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Intensification of CO_2 Capture by Monoethanolamine Solution in a Rotating Packed Bed Reactor Equipped with High Frequency Ultrasonic Transducers

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ABSTRACT

In this study, a carbon dioxide (CO_2) absorption process in a typical rotating packed bed (RPB) reactor equipped with blade packing and under a high frequency ultrasonic field has been studied. The utilized ultrasonic transducers were ultrasonic atomizer humidifiers with a frequency of 1.7 MHz. This reactor takes advantage of both controllable high gravitational force and induced effects of high frequency ultrasound, simultaneously, in a small volume. The overall volumetric gas side mass transfer coefficient (K_Ga) with and without ultrasound was investigated. The effects of different parameters such as rotational speed (400-1600 rpm), liquid flow rate (20- 120 L/h), monoethanolamine (MEA) concentration (1- 4 mol/L), gas flow rate (2500- 4000 L/h), and CO_2 concentration (1- 4 vol%) were investigated in the absence and presence of ultrasound. The obtained results showed that the removal efficiency increased with increasing gas and liquid flow rates, and rotational speed, as well as MEA concentration. With increasing CO_2 concentration, absorption efficiency decreased. The average arithmetic value of the relative volumetric gas-side mass transfer coefficient was enhanced 11.4% under the ultrasonic field. Moreover, the average CO_2 removal efficiency was enhanced from 27.4 % in the absence of ultrasound to 29.8% in the presence of ultrasound. Therefore, high frequency ultrasound can enhance CO_2 absorption, even in high efficiency equipment like RPBs.

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1. Introduction

Process intensification (PI) involves instruments and developing methods that leads to significant improvement in equipment and process designing and a substantial decrease in equipment size, and energy consumption, which finally results in lower cost and beneficial production, and higher production capacity [1]. PI is divided into two parts: equipment and methods. Rotating Packed Bed (RPB) reactor and ultrasonic waves are one of the most popular equipment and method of PI technologies [2]. The RPB as a high efficiency PI equipment was innovated in 1981 by Mallinson, and Ramshaw [3]. This device replaces a high centrifugal force with gravitational force and achieves high efficiency in a small volume. The RPB technology is suitable for separation processes like absorption [4], extraction [5], distillation [6], and other processes such as polymerization [7], oxidation [8] and nanoparticle synthesis [9]. By taking advantage of both high centrifugal force and rotational speed, the RPBs can be employed in systems involving viscous liquids and in reactions with short contact time [10]. In RPBs, due to high centrifugal forces, the liquid film changes into tiny droplets which leads to the enhancement of the interfacial area and, finally increase in the mass transfer rate [11].

In comparison with conventional packed beds, the RPBs have specific advantages such as higher efficiency [12, 13], significantly smaller apparatus

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volumes [13, 14], and lower energy consumption. As a result, from an economic point of view, The RPBs have the lower investment and operational cost comparing the conventional packed beds [13, 14]. Although various successful applications of RPBs have been reported in the industrial scale, the technology has not yet reached its full industrial maturity [15, 16]. The number of industrial-scale applications of RPBs is very limited [17], but rapidly growing, especially in gas absorption processes. A possible reason for the limited number of industrial applications of RPBs is the low number of scale-up investigations [17]. The scale-up investigation of the RPBs has been reported in a few studies [17, 18].

As mentioned, ultrasound is among the most popular methods of PI technologies [2]. Ultrasound is a mechanical wave with a frequency above 20 kHz [19-21]. Acoustic cavitation is the significant effect of the wave propagation in the liquid medium, which induces acoustic streaming, microstreaming and heating [22, 23]. Acoustic cavitation is the process of bubble formation, growth, and collapse due to variations in the wave pressure [24]. The size of the cavitation bubbles depends on the wave frequency [25, 26]. There are two types of cavitation bubbles: stable, and transient cavitation bubbles [27, 28]. Transient cavitation bubbles exist for less than one cycle [27], which are created by low frequency waves. Transient bubbles are not very small and they can decompose to smaller bubbles.

