

The influence of acoustic power on chemical absorption of CO₂ using Slow Kinetic Solvent

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Abstract. Ultrasonic Irradiation (UI) is an emerging technology that is used to assist the CO₂ absorption process. Even for the slow kinetic solvents without using any chemical promoter, high-frequency UI might enhance mass transfer during the absorption process. For this purpose, it is essential to study the performance of a high-frequency ultrasonic-assisted absorption system under varied operating conditions. The ultrasonic power is considered as one of the main parameters during the absorption of CO₂. Thus, in this paper, the influence of ultrasonic power is presented using Methyl diethanolamine (MDEA) as a chemical solvent. The ultrasonic power has been varied from 0 to 15.3 W. The results tend to show a significant absorption rate enhancement for higher ultrasonic power. Moreover, they prove that the high-frequency ultrasonic absorption system has high potential to be utilized to enhance the absorption using promoter-free MDEA.

1 Introduction

Natural gas has fewer environmental issues than the other types of fossil fuels. Hence, the demand for it is expected to rise by more than 60% until 2040 [1, 2]. Depends on reservoir geological conditions, natural gas has methane with other heavier hydrocarbons, including normal butane, propane, ethane, isobutene, and significant amounts of contaminating compounds such as CO₂, H₂S, and CO [3]. To increase the calorific value, meet the gas pipeline specifications, optimize operating and capital costs, environmental purposes, or even raise the selling price, such impurities have to be separated from the gas [4–6]. Hence, removing these acid gasses is an important and necessary step during the treatment of natural gas. It can be considered a steppingstone to the continuation of using this source of energy in various applications. Widely used processes to treat natural gas can be grouped as absorption, adsorption, cryogenic, and membrane. Among them, absorption is the most established process, but despite all recent developments for this process, some particular drawbacks such as the excessive footprint or operating/maintenance issues still exist, which need to be solved [5, 6].

The use of ultrasound as one of the intensification technologies has experienced rapid developments in the last decade. Among the various aspects that accelerate these developments, the increasing demand to introduce environmentally friendly and clean technology is crucial. Ultrasound defines as a sound with frequencies from 20 kilohertz up to several gigahertz [7]. It has several applications in various research fields such as the food industry, medical treatment, cleaning, materials processing, or even enhancement of multiphase reactions

[8, 9]. However, with such favourable applications, the use of ultrasound in gas separation technologies, particularly the CO₂ absorption process, is limited [10, 11].

Generally, the CO₂ absorption process has three main steps: vapor-liquid mass transfer, liquid-liquid mass transfer, and liquid chemical reaction [12]. Due to the presence of sonochemical and sonophysical effects, all these three steps can be affected by applying ultrasonic irradiation. Figure 1 shows the ultrasound-induced physical and chemical effects. The main sonophysical effects are streaming [13], ultrasonic fountain formation [14], and atomization [15, 16]. The streaming force refers to the liquid phase acceleration effect created by the high-frequency ultrasonic irradiation. In other words, the streaming can act as an agitator that creates turbulence within the liquid and results in the improvement of mass transfer [13, 17].

The fountain is created under sufficient ultrasonic power and can improve the mass transfer process by increasing the efficient surface area. Ultrasonic atomization refers to the phenomena in which the liquid's droplet is pinched out by the extreme vibration of the gas-liquid interfacial area. The generated fine liquid droplets subsequently provide the massive surface area needed the mass transfer. It is worth noting that the size of the droplet depends on the frequency of the sound field. The higher frequencies produce smaller droplets, increasing the efficient surface area needed for the mass transfer [18, 19].

Besides the physical effect of the ultrasonic irradiation, the sonochemical effect's existence might also enhance the chemical reactions occurring during the absorption.

