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Effect of Solution Height on Carbon Dioxide Absorption using High Frequency Ultrasonic Irradiation

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Technology for capturing CO_2 is gaining attention due to their potential in reducing the amount of CO_2 in the atmosphere and opportunities to reuse or transform the captured CO_2 into other valuable resources. This technology is commonly used during the welling for natural gas where the gas reservoir contains high concentration of CO_2. High frequency ultrasonic assisted absorption can be used to generate ultrasonic fountain that provides abundant fine droplets for CO_2 absorption. The height of the solution will affect the ultrasonic streaming velocity, which will then affect the amount of the generated droplet. In this paper, the high frequency ultrasonic assisted absorption was studied using different height of monoethanolamine (MEA) solution at varied ultrasonic power. The experiment was conducted using a pressurised batch process. The absorption rate was determined from the pressure drop profile. Based on the experiment results, the height of the solution has significantly affected the performance of the ultrasonic assisted absorption. The CO_2 absorption coefficient has been increased from 0.0388 to 0.0470 $kmol.m^{-3}.h^{-1}.kPa^{-1}$ by decreasing the height of solution from 50 mm to 25 mm under the ultrasonic power of 13.2 W. The thresholds of the ultrasonic fountain have been decreased from approximate 3 W to 1 W by decreasing the height of solution from 50 mm to 25 mm. Overall, the height of the solution in the ultrasonic reactor is one of the important factors to be considered for the ultrasonic assisted absorption process for CO_2.

1. Introduction

CO_2 capture technology plays a crucial role in reducing the amount of greenhouse gas (GHG) released to the atmosphere. CO_2 capture technology is also essential for the development of natural gas resource. Most of the undeveloped world gas reservoir contains high concentration of CO_2. The gas separation for CO_2 and natural gas is thus required to be applied at offshore site in order to inject the CO_2 back to the deep underground. Absorption is one of the most established technologies for industrial application. Chemical absorption method is widely used to capture CO_2 attributed to its high selectivity towards CO_2. Amine-based solvent, such as monoethanolamide (MEA), has been commercialised in industry due to their high absorption rate and high capacity for CO_2. The absorption process is usually conducted using packed bed (Tan et al., 2016) and bubble column method (Elhajj et al., 2014). These existed technologies involve huge unit operations and are not appropriate to be installed at the offshore site.

Several research studies have been conducted on the ultrasonic-assisted mass transfer process using low frequency ultrasonic irradiation, such as vapour-liquid mass transfer (Laugier et al., 2008), adsorption, (Schueller and Yang, 2001), and degassing (Gondrexon et al., 1997). In our previous research, high frequency ultrasonic irradiation is proposed to be one of the potential approaches in order to enhance the CO_2 absorption rate (Tay et al. 2016). The reported results show a great enhancement for the volumetric absorption rate due to the ultrasonic-assisted physical enhancement effect and this has largely reduce the size of absorption column required (Tay et al. 2016). Figure 1 shows physical enhancement effect using ultrasonic irradiation in the formation of ultrasonic streaming force (Rudenko, 1998) and ultrasonic fountain (Yasuda et al., 2005). The process involves the use of high pressure and an ultrasonic fountain caused by the ultrasonic irradiation. It is a form of physical absorption of CO_2 in a water or solution based system under elevated pressure.

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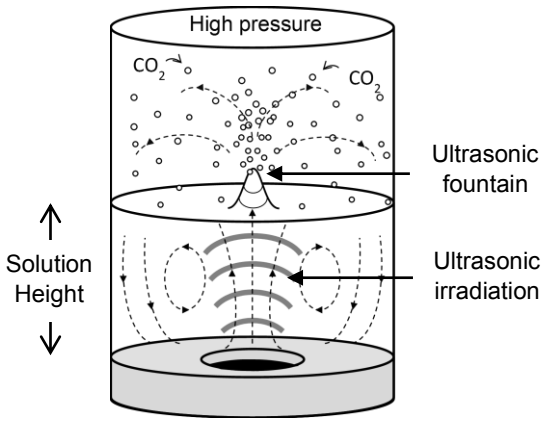