The frequency spectrum, above 1 MHz, is named “low power ultrasound” (< 10W) which does not affect the propagation medium, and is mainly utilized for medical diagnosis [27]. These waves increase turbulence and also generate stable cavitation bubbles, acoustic streams, and acoustic fountains [28, 29]. Stable bubbles do not collapse for a long time [27, 28]. The formation of the stable bubbles creates another important effect called microstreaming. The creation of the high speed unidirectional currents of the stable bubbles causes the momentum gradients and the fluid currents, which are called the acoustic streaming [27] These streams create the acoustic fountain (deformation) at the gas-liquid interface [26, 27, 29]. Whatever the ultrasonic frequency is higher, the liquid droplet will be smaller, and the interfacial area will increase, which causes enhancement in mass transfer rate [30] or intensification of the physical and chemical processes. Using high frequency ultrasound is an efficient way to enhance absorption processes.

Today, industrialization and the use of fossil fuels have caused the emission of greenhouse gases and global warming. Therefore, it is necessary to use alternative energy sources and reduce greenhouse gas emissions [31]. CO_2 is one of the most important emitted greenhouse gases. Therfore, CO_2 capture is an important issue, and finding new and efficient techniques to intensify it is crucial. As CO_2 absorption is a significant factor for reducing global warming, researchers are looking for efficient methods to capture and store CO_2 [32], such as absorption [33]. As mentioned above, respect to the specific characteristics of the RPB in absorption and separation, application of RPB in CO_2 capturing attracted wide attention [34-40]. In 2011, Cheng and Tan [34], studied about removal of CO_2 from indoor air by alkanolamines in an RPB reactor for a long time to decrease the CO_2 concentration from 1000 ppm to 100 ppm. They showed that the RPB was more efficient and preferable than a conventional packed bed. In 2014, Kang et al. [35] investigated the CO_2 capturing in an RPB using diluted ammonia. They found that the height of the transfer unit (HTU) of the RPB was smaller than a conventional packed bed, which was equivalent to that and ultimately resulted in a reduction in equipment size and costs. In 2017, Wu et al. [36], studied CO_2 capture from power plants by piperazine (PZ) and diethylenetriamine (DETA) solution in an RPB. Their experiments showed that the PZ and DETA solutions compared to traditional monoethanolamine (MEA) solutions were more efficient. In 2007, jassim et al. [37] studied about CO_2 absorption and desorption in an RPB by aqueous solution. They found that the CO_2 penetration in an MEA concentration up to 30 %w decreased.

Applications of ultrasound for increasing CO_2 absorption and saving CO_2 capture and storage (CCS) costs have been studied in recent years [2, 30, 41]. In 2017, Tay et al. [41] showed that the CO_2 absorption rate into MEA solution increases dramatically in the presence of high frequency ultrasound. In addition, they suggested that the high frequency ultrasounds lead to a high mass transfer coefficient in a compact design due to the formation of acoustic fountains and convective dynamics. In 2017, Tay et al. [30] studied the CO_2 absorption with a potassium carbonate without any promoter in the presence of high frequency ultrasound. They found that, the reaction rate assisted by ultrasonic waves was about 32 times higher than that without ultrasound. In addition, the reaction time to achieve 0.9 loadings (CO_2 mol/ K_2CO_3 mol) reduced to 400s. Finally, they showed that high frequency ultrasonic waves are an effective equipment for increasing the absorption rate in a slow kinetic reaction. In 2018, Luo et al. [2] studied the effective interfacial area and volumetric liquid-side mass transfer coefficient (K_La) using a NaOH-CO_2 mass transfer system in an RPB reactor under 20 kHz ultrasound. They reported that the $\overline{K_L a}$ under ultrasound field enhanced 5.5% compared with no ultrasound conditions.