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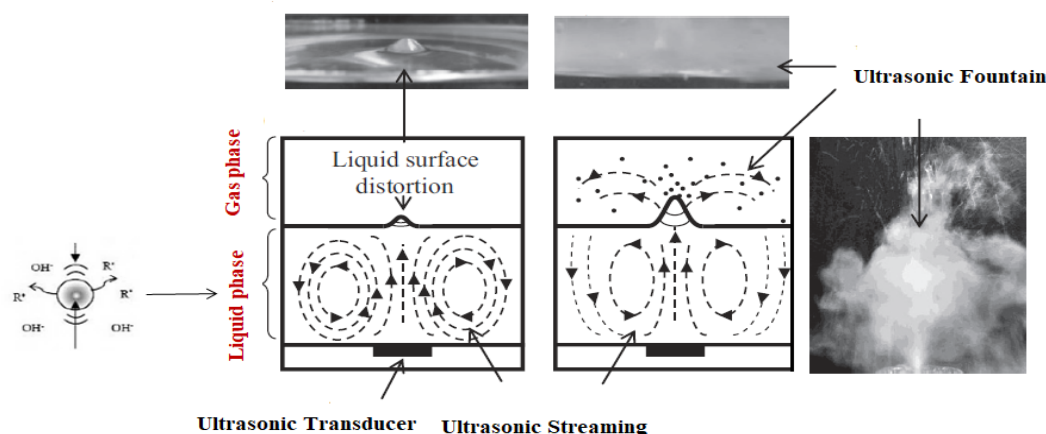


Fig. 1. Schematic diagram of physical and chemical effects created by high-frequency ultrasound [11].

Generally, the basic concept of the ultrasonic chemical effect is the generation of free reactive radicals that form due to cavitation and improve the chemical kinetic reaction rate [20–22]. Parameters such as frequency and power of the applied sound field, temperature, pressure, solvent properties, sonication time, or even the reactor's geometry may govern the sonochemical effect [8, 23].

Despite all the benefits, the relevant literature focusing on the ultrasonic-assisted absorption of CO₂ is scarcely reported. Thus, this research's key objective is to discuss the potential of using ultrasonic irradiation as an intensification technology to enhance the CO₂ absorption process. The scope of this research work focuses on investigating the effect of ultrasonic power as a critical parameter on CO₂ absorption using a slow kinetic solvent.

2 Methodology

2.1 Materials

Aqueous Methyldiethanolamine (MDEA, C₂H₇NO) solutions are extensively used to separate the acid gases from the natural gas.

The use of MDEA solutions was firstly reported by Frazier and Kohl in 1950 [24]. MDEA absorbent has a stable structure and does not quickly degrade, so it would not cause lots of corrosion problems. Despite the high CO₂ absorption capacity, since MDEA is a tertiary amine that does not have a hydrogen atom bonded to a nitrogen atom, the CO₂ reaction can occur just after the CO₂ dissolves into water to form the bicarbonate ion. The formed bicarbonate ion then undergoes an acid-base reaction with the amine to yield the overall CO₂ reaction [25, 26]. Such complicated steps ultimately lead to the low chemical reactivity with CO₂ and consequently low absorption rate, which could improve by applying ultrasonic. In the current work, MDEA (purity ≥ 99%) and CO₂ (99.9%) were respectively supplied by Merck Sdn. Bhd. and Air Sdn., Malaysia. The specifications that their suppliers provide are summed up in Table 1.

Table 1. The specifications of the used chemicals

Chemical	C ₂ H ₇ NO	H ₂ O	CO ₂
Molecular wt (gr)	119.16	18.015	44.01
Purity %	≥ 99%	99 %	99.99 %
Purification Method	-	Distillation	-
Supplier	Merck	-	Air Product

2.2 Experimental setup

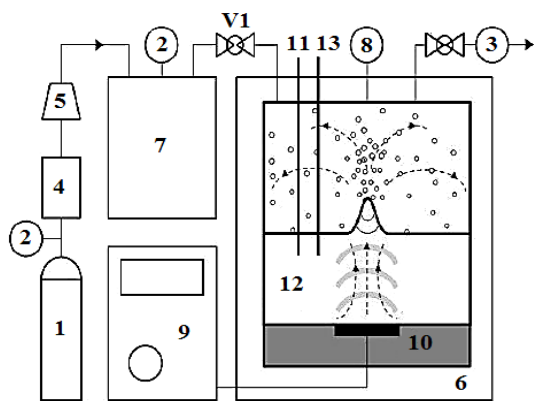
The experiments were performed in the stainless-steel batch reactor, as shown in figure 2.