Figure 1: Schematic diagram of ultrasonic assisted absorption

It is noted that effects of the operating parameters on the performance of ultrasonic assisted absorption are still required to be studied and optimised. Based on our previous research (Tay et al. 2016), ultrasonic fountain is the dominated enhancement effect for the absorption process. The ultrasonic fountain can only be generated under a sufficient ultrasonic intensity and streaming momentum on the liquid surface. The intensity of ultrasonic irradiation will be diverged and decreased by increasing the distance from ultrasonic transducer. The height of the solution is suggested to be one of the important factors that can affect the absorption performance. The objective of this work is to study the effect of solution height on the ultrasonic fountain threshold using different ultrasonic power, which ranged from 1.5 W to 13.2 W.

The study of CO_2-MEA reaction system has been well established by Danckwerts (1979), and further improved by Astarita et al. (1983). The overall reaction for the carbamate formation in CO_2-MEA system can be expressed as in Eq(1):

$$\mathrm{CO_2} + 2\mathrm{RNH_2} \leftrightarrow \mathrm{RNH_3}^+ + \mathrm{RNHCOO}^- \quad (1)$$

In the CO_2-MEA system, the CO_2 reacted with MEA to form the carbamate. This reaction can be described by the zwitterion mechanism. The zwitterion mechanism was first proposed by Caplow (1968) and reintroduced by Danckwerts (1979). It involves two step of reactions, firstly is the formation of complex ion, called zwitterion, followed by its deprotonation. The deprotonation of zwitterion is relatively faster than the carbomate reversion rate. Therefore, the chemical reaction is considered as irreversible. The CO_2 absorption rate can be expressed as in Eq(2):

$$\dot{n}_{co2} = E_{MEA} k_l^o a_e (C_{CO2}^* - C_{CO2}) \quad (2)$$

where

k_l^o = the mass transfer coefficient without chemical reaction, which can be determined from the experiment using physical solvent;

a_e = the effective surface area;

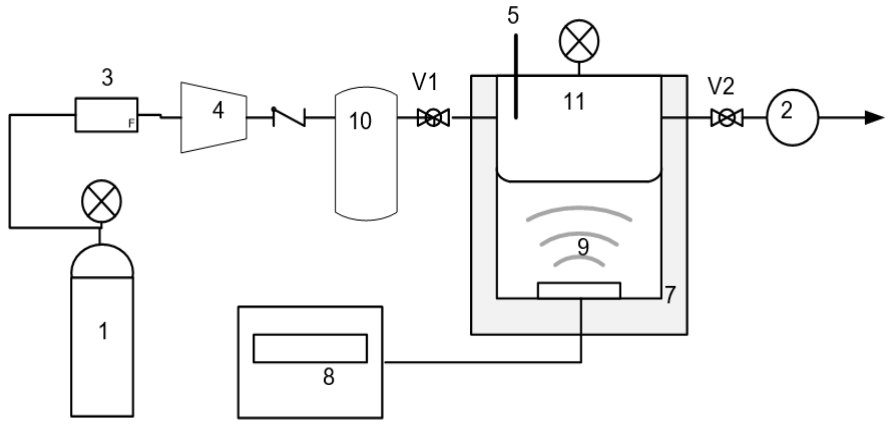
E_{MEA} = the chemical enhancement factor;

C_{CO2}^* = the concentration of CO_2 in the liquid interfacial and

C_{CO2} = the concentration of CO_2 in the liquid bulk.

2. Methodology

The experiment was set up in order to determine the absorption rate based on pressure drop profile (Tay et al., 2016). Figure 2 shows the schematic diagram of the experiment setup. Firstly, the operating temperature was maintained at 30 °C by placing ultrasonic assisted reactor in a temperature control bath. Then, the ultrasonic reactor was filled with different amount of MEA solvent (50 mL and 100 mL) with 30 wt% concentration. Subsequently, pure CO_2 gas was compressed into the gas storage vessel. Following this, the ultrasonic assisted reactor was pressurised by opening valve V1 and valve V2. The pressure of the ultrasonic assisted reactor was governed by a back-pressure regulator. The valve for the ultrasonic vessel was immediately closed after achieving the designed initial absorption pressure. Finally, the pressure of the CO_2 was recorded for every second using a data acquisition system.



