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Study of Mass Transfer Coefficient of CO_2 Capture in different Solvents using Microchannel: A Comparative Study

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Process intensification is the most demanding term in almost every process industry. Micro channels are known for the lowest production cost, high efficiency, safest-clean production rate, and energy-saving equipment. CO_2 emission is one of the big problems in several process industries. By replacing conventional equipment/channels with micro-level channels, CO_2 emission can be controlled efficiently with the comparatively high CO_2 removal rate. From prior literature, it is evident that CO_2 can be absorbed by water at low temperature and elevated pressure by physisorption which depends on Henry's coefficient but this method of absorption, CO_2 removal is comparatively low. However, the absorption rate can be enhanced by replacing the solvents, using some additive in existing solvents, varying the operating conditions such as flow rates (Q), concentration (C), etc., replacing physical absorption phenomena with chemical absorption, and by reducing the channel diameter to mini and micro level. In the present study, an effort has been made to compare the CO_2 absorption rate/removal rate in presence of different solvents like water, amino acids, mixture of amines, ammonia, NaOH, and KOH in terms of mass transfer coefficient that is based on different absorption phenomena and come-up with some guidelines. Also, an attempt is made to develop a correlation for the Sherwood number and the results obtained are compared with the predicted available co-relations in literature. This study concludes that the use of microchannels can enhance the mass transfer coefficient as well as CO_2 absorption rate several times in comparison to conventional channels and amines are proved to be a better solvent in comparison to other solvents for CO_2 removal in microchannels.

1. Introduction

In this era, industrial growth is on boom due to rapid advancement and implementation of new techniques at the same time its growth is hampered significantly due to CO_2 emissions. Thus, people are trying to develop new methods and techniques that can capture CO_2 before entering into the environment. There are several methods to remove CO_2 out of which absorption is the most successful method. Absorption of CO_2 in several solvents like water, amino acids (MEA, DEA, and MDEA), mixture of amines (Mackowiak et al., 2018), ammonia, NaOH, and KOH is possible physically as well as chemically both in conventional channel, micro-contactors, and micro-reactors. The need of different solvents arises in different contactors and reactors depends upon three factors mass transfer coefficients, reaction rate, and regeneration rate of the solvents. After an exhaustive literature survey on the use of different solvents in conventional, mini, and micro-channels, it is found that the chemisorption rate is lowest in conventional channels, and its value increases as the size of channel diameter decreases (Akkarawatkhoosith et al., 2020). The present study gives the information about the need of microchannels over conventional and mini contactors/reactors on the basis of mass transfer coefficients values. As a result, a comparison of the values of mass transfer coefficient for conventional channels ($D_h > 3$ mm are counter-current packed columns, co-current packed columns, bubble cap plate columns, sieve tray columns, bubble columns, trickle bed reactors, packed bubble columns, horizontal and coiled tube reactors, vertical tube reactors, spray columns, mechanically agitated contactors, static mixer, membrane contactors, Couette-Taylor reactor, impinging jet, mechanically agitated bubble reactors, submerged and plunging jet reactors, hydro cyclone reactors, and venturi reactors), milichannels (3 mm $\geqq D_h > 200$ μm), and microchannels (200 μm $\geqq D_h > 10$ μm) is done as shown in Figure 1 (a, b & c). Chemical absorption involves mass transfer rate and mass transfer coefficient can be enhanced by the parameters such as flow rates (Q), concentration (C), temperature (T), pressure (P) etc. (Al-Hindi et al., 2018).

2. Selection of system

2.1 Conventional, Mini, and Micro-channels

With the help of prior literature mass transfer coefficient (k_La) is represented on the scale. It can be easily seen in Figure 1 that microchannels and a special type of microchannel gives a higher value of mass transfer coefficient than the conventional and mini-channels for the absorption process.