The reactor had two parts a cylindrical body and a top cover. The reactor's volume, inner diameter, and height were 250 ml, 5 cm, and 17.8, respectively. The top cover had five specific outlet ports, including the temperature sensor, the pressure sensor, the gas inlet and outlet, and the probe for conductivity measurements.

The Hydroxyl radicals (·OH) are supposed to affect the conductivity; thus, in-situ measurement of conductivity is one of the possible options for elucidating the effect of radical species generated by the chemical absorption of CO₂.

A 1.7 MHz transducer with a 1.5 cm diameter was placed at the bottom of the ultrasonic reactor for direct contact with the solution. It is worth noting that the transducer converts the electrical power into ultrasonic irradiation. In the previous work, the calorimetric method was used to measure the ultrasonic power [27]. The reactor was filled with 100 ml of the MDEA aqueous solution. The solvent's temperature could be increased slightly due to the power dissipation by the ultrasound.

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1	CO ₂ gas cylinder	8	Pressure transducer
2	Pressure gauge	9	Ultrasonic power system
3	Back-pressure regulator	10	Ultrasonic transducer
4	Mass flow controller	11	Temperature transducer
5	Compressor	12	Ultrasonic reactor
6	Water bath	13	Conductivity probe
7	Gas storage vessel	V1, V2	Valve 1, 2

Fig. 2. Schematic diagram of the experiment setup [27].

Therefore, to maintain the solution temperature variation, the ultrasonic reactor settled in a water bath system. Then, the pure CO₂ gas was compressed into the gas storage vessel. After that, adjusting valve 1 and valve 2 with a back-pressure regulator enables us to pressurize and control the reactor pressure. For the ultrasonic-assisted batch experiments, the absorption rate (\dot{n}_{CO_2}) was calculated according to Equations 1:

$$\dot{n}_{CO_2} = (V/ZRT) (dp/dt) = K a_e (p - p^*) \quad (1)$$

where, V is vapor volume in the vessel, Z is compressibility factor, R is the ideal gas constant, T is temperature, and dp/dt is the pressure drop rate. Furthermore, K is the overall mass transfer coefficient, a_e is the effective surface area with ultrasonic irradiation, P is the pressure, and p^* is the vapor pressure of the CO₂ on the interfacial surface. At the initial stage p^* may be considered as zero.

3 Results & discussion

3.1 Effect of ultrasonic power on the absorption rate

Beyond the other parameters, the ultrasonic-assisted absorption system's performance is significantly dependent on the acoustic power and frequency. Thus, ultrasonic power is one of the main parameters for the current research work, reflecting on the pressure amplitude and supplies the required pressure for cavitation. It is worth noting that ultrasonic power is the output of the ultrasonic system, while the total power is the input of the electrical power system. The effects of

ultrasonic power on CO₂ pressure drop are shown in figures 3 and 4. The full-Time scale is presented in figure 3, while figure 4 displays the small scale for a clearer view. The initial pressure, temperature, and MDEA concentration were 11 bar, 343 K, and 50 wt%, respectively. The operating parameters ranges were chosen based on the limitation of the power supply, ultrasonic system, and solvent specifications [26, 28, 29].

The acoustic power that was determined calorimetrically varied from 0 to 15.3 Watt using a single ultrasonic transducer. Our previous work proved that almost 51% of the electrical power converted into ultrasonic power, which is acceptable based on the transducer specifications [27]. The selected range included the low, medium, and high power, to elucidate ultrasonic power's impact.