1. CO_2 gas tank
2. Back pressure regulator
3. Mass flow controller
4. Compressor
5. Temperature sensor
6. Pressure sensor
7. Water bath
8. Ultrasonic power system
9. Ultrasonic piezoelectric transducer
10. Gas storage vessel
11. Ultrasonic vessel

V1 Valve 1
V2 Valve 2

Figure 2: Experiment setup for the ultrasonic-assisted chemical absorption study

For an ultrasonic-assisted mass transfer experiment, the absorption rate ($\dot{n}_{co2}$) was measured using the expression as in Eq(3):

$$\dot{n}_{co2} = K_g a_e (p - p^*) = \frac{V}{ZRT}\frac{dp}{dt} \quad (3)$$

where,
V = vapour volume in the vessel;
Z = compressibility and
p^* = the CO_2 vapour pressure on the interfacial surface, which can be considered to be zero at initial stage.

The overall mass transfer coefficient can be expressed as in Eq(4):

$$\frac{1}{K_g} = \frac{1}{k_g} + \frac{H_{CO_2}}{E_{MEA} k_l^o} \quad (4)$$

If the gas phase resistance is too small, the first term for Eq(4) can be omitted. For k_l^o determination, k_l^o was measured based on physical absorption results by using water as the solvent. The ultrasonic enhancement can be obtained using the expression as in Eq(5):

$$E_{US} = \frac{(\dot{n}_{co2})_{us}}{(\dot{n}_{co2})_{nus}} \quad (5)$$

Where,
$(\dot{n}_{co2})_{us}$ = absorption rate with ultrasonic irradiation;
$(\dot{n}_{co2})_{nus}$ = the absorption rate without ultrasonic irradiation and
E_{US} = the targeted parameter, which is required to be determined from the experiment.

3. Result and discussion

The relationship between CO_2 absorption with different ultrasonic irradiation was plotted. Figure 3 shows the pressure drop profiles using different ultrasonic power up to 13.2 W at initial pressure of 11 bar for the solution with 25 mm height. By referring to Figure 3, the pressure drop is significantly faster at the higher ultrasonic power. The results have demonstrated that the absorption rate is significantly higher at higher ultrasonic power. Ultrasonic irradiation at high power of 13.2 W, 9.2 W, 5.5 W and 3.3 W all showed significant drop of pressure along the 200 s time. The drop in pressure occurred the fastest when an ultrasonic irradiation power of 13.2 W was used. The CO_2 pressure dropped significantly when the reaction begins. Low ultrasonic

irradiation power at 1.5 W showed constant CO^2 pressure where the CO_2 pressure decrease gradually along the reaction time of 200 s. At the end of experiment, the CO_2 pressure at the ultrasonic irradiation power of 1.5 W was significantly higher than the CO_2 pressure obtained for ultrasonic irradiation employing a high power of 13.2 W, 9.2 W, 5.5 W and 3.3 W.