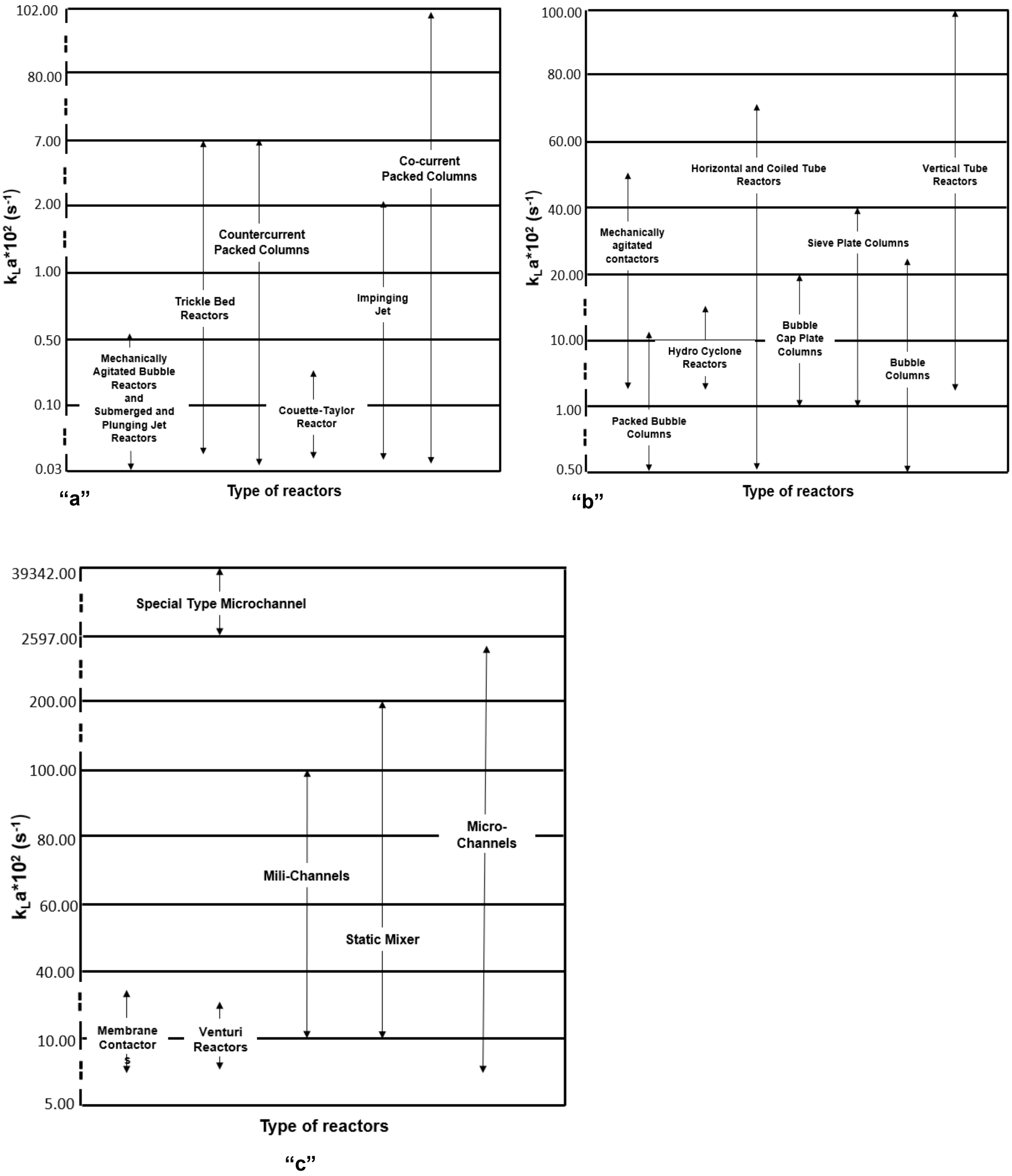

Figure 1: Schematic representation of mass transfer coefficient for different contactors and reactors.