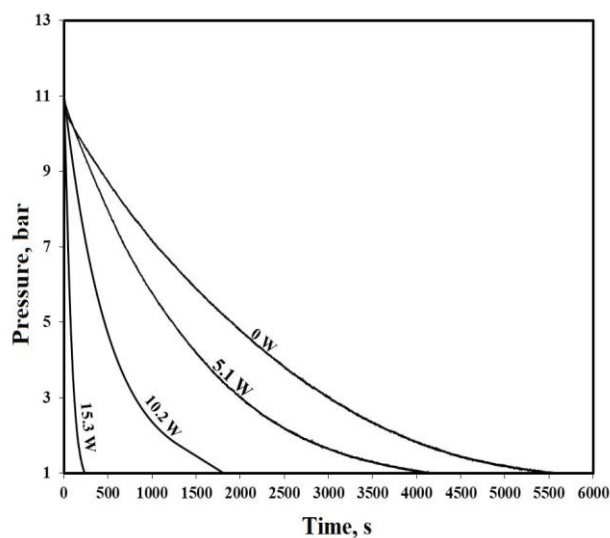


Fig. 3. The full-time scale of the experimental pressure drop profiles using various acoustic power.

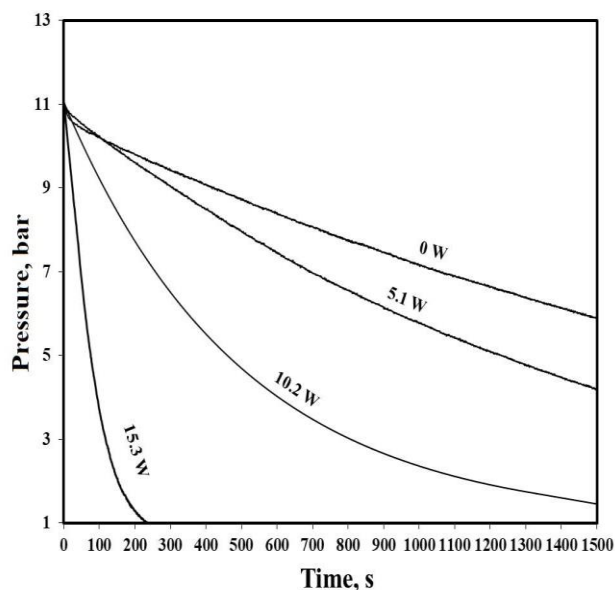


Fig. 4. The small scale of experimental pressure drop profiles under different ultrasonic power.

The results indicate that by applying higher ultrasonic power, the pressure drop is significantly faster. In the current work, the absorption rate was determined based on the CO₂ pressure drop profile. Thus, the faster pressure drop means that, the higher acoustic power would substantially enhance the CO₂ absorption rate, as shown in figure 5. This enhancement can be due to the fountain formation on one side and the free radical generation on the other side.

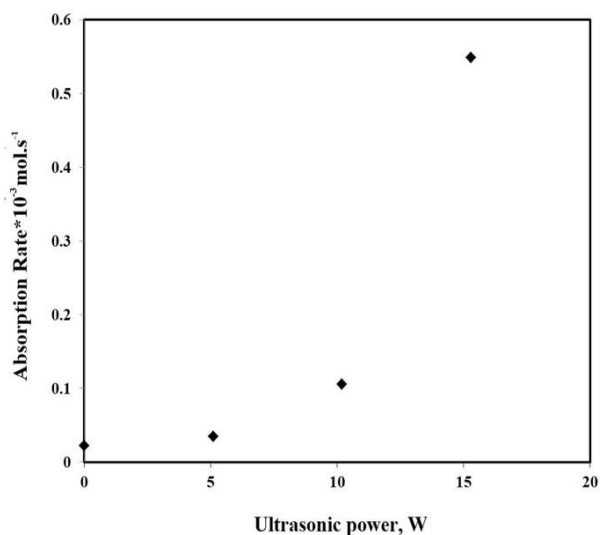


Fig. 5. Initial experimental absorption rate under different ultrasonic power.

The streaming force at lower power is not enough to overcome the liquid layer surface tension and to form the fountain. The absence of the ultrasonic fountain induces a minor change in the CO₂ absorption rate. Conversely, the streaming force is adequate for the higher ultrasonic power to force the liquid layer to form the fountain. In other words, by applying sufficient acoustic power, the ultrasonic fountain can be created. Specific amounts of liquid inside the fountain can be atomized as the fountain forms. Due to atomization, a significant number of liquid droplets cover the gas phase.

