) into Eq. (1), we have:

\[
\frac{d(C_{CO2})}{dt} = (k_L \times a) \times (C_{s,li} - C_{s,l})
\]  

(6)

in which \( C_{s,li} \) may be substituted by \( C_{CO2}^* \), as the carbon dioxide concentration at the saturation point, and \( C_{s,l} \) by \( C_{CO2} \), as it represents the measured carbon dioxide concentration at time of quantification.

Finally, we can integrate this equation from time zero, when the concentration of CO\(_2\) in the liquid phase is initial \( C_{CO2}^i \), until time \( t \), where the concentration of CO\(_2\) in the liquid phase is \( C_{CO2} \). Then:

\[
\int_{C_{CO2}}^{C_{CO2}^i} \frac{d(C_{CO2})}{(C_{CO2}^* - C_{CO2})} = (k_L \times a) \times \int_{t}^{0} dt
\]

(7)

\[
\ln \left( \frac{C_{CO2} - C_{CO2}^i}{C_{CO2}^* - C_{CO2}} \right) = -k_L \times a \times t
\]

(8)

We can replace the time by:

\[
t = \frac{L}{v_L}
\]  

(9)
Finally, the final equation of the convective coefficient of volumetric mass transfer is given by (Tortopidis, 1997):

$$k_L \cdot a = \frac{v_L}{L} \cdot \ln \left( \frac{C_{CO2}^* - C_{CO2}^i}{C_{CO2}^* - C_{CO2}} \right)$$  \hspace{1cm} (10)

Table 2 lists the volumetric mass transfer coefficient values calculated for each operating condition.

| T (°C) | J_L (m/s) | J_G (m/s) | k_L·a (s⁻¹) [·10⁻²] |
|-------|-----------|-----------|---------------------|
| 10    | 0,035     |           |                     |
|       | 0,460     | 11,49     |                     |
|       | 0,920     | 23,07     |                     |
|       | 1,381     | 55,00     |                     |
|       | 1,841     | 116,01    |                     |
|       | 0,071     |           |                     |
|       | 0,460     | 27,57     |                     |
|       | 0,920     | 62,55     |                     |
|       | 1,381     | 142,22    |                     |
|       | 1,841     | 282,72    |                     |
| 15    | 0,035     |           |                     |
|       | 0,460     | 6,29      |                     |
|       | 0,920     | 28,53     |                     |
|       | 1,381     | 66,50     |                     |
|       | 1,841     | 202,17    |                     |
|       | 0,071     |           |                     |
|       | 0,460     | 30,50     |                     |
|       | 0,920     | 66,68     |                     |
|       | 1,381     | 195,58    |                     |
|       | 1,841     | 317,87    |                     |
| 20    | 0,035     |           |                     |
|       | 0,460     | 5,87      |                     |
|       | 0,920     | 33,06     |                     |
|       | 1,381     | 86,27     |                     |
|       | 1,841     | 226,61    |                     |
|       | 0,071     |           |                     |
|       | 0,460     | 26,13     |                     |
|       | 0,920     | 83,38     |                     |
|       | 1,381     | 226,11    |                     |
|       | 1,841     | 380,13    |                     |

Table 2 – Volumetric mass transfer coefficient calculated for each operating condition.

Based on the experimental data presented in Table 2, we can first notice that when we set the velocity of carbon dioxide and increase the velocity of water, the mass transfer coefficient increases. Similarly, as can be observed in Fig. 4, when we perform the opposite operation, that is, we fix the velocity of the liquid and increase the velocity of the gas, the coefficient of mass transport also increases.
Fig. 4 – Comparison of volumetric mass transfer coefficient calculated in this study for different values of liquid and gas superficial velocities at: (a) $T = 10 \, ^\circ C$; (b) $T = 15 \, ^\circ C$; (c) $T = 20 \, ^\circ C$.