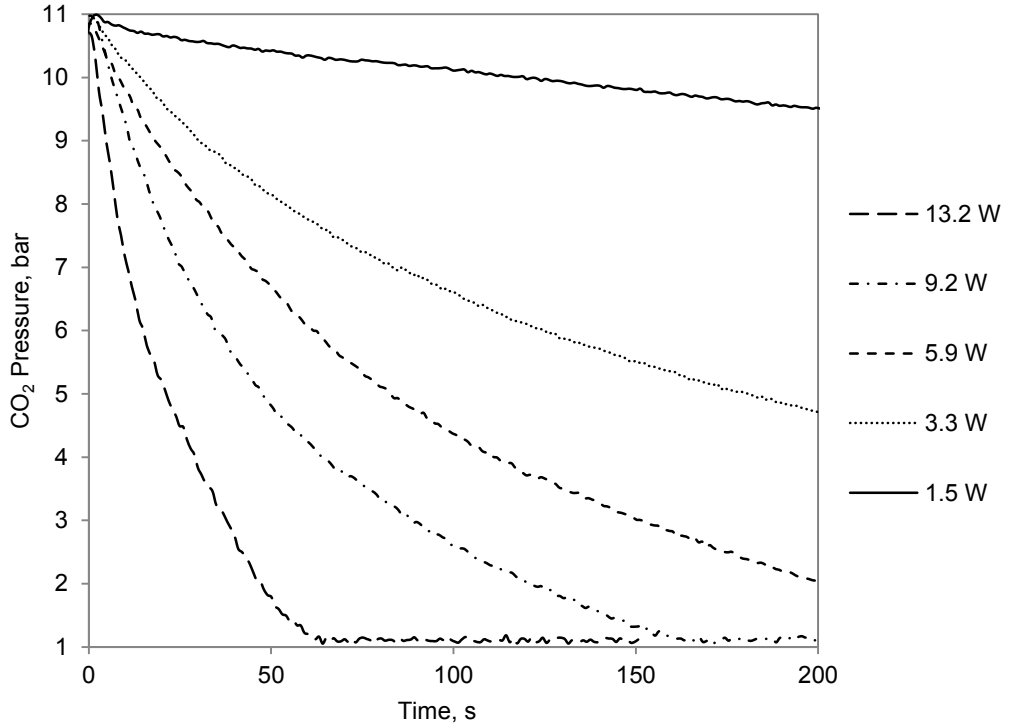


Figure 3: The time dependent pressure profiles during the CO_2 absorption processes with 11 bar and 25 mm height of solution as the initial pressure under different ultrasonic irradiation.

Figure 4 shows the mass transfer coefficient and ultrasonic enhancement using solution with 25 mm height. It can be seen from Figure 4 that the intercept of ultrasonic power indicates the thresholds of the ultrasonic power for absorption process. The ultrasonic fountain might not be formed below the thresholds of ultrasonic power. It is attributed to the insufficient ultrasonic streaming velocity in order to overcome the surface tension of the solution for the formation of the ultrasonic fountain. For the case in which ultrasonic power higher than the threshold, ultrasonic fountain can be generated, and thus, a large amount of the liquid droplet can be produced. Subsequently, the absorption rate can be greatly enhanced.

In order to determine the effect of the height of the solution on the efficiency of CO2 absorption rate, a comparison of the CO_2 absorption rate between a 25 mm and 50 mm of height of solution was carried out. Figure 5 shows the comparison result of the absorption coefficient between the solution with the height of 25 mm and 50 mm. Under the ultrasonic power of 13.2 W, an absorption coefficient of 0.0470 $kmol.m^{-3}.h^{-1}.kPa^{-1}$ is obtained using 25 mm height of solution. Lower absorption coefficient of 0.0388 $kmol.m^{-3}.h^{-1}.kPa^{-1}$ is obtained using a higher height of solution (50 mm). This shows that lower height of solution can be more effective in the absorption of CO_2 due to the shorter distance travelled by the ultrasonic irradiation. This is mainly due to the increasing ultrasonic threshold with the height of solution. The ultrasonic power threshold can be determined from the intercept of the ultrasonic power as shown in Figure 5.

Referring to Figure 5, the ultrasonic fountain thresholds for the solution height of 25 mm and 50 mm are approximate to 1 W and 3 W. The increase in ultrasonic threshold with the increasing solution height is due to the reduction of the ultrasonic irradiation intensity, which is caused by the longer distance between ultrasonic transducer and the liquid surface. Therefore, the ultrasonic streaming velocity is reduced. This has resulted in a requirement of higher ultrasonic power in order to generate the ultrasonic fountain on the surface of solution. Less intensity of ultrasonic power is required when the solution height is lower due to efficient transduction of the ultrasonic power.