In the above studies, the use of RPB and ultrasound in the CO_2 absorption process has been investigated separately, except in the work of Luo et al. [2]. In their study, low frequency ultrasound was used in an RPB reactor. Continuing our previous work [42] and the work of Luo et al. [2], we decided to use high frequency ultrasonic waves in an RPB with different packing and different CO_2 absorbent. For this purpose, ultrasonic transducers with a frequency of 1.7 MHz were selected, which are known as ultrasonic atomizer humidifiers. These transducers creat cold boiling [28].

In this study, we aimed to investigate the simultaneous effect of an RPB equipped with blade packing as PI equipment, and high frequency ultrasound as a PI method on the CO_2 absorption process. Therefore, measurement of CO_2 absorption percent (E), and overall volumetric gas-side mass transfer coefficient (k_Ga) under different operational conditions was performed using an MEA-CO_2 mass transfer system.

2. Material and Experimental Methods

2.1. Development of RPB Reactor

In this study, an RPB reactor with a plexiglass casing was used. The rotor is equipped with 12 stainless steel blades. Four 1.7 MHz ultrasonic transducers with a diameter of 20 mm were embedded at the bottom of the reactor. To ensure the presence of liquid on the transducers and to benefit from ultrasound effects such as cavitation, acoustic fountain, and fogging, the rotor was installed upside down. Another advantage of this structure is comfortable sealing. Two distributers with a hole diameter of 1 mm were used to distribute the liquid in the packing symmetrically. Figure 1 shows the different parts and dimensions of the RPB reactor.

2.2. Experimental Method

The schematic of the experimental setup for the CO_2 absorption process is depicted in Figure 2. The utilized absorbent was MEA with a concentration of 1-4 mol/L and a volume flow rate of 20-120 L/h. The gas mixture containing CO_2 and N_2, with a volume flow rate of 2500-4000 L/h, entered the reactor and reacted with the absorbent. The inlet CO_2 concentration was 10000-40000 ppm. The outlet gas was analyzed by CO_2 sensor (s8 alarm 5% (senseair)). The CO_2 amount in the outlet gas was used to calculate the overall volumetric gas-side mass transfer coefficient by the following equation [42– 45]:

$$K_G a = \frac{Q_G}{\pi \times (R_o^2 - R_i^2) \times Z_b} ln \frac{C_i}{C_o} \quad (1)$$

where Q_G is the volumetric gas flow rate, Z_b is the height of packing, and R_i and R_o are the inner and outer diameter of packing, respectively. Moreover, C_i and C_o are the CO_2 concentration in the inlet and outlet gas streams. The CO_2 removal efficiency can be determined by equation (2):

$$E\ (\%) = \frac{C_i - C_o}{C_i} \times 100 \quad (2)$$

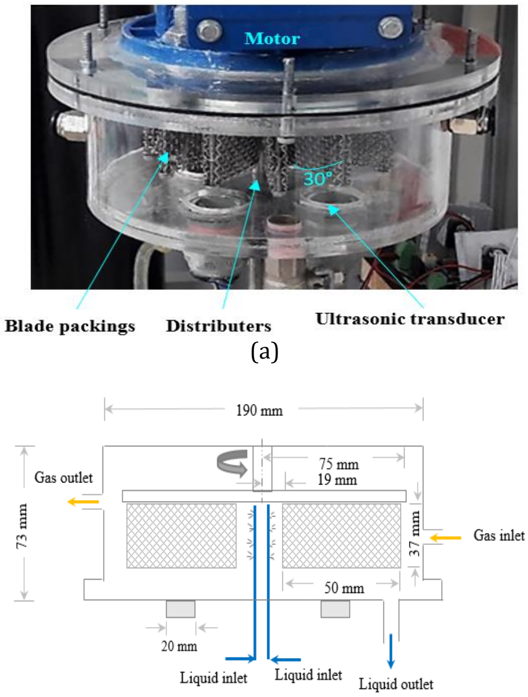
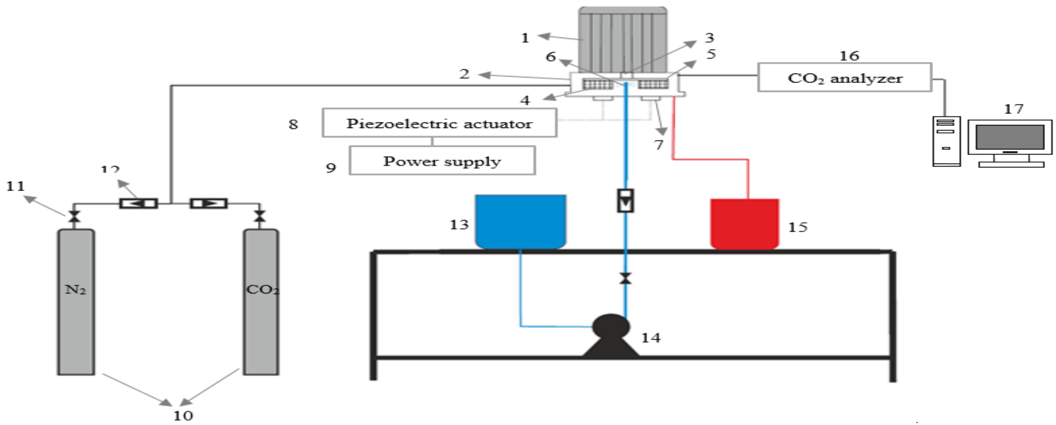