2.2 Physical/Chemical Absorption

The absorption of CO_2 is possible in different solvents, either by the physical absorption or chemisorption process. Dong et al., (2020) shows that the chemisorption of CO_2 has an absorption rate around 3 to 10 times greater than the physical absorption. As the diameter of the channels decreases (micro-channels) chemisorption always gives higher values of mass transfer coefficient over physical absorption (Akkarawatkhoosith et al., 2020). For microreactors, the range of mass transfer coefficient for physical absorption comes under the 0.3 - 7 s^{-1} and for chemisorption, the range extended from 0.7 - 100 s^{-1} for CO_2 absorption in amines (Yao et al., 2017).

2.3 Solvents

The study of CO_2 absorption for different solvents in different channels is available widely in literature. The solvents shown in Table 1 gives good absorption rate for conventional channels. These solvents can also give good absorption rate in mini and micro-channels as well provided channels should be clean before vaporization of solvents. Because absorption of CO_2 in water, NaOH, KOH, and ammonia forms soluble salts (Carbonates and Bicarbonates) may block the channels of small diameters. Thus, the amines and its blended solutions can be proved to be good solvents for the CO_2 absorption process in microchannels and a special type of microchannel (Figure 1).

Table 1: CO_2 absorption in different type of solvents reported in literature.

Authors	Absorption	Absorption mechanism
Elhajj et al., (2020)	Water	$CO_2 + H_2O \rightleftharpoons H_2CO_3;\ H_2CO_3 \rightleftharpoons HCO_3^- + H^+;\ HCO_3^- \rightleftharpoons CO_3^{2-} + H^+$
Darmana et al., (2007)	NaOH	$CO_2 + OH^- \rightleftharpoons HCO_3^-;\ HCO_3^- + OH^- \rightleftharpoons CO_3^{2-} + H_2O$
Kraub et al., (2017)	NaOH	$CO_2 + 2OH^- \rightleftharpoons CO_3^{2-} + H_2O$
Liu et al., (2009)	Ammonia	$NH_3 + CO_2 + H_2O \rightleftharpoons NH_4HCO_3$
	Amines	
Kim et al., (2014)	MEA	$CO_2 + 2MEA \rightleftharpoons MEAH^+ + MEACOO^-$
Rinker et al., (1996)	DEA	$CO_2 + R_1R_2NH \rightleftharpoons R_1R_2NH^+COO^-$ $R_1R_2NH^+COO^- + B \rightleftharpoons R_1R_2NCOO^- + BH^+$ Where R_1R_2NH is any secondary amine e.g., DEA
Donaldson et al. (1980)	MDEA	$(R)_3N + CO_2 + H_2O \rightleftharpoons (R)_3NH^+ + HCO_3^-$ Where $(R)_3N$ is any tertiary amine e.g., MDEA
Conway et al. (2015)	Mixture of Amines	$Amine_1 + Amine_2 + CO_2 \rightleftharpoons Amine_1CO_3^- + Amine_2H^+$

2.4 Concentration

When CO_2 absorbs in any solvent, several parameters affect the absorption rate like concentration, flow rates, temperature, pressure, channel diameter, type of absorption, and solvents. Section 2.1, 2.2, and 2.3 helps to decide the channel diameter range, type of absorption (physical/chemical absorption), and solvents. Yue et al., (2009) has given sufficient data about the absorption of this gas in the microchannel Aghel et al., (2019) explained that a higher concentration of the solute gives a higher mass transfer rate in the solvent. Available data (Table 2) shows low temperature and high pressure is a favorable condition for CO_2 loading (means mole of CO_2 loading per mole of solvent)/chemisorption in monoethanolamine (MEA), diethanolamine (DEA), and methyldiethanolamine (MDEA). Table 3 represent that higher absorption efficiency can be achieved with the higher solvent rate and low value of CO_2 flow rate.

