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PERFORMANCE OF HOLLOW FIBER MEMBRANE GAS-LIQUID CONTACTORS TO ABSORB CO_2 USING DIETHANOLAMINE (DEA) AS A SOLVENT

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Abstract

This study uses DEA solution to absorb CO_2 from the gas flow through the hollow fiber membrane contactors. This study aims to evaluate the performance of hollow fiber membrane contactors to absorb CO_2 gas using DEA solution as solvent through mass transfer and hydrodynamics studies. The use of DEA solution is to reduce the mass transfer resistance in the liquid phase, and on the other side, the large contact area of the membrane surface can cover the disadvantage of membrane contactors; additional mass transfer resistance in the membrane phase. During experiments, CO_2 feed flows through the fiber lumens, while the 0.01 M DEA solution flows in the shell side of membrane contactors. Experimental results show that the mass transfer coefficients and fluxes of CO_2 increase with an increase in both water and DEA solution flow rates. Increasing the amount of fibers in the contactors will decrease the mass transfer and fluxes at the same DEA solution flow rate. Mass transfer coefficients and CO_2 fluxes using DEA solution can achieve 28,000 and 7.6 million times greater than using water as solvent, respectively. Hydrodynamics studies show that the liquid pressure drops in the contactors increase with increasing liquid flow rate and number of fibers in the contactors. The friction between water and the fibers in the contactor was more pronounced at lower velocities, and therefore, the value of the friction factor is also higher at lower velocities.

Keywords: hydrodynamics, mass transfer, membrane contactor

1. Introduction

Carbon dioxide (CO_2) is usually found in natural gas where the composition varies from 30 to 80%. CO_2 in natural gas reduces the quality of natural gas, especially in terms of heating value and may cause damage to the material system especially in piping system and process equipment. At present, many of CO_2 removal processes in the industries are through amine processes using solvents such as MEA and DEA [1]. Gas absorption or desorption processes to or from liquid in gas and petrochemical industries in the 20th century are mostly carried out in absorption column which is subject to entrainment, flooding and unloading [2,3]. In addition, problems also arise as a result of gas-liquid mixing to form foam which will reduce the gas-liquid contact area drastically. These problems can be eliminated if the process is performed in hollow fiber membrane gas-liquid contactor as there no mixing between gas phase and liquid phase in the contactor.

The CO_2 removal process is conventionally carried out in conventional column such as packed tower, spray tower, and bubble column based on the reactions that occur between dissolved CO_2 and amine solutions or alkaline metal salt as absorbent/solvent. In the conventional entrainment and even flooding occurs if gas flow rate is much higher than liquid flow rate, meanwhile, unloading occurs if liquid flow rate is much higher than gas flow rate. In addition, the size of column needed for such process is large and costly in terms of installation and maintenance [4].

Membrane-based gas-liquid contactors can be used as an alternative to replace conventional absorption column. These membrane-based contactors have larger surface area for gas-liquid contact than conventional column. For example, packed absorption columns have specific surface area in the range of 30 – 300 m^2/m^3, while membrane-based contactors have specific surface area in the range of 1600 – 6600 m^2/m^3, or even can achieve as high as 33,000m^2/m^3 for hollow fiber membrane contactors [5].

Gas absorption or desorption from or to liquid through the membrane can be described as phase extraction of gas phase with or from liquid [6]. Concentration gradient between gas phase and liquid phase act as a driving force for diffusion transfer through membrane pores. There are two types of membrane, micro-porous and dense membranes. These membranes are used to prevent gas-liquid mixing in the contactors, and simultaneously allow the transfer of volatile component across the membrane pores.

Micro-porous membrane has pore size in the range of 0.1 to 10 μm, while dense membrane has pore size smaller than 50 Å. In many membrane processes, the selection is based on the membrane pore size. However, gas absorption and removal to or from liquid in the membrane-based gas-liquid contactors, this parameter is not adequate. Interaction of gas and liquid is an important parameter in the contact process so that hidrophobicity of the membrane material also needs to be known [7].