Figure 1. (a) The real view of the RPB reactor, and (b) schematic view of the reactor and dimensions (mm).



1. Motor, 2. The casing of the RPB reactor, 3. Shaft, 4. Blade packings, 5. Rotating bed, 6. Distributers, 7. Ultrasonic transducers, 8. Piezoelectric actuator, 9. Power supply, 10. CO_2 and N_2 gas cylinders, 11. Valves, 12. Rotameter, 13. MEA solution tank, 14. Pump, 15. Waste tank, 16. CO_2 analyzer,17. Computer.

Figure 2. Schematic diagram of the CO_2 capture system

The operating pressure of the RPB reactor was 1 atm. All experiments were carried out at 25±2 °C with three repetitions. The ultrasonic power was measured by a calorimetric experiment in a batch flow mode [19, 20]. The concentration of CO_2 in the outlet stream was measured with a standard deviation of less than 3 %. Effects of the rotational speed of the bed (N), liquid flow rate (Q_L), absorbent concentration (C_{MEA}), gas flow rate (Q_G), and CO_2 concentration on K_Ga and E were investigated.

3. Result and Discussion

3.1. Effect of Rotational Speed on K_Ga

Figure 3 presents the effect of N on K_Ga. In these experiments, Q_G, C_{MEA}, CO_2 percent, Q_L, and P_{us} were set at 3500 L/h, 1 mol/L, 2 vol%, 50 L/h, and 22.5 W, respectively. As can be seen, enhancing the rotational speed increases the volumetric gas side mass transfer coefficient. To describe this issue, we can mention that, higher rotational speed causes more collisions between rotating packed bed and absorbent solution that leads to thinner liquid films and their better distribution. These tiny droplets and the thin liquid film increase the interfacial area and improve the overall mass transfer coefficient. Moreover, the CO_2 absorption increased in ultrasonic presence. These waves cause creating tiny droplets, more interfacial area, and finally increase the removal efficiency. Also, a convergence in the diagram at high rotational speeds, it means that the centrifugal force is overcoming ultrasonic effects. For a better evaluation of the effects of high frequency ultrasound on CO_2 absorption, the relative volumetric gas-side mass transfer coefficient, σ_{K_Ga}, is defined as follows [2]:

$$\sigma_{K_Ga}(\%) = \frac{K_Ga_{us} - K_Ga_0}{K_Ga_0} \times 100 \quad (3)$$

where 0 and us are referred to as no ultrasound and ultrasound conditions. In these experiments, for N of 400 –1600 rpm, the values of σ_{K_Ga} change from 32.3 % to 3.3 %, while E varies from 14.8 % to 28.9% for non-ultrasonic assisted RPB reactor (NUAR) to 19.1%-29.7% for ultrasonic assisted RPB reactor (UAR).