Table 2: CO_2 absorption in amines at different temperature and pressure.

Authors	Amines	Temperature (K)	Pressure (kPa)	CO_2 loading (mole/mole)
Prachi Singh (2011)	MEA	313	15.70	0.56
Guevara et al., (1993)	MEA	373	30.40	0.238
Yeh et al., (1999)	MEA	313-373	-	0.35-0.40 CO_2 kg/MEA kg
Lee et al., (1972)	DEA	298	6.89	0.57
Benamor et al., (2005)	DEA	313	10.70	0.59

Svensson et al., (2013)	MDEA	308	31.0	0.182
		318	34.6	0.186
		328	34.9	0.098
		338	37.4	0.028
Chowdhary et al., (2013)	MDEA	313	101.325	0.46-0.58

Table 3: CO_2 absorption in amines at different flow rates.

Authors	Amines	Liquid load (m^3/m^2-h)	Gas flow rate (L/h)	Removal Efficiency (%)
Aroonwilas et al., (2004)	MEA	10		90
	DEA		-	54
	MDEA			04
Aroonwilas et al., (2004)	MEA	4.8		43
	DEA		-	35
	MDEA			03
Pan et al., (2014)	MDEA	-	100, 200, 300, 400	< 40, ≈ 20, < 10, < 05

2.5 Sherwood number

In order to achieve maximum CO_2 absorption, the information is collected for the suitable system. It can be concluded from the data given in section (2.1-2.4) that chemisorption in microchannels enhance mass transfer coefficient/absorption rate by using high flow rate of solvent (amines), low flow rate of CO_2, low temperature, and high pressure. The mass transfer coefficient in terms of Sherwood number (Sh) for the mass transfer in gas liquid-phase is used in the present work. There are several co-relations available in the literature for CO_2 absorption in amines using microchannel for different ranges of Reynolds number (Re). Several researchers gave correlation of Sherwood number in terms of different factors and dimensionless numbers e.g. Sherwood number can be a function of Hatta number (Niu et al., 2009), Enhancement factor (Jiang et al., 2017), Damkholer number, and Capillary number (Yin et al., 2019) other than the Reynolds number (Yin et al., 2019), and Schmidt number (Ganapathy et al., 2013). In this work, we use the method of Buckingham Pi theorem in which Sherwood number is a function of gaseous Reynolds number (Re_G), Reynolds number of liquid (Re_L), Schmidt number for gas (Sc_G), and the ratio of liquid to gas velocity (V_L/V_G) as given by Eq. (1 & 2).

$$Sh = f.(Re_G.Re_L.Sc_G.\frac{V_L}{V_G}) \tag{1}$$

$$Sh = n_1.Re_G^{n_2}.Re_L^{n_3}.Sc_G^{n_4}.\frac{V_L}{V_G}^{n_5} \tag{2}$$

In order to obtain a more accurate Sherwood number, the Schmidt number of gas is introduced in Eq. (2) in which n_1, n_2, n_3, n_4, and n_5 are the fitting parameters and their values taken are 0.084, 0.12, 0.385, 0.3, and 1 respectively. These values are obtained with the help of experimental data, performed in the laboratory and the parameters used in the experiment are given in Table 4. The final modified form of the equation is given by Eq. (3).

$$Sh = 0.084\,.Re_G^{0.12}Re_L^{0.385}Sc_G^{0.3}\frac{V_L}{V_G} \tag{3}$$

Table 4: Parameters used for the present study of experimental and theoretical work.