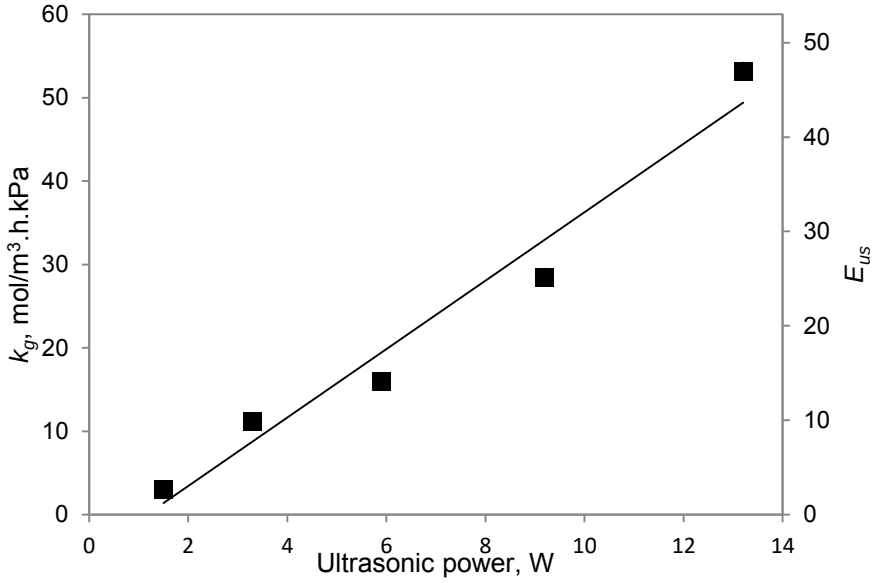


Figure 4: Mass transfer coefficient and ultrasonic enhancement of 25 mm height of solution at varied ultrasonic power.

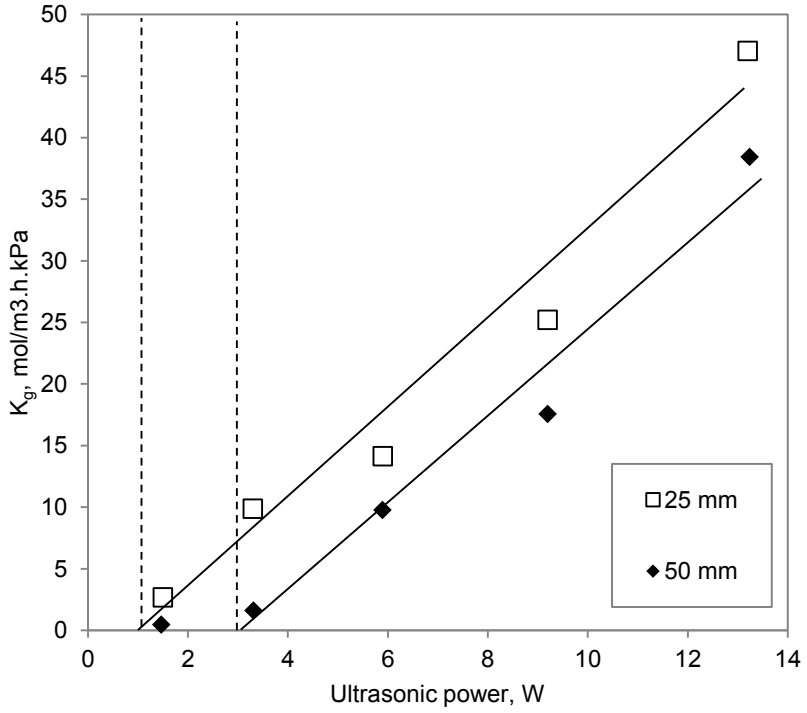


Figure 5: Comparison of absorption coefficient for 25 mm and 50 mm height of solution

4. Conclusion

This study investigated the effect of ultrasonic irradiation under different intensity on the efficiency of CO_2 absorption. The study also compared the efficiency of CO_2 absorption between a high and low solution height. In conclusion, the effect of the solution height on the CO_2 absorption has been investigated using 25 mm and 50 mm height of solution. The absorption rate was determined from the pressure drop profile. Based on the

results, the CO_2 absorption coefficient has been increased from 0.0388 to 0.0470 $kmol.m^{-3}.h^{-1}.kPa^{-1}$ by decreasing the height of solution from 50 mm to 25 mm. This shows that the CO_2 absorption coefficient is significantly affected by the height of solution of the ultrasonic fountain. The thresholds of the ultrasonic fountain have been decreased from approximate 3 W to 1 W by decreasing the height of solution from 50 mm to 25 mm. Overall, the height of the solution in the ultrasonic reactor is essential to be considered for efficient ultrasonic assisted absorption process. This study provides a better insight on the relationship among CO_2 absorption efficiency, intensity of ultrasonic irradiation and the height of solution of the ultrasonic fountain. More systematically study is still required in order to optimise the CO_2 absorption performance based on the effect of the solution height.

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