Hollow fiber membrane contactors have been verified by several researchers in a wide range of applications such as in gas absorption or removal to or from gas to liquid. In the hydrophobic membrane, the pores are gas filled so that the mass transfer resistance in the membrane phase can be ignored to the overall mass transfer resistance in the contactor [8,9]. Alkanol Amine groups are usually used in amine process to remove acid gas from natural gas stream. Yan et al. studied on CO_2 removal from flue gas using polypropylene (PP) hollow fiber membrane contactors using monoethanolamine (MEA) and methyldiethanolamine (MDEA) in the experiments. Experiment results show that CO_2 removal efficiency was above 90%. It indicates that the hollow fiber membrane contactor has a great potential in the area of CO_2 separation from flue gas when absorbent's concentration and liquid–gas pressure difference are designed elaborately [10]. This research aims to evaluate the effectiveness of hollow fiber membrane contactors in CO_2 absorption process into water and DEA solution. There are two major studies conducted in this research; mass transfer and hydrodynamics studies.

2. Experimental

There are three hollow fiber membrane contactors 1.6x40 cm used in this study. The hollow fiber membrane in the contactor has outside diameter 2 mm, pore size 0.1 μm and made of polypropylene. A 99.5% grade DEA of Merck was dissolved in reverse osmoses (RO) water to prepare 0.001 M solutions. The experimental set up is shown in Figure 1. Pure CO_2 (Samatour Gas) was used as the feed gas while RO water or aqueous 0.001 M DEA solution was employed as the absorbent. The gas passed through the lumen-side and the liquid flowed counter-currently through the shell side of the hollow fibers. In a typical experiment, the feed gas was introduced into the system from compressed gas cylinders and the flow rate was adjusted by mass flow controllers (Sierra Top-Trak 820 Series). The gas pressures were indicated by the pressure gauges at the gas cylinder. A needle valve at the outlet cylinder was used to regulate the gas pressures. Pressure drop between water or DEA 0.001 M solution inlet and outlet contactor was measured using digital manometer while the pH inlet and outlet water or 0.001 M DEA solution to or from contactor was measured using Orion Tri Star pH meter.

During the experiment water or 0.001 M DEA solution flow rates are varied in the range of 100 to 300 L/hour to give Reynolds number in the range of 3000 to 9000. Based on the CO_2 concentration difference in water and 0.001 M DEA solution before and after passing the contactor, overall mass transfer coefficients can be calculated (k_L) using Eq. (1).

$$K_L = \frac{Q_L}{A} \ln \frac{C^* - C_0}{C^* - C_1} \tag{1}$$

The mass-transfer performance is usually correlated as Sherwood number, *Sh*, and is obtained from experimental data as a function of Reynolds number, *Re*, and Schmidt number, *Sc* [11] in the form

$$Sh = a\, Re^{b}\, Sc^{c} \tag{2}$$

where a, b, and c are empirically determined constants. The constants a is related to the system geometry, whereas the constant b varies from 0.33 to 1 as reported in the literature related to the flow characteristics of fluids in the membrane contactors [9]. The exponent of Reynolds number, *b*, is an indication of mass transfer regime, where the higher the *b* the more turbulence the mass transfer regime [12].

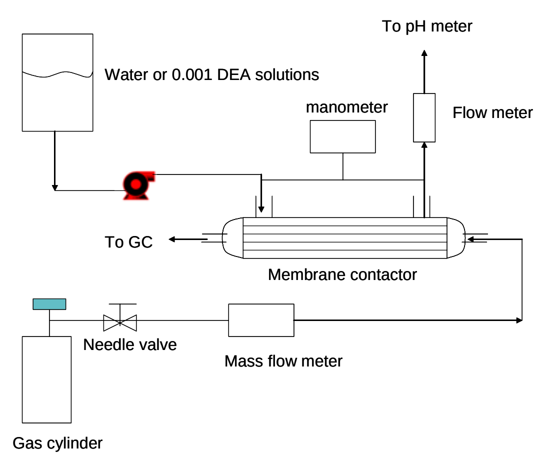

Figure 1. Experimental Setup of CO_2 Absorption in a Membrane Contactor

In the experiment the fluid viscosity is not varied so that the exponent for the Schmidt number to be c=0.33, as commonly accepted for similar systems [9], and Eq. (2) can be simplified as

$$Sh = f(\varphi)\, Re^{b} \quad (3)$$

where $f(\varphi)$ is geometry dependency as a function of contactor packing fraction, φ.