Parameters	Values	Parameters	Values	Parameters	Values
Gas	CO_2	Temperature	298, 313, and 338 K	Schmidt number	5891.684, 4359.327, and 3969.112
Amine	Aq. MEA	Model	Homogeneous mixture model	Reynolds number	$13 < Re_G < 193$ $90 < Re_L < 540$
Pressure	1 atm.	V_L/V_G	10	System	Microchannel

3. Results and Discussion

Figure 2 and Figure 3 shows the comparison of modified correlation given by Eq. (3) with that of Niu et al., (2009) and Ganapathy et al., (2013) which is mentioned in Eq. (4) and (5) respectively.

$$Sh.a.d_h = 0.11 Re_G^{0.39} Re_L^{0.7} Sc_L^{0.5} \quad (4)$$

$$Sh.a.d_h = 10.201 Re_G^{0.206} Re_L^{0.218} Sc_L^{0.5} \quad (5)$$

It can be clearly seen that the modified correlation outperforms in comparison to the others and gives higher values of Sherwood number throughout the range of Re_G (0-200) (Figure 2). It is also observed that replacing Sc_L with Sc_G gives the higher value of Sherwood number and thereby higher value of mass transfer coefficient as well. Figure 3 indicates that with increase in temperature, the value of the Sherwood number increases as a result the mass transfer coefficient also increases at higher temperature values.

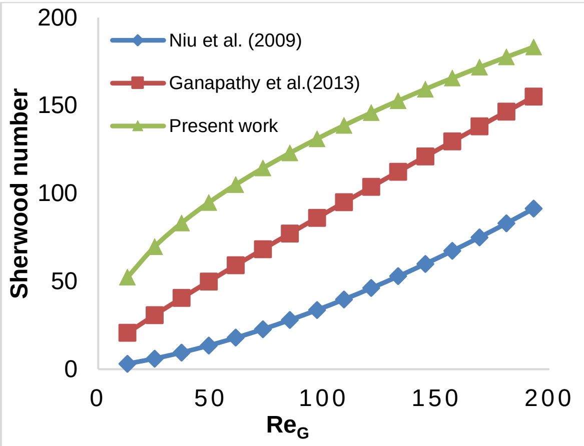

Figure 2: Comparison of present work with the other empirical co-relations.

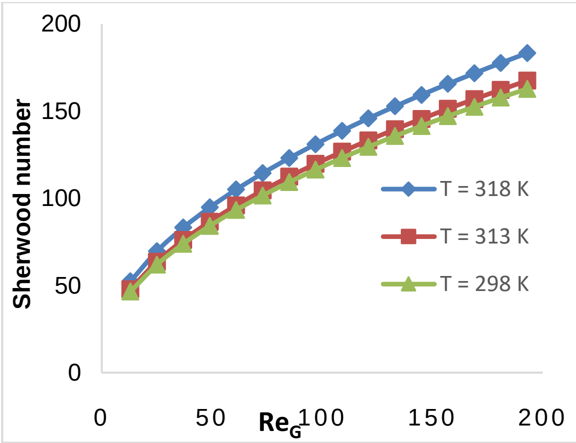

Figure 3: Study of Sherwood number at a different temperature, for CO_2 absorption in aq. MEA.

4. Conclusions

The presented work investigates the conditions to enhance the higher rate of CO_2 removal and solvent selection, and equipment's based on literature review and also suggest a modified correlation of Sherwood number that gives higher values of mass transfer coefficient. The main findings are as follows:

- In conventional, mini and microchannel, it is found that microchannels/special type of microchannels provides a high value of mass transfer coefficient.
- It is found that chemisorption with amine solution gives high rate of absorption with low flow rate of CO_2, high flow rate of solvents, low temperature and high pressure. Amines are proved to be good solvent for microchannels because it doesn't form carbonates and bi-carbonates and thereby prevent choking problems in channels.
- On that basis of developed empirical co-relation, it can say that that the selection of Sc_G over Sc_L provides much better results than the other empirical co-relations which are also verified with the help of experimental work.
- At constant pressure, as the temperature increases the value of Sherwood number increases which leads to increase in the value of mass transfer coefficient. Thus, it can be concluded that the 318 K favours the chemisorption process of CO_2 in MEA.

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