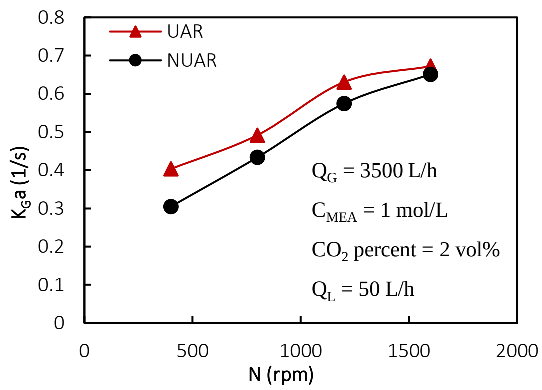


Figure 3. Effect of rotational speed on K_Ga.

3.2. Effect of Liquid Flow Rate on K_Ga

Figure 4 showed the effect of Q_L on K_Ga, while Q_G, C_{MEA}, CO_2 percent, N, and P_{us} were set at 3500 L/h, 1 mol/L, 2 vol%, 1200 rpm, and 22.5 W, respectively. K_Ga and the removal efficiency are enhanced by increasing the liquid flow rate due to decreasing liquid-side mass transfer resistance. In addition, the wetting of each unit of packing increases which is favorable for CO_2 absorption. On the other hand, increasing the liquid flow rate also has a negative aspect that reduces the residence time, but it's negligible. Also, the acoustic fountain formed by ultrasonic waves can return the absorbent solution from the bottom of the reactor to the packing zone and increases each unit of wetted packing, that is desirable for capturing process. In these experiments, for Q_L of 20 –120 L/h, the values of σ_{K_Ga} change from 16.9 % to 5.0 %, while E varies from 24.1 %– 33.2 % for NUAR to 27.6%- 34.6% for UAR.

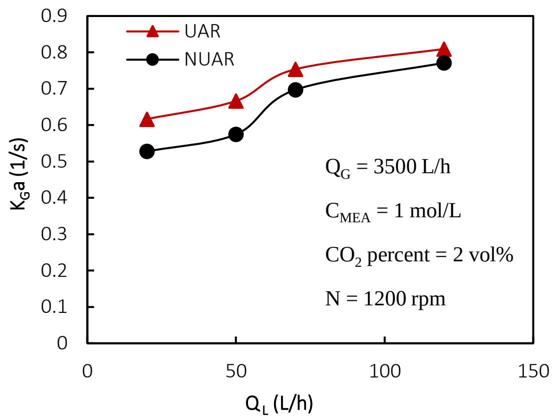


Figure 4. Effect of liquid flow rate on K_Ga.

3.3. Effect of MEA Concentration on K_Ga

Figure 5 summarizes the effect of MEA concentration on K_Ga, while Q_G, Q_L, CO_2 percent, N, and P_{us} were set at 3500 L/h, 50 L/h, 2 vol%, 1200 rpm, and 22.5 W, respectively.

The results show that by increasing the C_{MEA}, the K_Ga and removal efficiency enhance. For the constant amount of CO_2, there is more amount of MEA that causes enhancement in mass transfer driving force and finally a significant increase in removal efficiency.

On the other hand, ultrasound can spread the MEA droplets, which still didn't react completely with CO_2, to the packing zone by the acoustic fountain and fogging phenomena, and finally assist in improving the removal efficiency and K_Ga.

In these experiments, for C_{MEA} of 1- 4 mol/L, the values of σ_{K_Ga} change from 9.8 % to 6.7 %, while E varies from 26.0 %– 37.7 % for NUAR to 28.1%- 39.6% for UAR.