3. Results and Discussion

The results obtained from the experiments were analyzed in terms of mass transfer and fluid flow. Number of fibers and liquid flow rates in the contactor were varied to find their effects on the mass transfer performance. Meanwhile, flow and pressure drop data were analyzed to determine the degree of turbulence of the mass transfer process in the membrane contactor.

Figure 2 illustrates the variation of the overall mass transfer coefficients as a function of liquid velocity in the contactors. The overall mass transfer coefficient increased with liquid velocity since a higher liquid velocity could improve the mass transfer in liquid and therefore reduce effectively the total mass transfer resistance. It can be seen that the CO_2 absorption rates in the contactor using 0.001 M DEA solutions were around 28000 times higher than those of using water as a sorption liquid. The deteriorated performance in the contactor using water as sorption liquid was mainly attributed to the mass transfer resistance in the liquid phase at the shell side of the contactor. The same phenomenon also occurs for the flux of CO_2 through the membrane contactor as shown in Figure 3. It can also be seen that CO_2 flux rates in the contactor using 0.001 M DEA solutions were 7.6 million times higher than those that use water as sorption liquid.

Figure 4 shows the experimental mass transfer coefficients in the shell side for the contactor, using 0.001 M DEA solution as sorption liquid, as a log–log plot of Sherwood number versus the Reynolds number. An increase in Reynolds number, due to an increase in the liquid flow rate, increases the mass transfer coefficient, and consequently, the Sherwood number. This behavior is observed for liquid flow rates up to 300 liter/hour (Res = 3000). Eq. (3) can be used to fit the experimental data by linear regression, and parameters $f(\varphi)$ and b can be estimated to give:

$$Sh = 8.38\varphi^{-1.81}\, Re^{0.66} \quad (4)$$

The Reynolds exponent in Eq. (4) is 0.66, which can well fit the experimental data. A value larger than 0.33 indicates that the transverse flow module provides a component of velocity perpendicular to the hollow fiber surface, which results in a higher mass transfer coefficient than that achieved with the parallel flow module [13]. In literatures, similar Reynolds exponents were also reported by Prasad and Sirkar [14] ($Sh \approx Re^{0.6}$), Mavroudi [15] ($Sh \approx Re^{0.67}$) and Vladisavljevic [11] ($Sh \approx Re^{0.67}$). Absorption capacity ($k_L a$) of hollow fiber membrane contactor achieved in this study is as high as 36 times as absorption capacity of pack column using the same absorbent solution (DEA).

Based on hydrodynamics study, as expected, an increase in water flow rate, or fiber packing density increased energy losses in the contactor as shown in Fig. 5. Since the fiber packing densities of the modules were close, differing by less than 10%, application of the friction factor correlation to packing densities varying greatly from 18.8 to 28.1% of the pipe cross-sectional area should be made with care. Therefore, the application of the fiber friction factor correlation to other fiber types and contactor designs may require

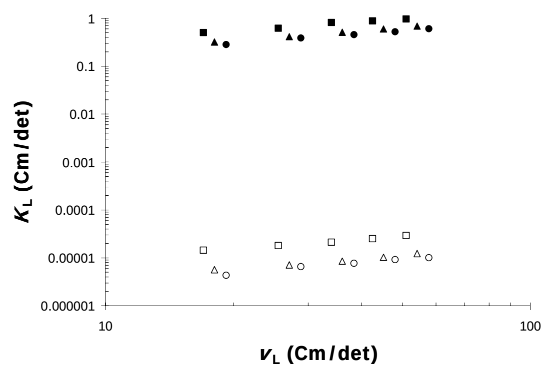

Figure 2. Overall Mass Transfer Coefficients, K_L, Under Various Number of Membrane Fibers in the Contactor of (■) 12, (▲) 15 and (●) 18 using DEA Solution as Absorbent, and (□) 12, (Δ) 15 and (○) 18 using Water as Absorbent

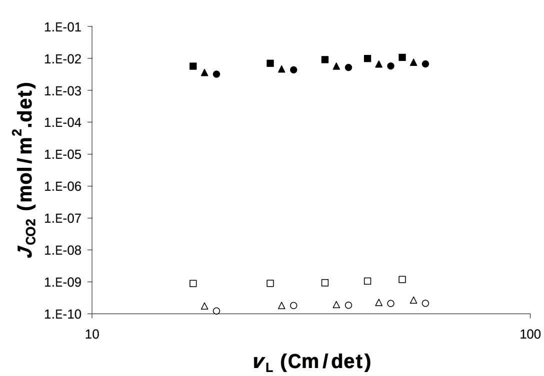

Figure 3. Fluxes of CO_2, J_{CO2}, under Various Number of Membrane Fibers in the Contactor of (■) 12, (▲) 15 and (●) 18 using DEA Solution as Absorbent, and (□) 12, (Δ) 15 and (○) 18 using Water as Absorbent

