Effect of Monoethanolamine Concentration on CO_2 Removal using Continuous High Frequency Ultrasonic Reactor

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Chemical absorption is the most common technology used for the process of CO_2 removal from natural gas due to its ability to reduce the level of the acid gas to a very low level as well as higher mass transfer performance due to the chemical kinetic reaction. Higher solvent concentration resulted in higher capacity to remove CO_2, however reduces the diffusivity as well as its ability to spread on the packing due to its high solvent viscosity. This has hindered the used of high viscosity solvent in the operation of conventional packed column. However, recent technology of using high frequency ultrasonic irradiation for absorption process has shown possibility to operate high viscosity solvent due to the absence of packing in the reactor. From this work, it is shown that high viscosity solvent does not affect the mass transfer performance under ultrasonic irradiation due to the presence of acoustic streaming, fountain and atomization that reduced the film resistance thus increases the diffusivity. This work has demonstrated that this technology has high flexibility of solvent selection for the absorption process.

1 Introduction

In the chemical absorption process, CO_2 gas is removed from natural gas by reacting with chemical solvent and form intermediate compounds. Chemical absorption is mainly used in the separation process because it can reduce the level of acid gas to a very low level. There are a number of solvents that have been used for the chemical absorption of CO_2 such as amines, ammonia and potassium carbonate [1-7]. Among those solvents, amines especially monoethanolamine (MEA) is the most well-developed and commonly used for CO_2 removal from gas streams such as sour natural gas [2, 8, 9].

Other than having higher reactivity than secondary and tertiary amine, MEA also exhibited higher mass transfer performance than ammonia [1, 5]. Besides, MEA is also used for the absorption process due to its high absorption capacity and low cost [10, 11]. Higher MEA concentration provides higher amount of active MEA in the bulk solvent thus increasing the mass transfer performance. However, the increase in MEA concentration resulted in the increase of the solvent viscosity. This affects the mass transfer performance of the absorption process due to slower diffusion of amine from the bulk solvent to the gas-liquid interfacial area and also slower diffusion of the gas to the reactive boundary layer. Furthermore, for packed column operation, the use of high viscosity solvent resulted in decrease of absorption performance due to the reduced in turbulence on the packing surface as well as lower solvent ability to spread on the packing [12, 13]. In addition, high viscosity solvent caused operational issue for packed column operation [14]. Thus, the operation of the packed column for high viscosity solvent is less feasible.

Recently, a new technology of using high frequency ultrasonic irradiation has shown higher CO_2 absorption performance as compared to the packed column [4, 15-17]. This technology successfully enhanced the mass transfer process due to the physical effects of ultrasonic irradiation that increases the gas-liquid interfacial area without having any packing in the column. Due to the absence of the packing, the use of high viscosity solvent for the absorption process might be possible. Therefore, in this work, the effect of MEA concentration on the mass transfer performance of CO_2 absorption using high frequency ultrasonic irradiation in a continuous system is reported.

2 Experimental Procedure

2.1. Chemical and Materials

MEA used in the experiment is of 99% purity purchased from MERCK. CO_2 gas purchased from Air Products is of 99% purity. The natural gas (NG) purchased from Petronas Dagangan Bhd. is of 97% purity.

2.2 Experimental Set Up and Operation

The detailed experimental set up and operation has been described in our previous work [17]. The high frequency ultrasonic reactor used in this study consists of 4 unit of

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ultrasonic transducers and no packing. Table 1 shows the constant process parameters in this work. The MEA concentration was varied at 30 wt.% and 70 wt.%.

Table 1 Constant operating parameters

Parameter	Value
CO_2 concentration (%)	20
Pressure (bar)	50
Temperature (°C)	30
Ultrasonic Frequency (MHz)	1.7
Ultrasonic Power (W)	33

2.3 Overall Volumetric Mass Transfer Coefficient

The overall volumetric mass transfer coefficient calculation followed the work done by A. Setameteekul et al.(2008) as expressed in Eq. (1) and Eq. (2) [12].