Consequently, the sufficient interfacial area between gas and liquid is provided, which leads to the mass transfer improvement. Also, the importance of ultrasonic power has been reported by Tingaud et al. [30], where they have mentioned that ultrasonic power is a crucial parameter in forming a jet-like fountain at the liquid surface. Later on, Kudo [31] and Garreton [32] found that in addition to the formation of the liquid fountain, ultrasonic atomization is also beneficial for enhancing the mass transfer process. They reported that ultrasonic atomization occurs when a liquid is irradiated with sufficient ultrasonic power, and fine liquid droplets are generated at the gas-liquid interface. The generation of these droplets is advantageous for the enhancement of the mass transfer process.

On the other side, when ultrasound is incident upon a liquid, it can trigger sonochemical active bubbles and facilitate chemical bonds breaking via the process of

sonolysis. In other words, increasing ultrasonic power enormously can disrupt bubble dynamics as it helps bubbles grow abnormally during expansion cycles. The implosion of these bubbles provides a unique environment for chemical reactions as highly reactive free radicals are formed. The generated free radicals can react quickly, recombine with other presented species in the solution or even diffuse into the liquid bulk. Thus, they can initiate, promote or alter the various reaction pathways occurring during the CO₂ absorption process [33, 34]. Though, more detailed research work is required to clarify this part. The mass transfer and CO₂ absorption rate enhancement are summarized in Table 2.

Table 2. CO₂ absorption rate comparison between the case without ultrasound and the case with high power ultrasound

Ultrasonic power, W	0	15.3
PCO ₂ , kpa	1100	1100
T, K	343.15	343.5
Absorption rate *(10 ⁻³ mol. s ⁻¹)	0.022	0.55
Volumetric absorption coefficient *(10 ⁻⁵ mol.pa-1.m ³ . s ⁻¹)	0.008	0.199

4 Conclusion

High-frequency ultrasound is a new promising technique to assist the absorption process of CO₂. Particularly, the use of an ultrasonic-assisted CO₂ absorption system has the high potential to improve the absorption rate of the slow kinetic solvents without adding any rate promoters. The efficiency of the ultrasonic-assisted absorption cell extensively relies on acoustic power and frequency. The influence of acoustic power on the CO₂ absorption rate by using Methyl-diethanolamine was discussed in this paper.

The batch experimental results indicate the high-frequency ultrasonic irradiation of 1.7 MHz can significantly boost the CO₂ absorption rate in un-promoted MDEA solvent by increasing the ultrasonic power. Based on the findings, the high power ultrasonic-assisted absorption system using 50 wt% un-promoted MDEA provides 25-fold times higher absorption rate than the case without ultrasonic irradiation. This improvement is due to both the physical and chemical effects of ultrasonic irradiation. The sonophysical effect improves the absorption through the fountain formation and atomization, while because of the formation of highly reactive radical species, the sonochemical effect alters the absorption mechanism and reaction pathway.