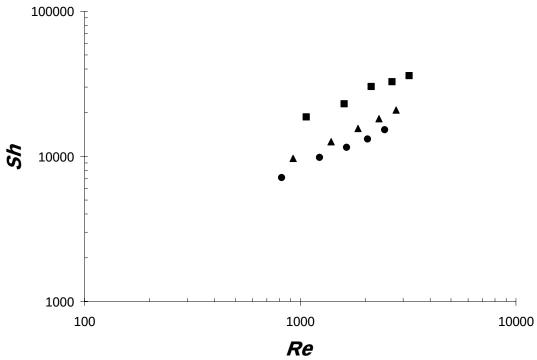

Figure 4. **Sherwood Number, *Sh*, under Various Number of Membrane Fibers in the Contactor of (■) 12, (▲) 15 and (●) 18 using DEA Solution as Absorbent, at Various Reynolds Number, *Re***

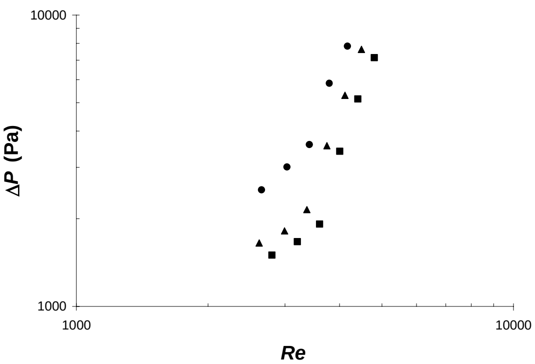

Figure 5. **Liquid Pressure Drop, Δ*P*, under Various Number of Membrane Fibers in the Contactor of (■) 12, (▲) 15 and (●) 18, at Various Reynolds Number, *Re***

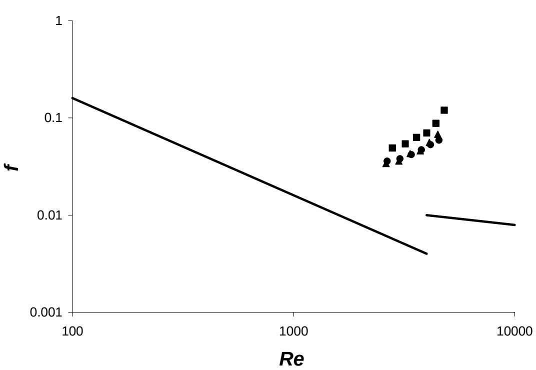

Figure 6. **Friction Factor, *f*, under Various Number of Membrane Fibers in the Contactor of (■) 12, (▲) 15 and (●) 18, at Various Reynolds Number, *Re***

further investigation. It is evident from the relative values of the fiber friction factor and the Moody chart values for smooth pipes, as shown in Fig. 6, that the fiber surface did not behave as a 'smooth-pipe' within the experimental velocity range. The friction between water and the fibers in the contactor was more pronounced at lower velocities. As a result, the value of the friction factor is also higher at lower velocities.

4. Conclusion

Experiments have been conducted to study the mass transfer and fluids hydrodynamics of CO_2 absorption through hollow fiber membrane contactor using water and 0.001 M DEA solution as sorption liquids. This experimental results show that the mass transfer coefficients and fluxes of CO_2 increase with an increase in both water and DEA solution flow rates. Increasing the amount of fibers in the contactors will decrease the mass transfer and fluxes at the same DEA solution flow rate. Mass transfer coefficients and CO_2 fluxes using DEA solution can achieve 28000 and 7.6 million times greater than using water as solvent, respectively. Hydrodynamics studies show that the liquid pressure drops in the contactors increase with increasing liquid flow rate and number of fibers in the contactors. The friction between water and the fibers in the contactor was more pronounced at lower velocities, and therefore, the value of the friction factor is also higher at lower velocities.

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