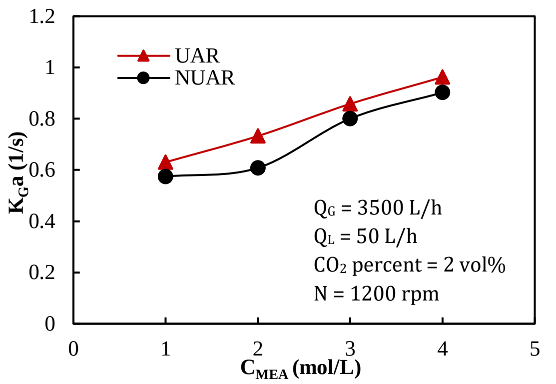


Figure 5. Effect of MEA concentration on K_Ga.

3.4. Effect of Gas Flow Rate on K_Ga

Figure 6 shows the effect of Q_G on K_Ga. In this section, CO_2 percent, Q_L, C_{MEA}, N, and P_{us} were 2 vol%, 50 L/h, 1 mol/L, 1200 rpm, and 22.5 W, respectively. The gas flow rate enhancement has both positive and negative effects. Decreasing the gas-side mass transfer resistance is the positive effect and decreasing the residence time is the negative effect, but it's negligible. Moreover, ultrasound can enhance the K_Ga. Ultrasonic waves return some of the MEA solution to the packing zone, which reduces the negative effect of gas flow rate enhancement (residence time) by increasing the volume of liquid in the packing zone, in the time interval between gas entering and exiting the reactor. In these experiments, for Q_G of 2500 -4000 L/h, the values of σ_{K_Ga} change from 9.3 % to 13.4 %, while E varies from 31.3 % - 23.9 % for NUAR to 33.6%- 26.6% for UAR.

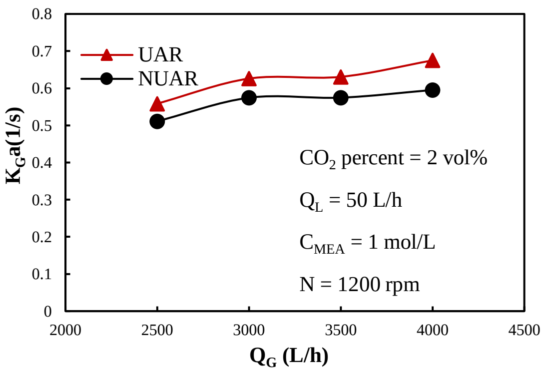


Figure 6. Effect of gas flow rate on K_Ga.

3.5. Effect of CO_2 Concentration on K_Ga

Figure 7 illustrates the effect of CO_2 concentration on K_Ga, while Q_G, Q_L, C_{MEA}, N, and P_{us} were set at 3500 L/h, 50 L/h, 1 mol/L, 1200 rpm, and 22.5 W, respectively. The effect of increasing this parameter on CO_2 absorption is the opposite of increasing MEA concentration. It means that for a specific amount of MEA, there are more CO_2 molecules that should be captured. In this case (high CO_2 concentration), thermodynamics (solubility) would govern the removal efficiency instead of kinetics which finally caused a decrease in removal efficiency [46]. Furthermore, ultrasonic effects on process efficiency are similar to other parameters. In these experiments, for CO_2 concentration of 10000 - 40000 ppm (1%-4%), the values of σ_{K_Ga} change from 7.4 % to 16.3 %, while E varies from 36.4 % to 22.0 % for NUAR to 38.5%- 25.0 % for UAR.

According to the above evidence, despite the very low residence time of the liquid droplets in the RPB reactor, 1.7 MHz ultrasound can significantly enhance CO_2 absorption and K_Ga. Using the above results, $\overline{\sigma_{K_Ga}}$ (the average arithmetic value of σ_{K_Ga}) was obtained 11.4%, respectively.

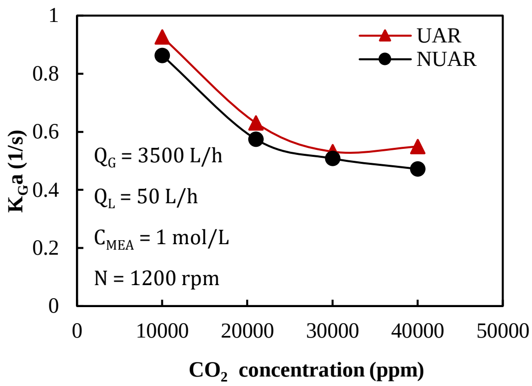


Figure 7. Effect of CO_2 concentration on K_Ga.