$$K_G a_v = \frac{\dot{n}_{CO2}}{\Delta P_{LM}} \quad (1)$$

Where

$$\Delta P_{LM} = P\left[\frac{(y_{CO2,G}-y^*_{CO2,G})_{in}-(y_{CO2,G}-y^*_{CO2,G})_{out}}{\ln\frac{(y_{CO2,G}-y^*_{CO2,G})_{in}}{(y_{CO2,G}-y^*_{CO2,G})_{out}}}\right] \quad (2)$$

where $\dot{n}_{CO2}$ is the absorption rate in mol/m^3.hr; P is the total pressure of the system in kPa; $Y_{co2,in}$ and $Y_{co2,out}$ are the CO_2 concentration in the gas entering and leaving the absorber respectively; $y_{co2,G}$ is the CO_2 mole fraction in the gas phase while $y^*_{co2,G}$ is the equilibrium mole fraction. A log mean driving force was used since the liquid phase is assumed to be well-mixed due to the ultrasonic acoustic streaming and the gas phase is assumed as plug flow since the composition changes significantly from inlet to outlet.

2.3 Physical Properties of MEA

The viscosity of the solvent was obtained based on the study reported by Arachchige et al. (2013) while the diffusivity was acquired based on the work reported by Versteeg et al. (1988) [18, 19].

3 RESULTS AND DISCUSSION

Table 2 and Table 3 present the viscosity and diffusivity of 30 wt.% and 70 wt.% MEA. As shown in tables, increased in MEA concentration from 30 wt.% to 70 wt.% resulted in significant increase of the viscosity and reduction of the chemical diffusivity. The high solvent viscosity resulted in the lower liquid flow rate and low mixing effect. In addition, lower diffusivity resulted in lower mass transfer in the liquid phase.

Table 2 Viscosity of MEA solvent

MEA Concentration (wt.%)	Viscosity (mPa.s)
30	2.48
70	12.46

Table 3 Diffusivity of MEA solvent

MEA Concentration (wt.%)	Diffusivity ($x10^{-9}$ m^2.s^{-1})
30	1.39
70	0.60

Figure 1 shows the effect of MEA concentration on the mass transfer coefficient. As shown in the figure, higher MEA concentration resulted in higher mass transfer performance.

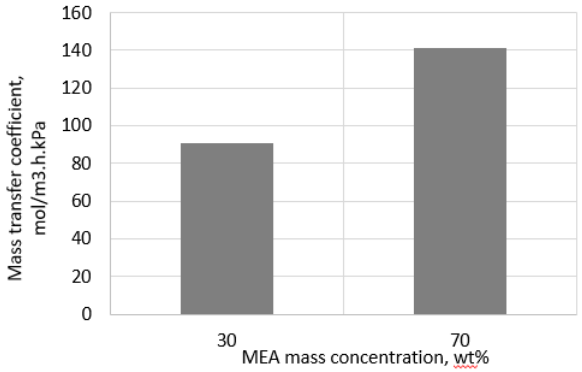

Figure 1 Effect of MEA concentration on mass transfer coefficient

This is due to the higher capacity of active MEA in the solution to remove the CO_2 from the gas stream [9, 12] . The increase of MEA concentration from 30 wt.% to 70 wt.% resulted in increased of mass transfer coefficient from 90.6 mol/m^3.kPa.hr to 141.4 mol/m^3.kPa.hr. In the study done by Setameteekul et al.(2008), the increase in MEA concentration at low temperature of 30°C resulted in low increment of mass transfer coefficient, which is due to low spreading ability in the packing surface [12]. Due to this, low MEA concentration of 30 wt.% has been used in most of the studies conducted on CO_2 absorption in a packed column [1, 6, 8].

Even though higher MEA concentration has higher active MEA to capture the CO_2, the increase in viscosity and lower diffusion coefficient resulted in lower mass transfer increment and increase the operating difficulty of the packed column. However, from this work, it has been proved that the high viscosity does not affect the mass transfer performance under ultrasonic irradiation. This is due to the presence of high frequency ultrasonic irradiation, which generates acoustic streaming, fountain and atomization which reduces the film resistance through the mixing effect, thus increase the diffusivity. Furthermore, internal design of the reactor is empty,

unlike the conventional packed column, which is filled with packing that limits the usage of high viscosity solvent.

4 CONCLUSION

This work has shown that the increase in solvent viscosity does not affect the mass transfer performance of the absorption process using the high frequency ultrasonic irradiation system. Therefore, this technology has great potential for the enhancement of absorption technology since it can provide higher mass transfer performance as well as higher flexibility of solvent selection as compared to the conventional packed column.

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