The increase in the volumetric mass transfer coefficient with increasing liquid velocity is in accordance with the discussion at the beginning of this section. As the velocity of the liquid phase rises, the number of Reynolds increases. When the values in which the flow regime happens to be in the transition range, between the laminar and the turbulent one, it translates into a greater efficiency in the mass transfer in the liquid phase of our gas-liquid system. This happens because of the increased turbulence, which promotes a higher rate of mass transfer. In addition, the more the liquid velocity increases, the greater the surface area, the greater the film thickness formed between the phases, promoting greater mass transfer.

Finally, the mass transfer coefficient was analyzed based on the changes in the temperature value of the system. Table 2 gives us enough data to realize that when the system is in fixed flow velocity conditions, the increase in temperature causes an increase in the mass transfer coefficient. This effect can be observed in Fig. 5.
At low speeds, the influence of temperature is not of great significance. However, as the operating conditions promote an increase in Reynolds number, the values of the mass transfer coefficient distances comparatively to their operating temperatures.

Analyzing the solubility of carbon dioxide in water, it increases as the temperature of the system decreases. This coefficient was expected to increase as the system temperature declined, since the availability of water to absorb the gas increases. However, it is necessary to observe other variables that may influence this parameter of mass transfer.

The increase in temperature causes the viscosities of the fluids to decrease, further causing an increase in the molecular diffusivity of the solute, which is inversely proportional to the dynamic viscosity. In addition, we have the increase of Reynolds, which promotes mass transfer. Finally, the increase in temperature decreases the density, as already discussed above, also decreasing the surface tension of the liquid and this decreases the resistance to mass transfer.

Based on these arguments, it was observed that these physical properties influence the mass transfer coefficient more than the gas solubility as a function of temperature. This is common in many mass transfer systems and occurs in this system because of the geometric characteristics of the channel are not as micro as it need to be. It is also important to note that a higher volumetric mass transfer coefficient \((k_L \cdot a)\) does not always mean a larger mass transfer, since this is also a function of the interfacial area.

4. CONCLUSION

In this study, the gas-liquid mass transfer between carbon dioxide and distilled water was studied in a microchannel with a hydraulic diameter of 0.008 m. The experimental analysis was performed at temperatures of 10, 15 and 20 degrees Celsius. Phase velocities ranged from 0.035 m/s to 0.071 m/s for water and 0.46 m/s to 1.84 m/s for carbon dioxide. The following conclusions were obtained from the results of this article:

![Fig. 5 – Temperature influence in the volumetric mass transfer coefficient to: (a) \(V_L = 0.035\) m/s; (b) \(V_L = 0.071\) m/s.](image-url)
The produced microchannel has reached the parallel regime, also known as stratified flow, in the fluid velocity ranges of 0.035 m/s to 0.071 m/s for water and 0.46 m/s to 1.84 m/s for carbon dioxide.

With the velocity of the gas phase fixed and varying the velocity of the liquid phase, the volumetric mass transfer coefficient increases. Analogously, by setting the velocity of the liquid phase and varying the velocity of the gas phase, the volumetric mass transfer coefficient also increases, though not as significant as the first case.

Increasing the temperature of the mass transfer system resulted in an increase in the volumetric mass transfer coefficient, although we know that with the decrease in the temperature of the CO2 absorption system, its solubility decreases in water. This showed that the geometric characteristics of the microchannel produced is not enough for the solubility effect to override the physical effects caused by the temperature change.

The promoted mass transfer in the microchannel produced is satisfactory, reaching from 0.059 s\(^{-1}\) to 3.80 s\(^{-1}\). To show the superiority of this system before common equipment that promote mass transfer, we can compare the values of the mass transfer coefficient volumetric. Examples such as a bubble tower, which reach 0.24 s\(^{-1}\), a spray tower with coefficient of up to 0.2 s\(^{-1}\), a spray absorber, reaching up to 1.22 s\(^{-1}\), or even a static mixer, which reaches up to 2.5 s\(^{-1}\), have lower coefficients than the microchannel of this article (Dan Li, 2015).

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