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References

1. B. Shimekit and H. Mukhtar, *Adv. Nat. Gas Technol.* (2012)
2. S. Kumar, J. H. Cho, and I. Moon, *Int. J. Greenh. Gas Control* **20**, 87 (2014)
3. L. S. Tan, K. K. Lau, M. A. Bustam, and A. M. Shariff, *J. Nat. Gas Chem.* **21**, 7 (2012)
4. R. K. Abdulrahman and I. M. Sebastine, *J. Nat. Gas Sci. Eng.* **14**, 116 (2013)
5. F. Ahmad, K. K. Lau, A. M. Shariff, G. Murshid, and L. K. Keong, *Comput. Chem. Eng.* **36**, 119 (2012)
6. A. A. Olajire, *Energy* **35**, 2610 (2010)
7. T. Leong and S. Kentish, *THE FUNDAMENTALS OF POWER ULTRASOUND-A REVIEW* (2011)
8. R. J. Wood, J. Lee, and M. J. Bussemaker, *Ultrason. Sonochem.* **38**, 351 (2017)
9. P. R. Gogate, V. S. Sutkar, and A. B. Pandit, *Chem. Eng. J.* **166**, 1066 (2011)
10. W. H. Tay, K. K. Lau, and A. M. Shariff, *Sep. Purif. Technol.* **183**, 136 (2017)
11. W. H. Tay, K. K. Lau, and A. M. Shariff, *Ultrason. Sonochem.* **33**, 190 (2016)
12. H. Karlsson and H. Svensson, in *Energy Procedia* (Elsevier Ltd, 2017), pp. 2009–2023
13. B.-G. Loh, S. Hyun, P. I. Ro, and C. Kleinstreuer, *J. Acoust. Soc. Am.* **111**, 875 (2002)
14. Z. Xu, K. Yasuda, and X. Liu, *Ultrason. Sonochem.* **32**, 241 (2016)
15. K. Yasuda, Y. Bando, S. Yamaguchi, M. Nakamura, A. Oda, and Y. Kawase, in *Ultrason. Sonochem.* (Elsevier B.V., 2005), pp. 37–41
16. K. Yasuda, H. Honma, Z. Xu, Y. Asakura, and S. Koda, *Jpn. J. Appl. Phys.* **50**, 07HE23 (2011)
17. S. J. Lighthill, *J. Sound Vib.* **61**, 391 (1978)
18. B. Avvaru, M. N. Patil, P. R. Gogate, and A. B. Pandit, *Ultrasonics* **44**, 146 (2006)
19. T. Kudo, K. Sekiguchi, K. Sankoda, N. Namiki, and S. Nii, *Ultrason. Sonochem.* **37**, 16 (2017)
20. Y. G. Adewuyi, *Ind. Eng. Chem. Res.* **40**, 4681 (2001)
21. L. H. Thompson and L. K. Doraiswamy, *Ind. Eng. Chem. Res.* **38**, 1215 (1999)
22. P. Riesz, D. Berdahl, and C. L. Christman, *Environ. Health Perspect.* **VOL. 64**, 233 (1985)
23. N. Pokhrel, P. K. Vabbina, and N. Pala, *Ultrason. Sonochem.* **29**, 104 (2016)
24. H. D. Frazier and A. L. Kohl, *Ind. Eng. Chem.* **42**, 2288 (1950)
25. S. Mokhatab, W. A. Poe, and J. Y. Mak, in *Handb. Nat. Gas Transm. Process.* (Elsevier, 2019), pp. 231–269
26. P. Pal, A. AbuKashabeh, S. Al-Asheh, and F. Banat, *J. Nat. Gas Sci. Eng.* **24**, 124 (2015)
27. F. Shokrollahi, K. K. Lau, W. H. Tay, and L. S. Lai, *Int. J. Eng. Technol.* **7**, 106 (2018)
28. A. Rosli, A. Latif Ahmad, J. K. Lim, and S. C. Low, *J. Phys. Sci.* **28**, 121 (2017)
29. C. Gouedard, D. Picq, F. Launay, and P. L. Carrette, *Int. J. Greenh. Gas Control* **10**, 244 (2012)
30. F. Tingaud, S. Ferrouillat, S. Colasson, A. Bontemps, and O. Bulliard-Sauret, *Ultrason. Sonochem.* **20**, 1046 (2013)
31. T. Kudo, K. Sekiguchi, K. Sankoda, N. Namiki, and S. Nii, *Ultrason. Sonochem.* **37**, 16 (2017)
32. L. Gaete-Garretón, D. Briceño-Gutiérrez, Y. Vargas-Hernández, and C. I. Zanelli, *J. Acoust. Soc. Am.* **144**, 222 (2018)
33. M. Lupacchini, A. Mascitti, G. Giachi, L. Tonucci, N. d’Alessandro, J. Martinez, and E. Colacino, *Tetrahedron* **73**, 609 (2017)
34. S. Merouani, O. Hamdaoui, Y. Rezgui, and M. Guemini, *Ultrason. Sonochem.* **20**, 815 (2013)