3.6. Comparison of the RPB Performance in This Study with Other Works

The comparison of the performance of the present reactor with other existing reactors with almost similar structures and conditions is done in Table 1. Although the comparison is very difficult due to the different structures of the reactors and their internal components and different operating conditions. As it was mentioned, for the first time, Luo et al [2] studied the effect of ultrasound on mass transfer coefficient using a NaOH-CO_2 system in an RPB reactor. As they reported K_La, it was not possible to compare our study with them. From Table 1, it can be seen that in almost similar conditions, relatively more K_Ga was obtained in the present work. For example, the maximum K_Ga of the present study is 20% more than the K_GA in the work of Lin et al. [48], which is a significant increase considering as the percentage of carbon dioxide in the present study is twice. In addition, in terms of comparison with the work [42], in the same conditions, the reactor equipped with ultrasonic waves has achieved higher coefficients. Also, in the almost similar conditions, the maximum K_Ga of the present study is 4.17 times the K_Ga reported in the work of Mohammadi and Heidari [45].

Table 1. Comparison of the RPB performance in this study with other works

Type of packing	RPB rotational speed (rpm)	Liquid flow rate (L/h)	Gas flow rate (L/h)	Absorbent concentration (mol/L)	CO_2 (%Vol)	K_Ga (1/s)		Efficiency (%)		Ref.
Structured Packing	[375- 1735]	2.52	[264- 786]	NaOH [0.2- 1]	1	[0.1- 0.75]		—		[47]
Blade Packing	[600- 1800]	[12- 30]	[540- 3960]	MEA [1]	1	[0.12- 0.8]		—		[48]
Blade Packing	[400- 1600]	[20- 120]	[2500- 4000]	MEA [1- 4]	2	[0.3- 0.9]		[14.8- 37.7]		[42]
Arc Blade	[400- 1600]	[18- 36]	3360	MEA [3- 12]	2	[0.13- 0.23]		[49- 78]		[45]
Blade packing	[400- 1600]	[20- 120]	[2500- 4000]	MEA [1- 4]	2	Without US [0.3- 0.9]	With US [0.4- 0.96]	Without US [14.8- 37.7]	With US [19.1- 39.6]	This work

4. Conclusions

In this study, the investigation of the effect of high frequency ultrasonic waves on CO_2 absorption in the RPB reactor with twelve blade packings was done. Four 1.7 MHz ultrasonic transducers were embedded in the bottom of the reactor. All experiments were repeated in the absence and presence of ultrasound. The CO_2 absorption percent (E), and the volumetric gas-side mass transfer coefficient (k_Ga) were investigated by using an MEA-CO_2 mass transfer system. The effects of different parameters such as rotational speed (400–1600 rpm), liquid flow rate (20 –120 L/h), MEA concentration (1- 4 mol/L), gas flow rate (2500- 4000 L/h), and CO_2 concentration (1- 4 vol%) have been investigated. According to the evidence, despite the very low residence time of the liquid droplets in the RPB reactor, 1.7 MHz ultrasound can significantly enhance CO_2 absorption. The following results have been obtained:

- The removal efficiency increased with increasing gas and liquid flow rate, and rotational speed, as well as MEA concentration.
- With increasing CO_2 concentration, absorption efficiency decreased.
- $\overline{\sigma_{K_Ga}}$ (The average arithmetic value of σ_{K_Ga}) was obtained 11.4% under the ultrasound field.
- The average CO_2 removal efficiency was enhanced from 27.4 % in the absence of ultrasound to 29.8% in the presence of ultrasound.
- High frequency ultrasound can enhance CO_2 absorption, even in high efficiency equipment like RPBs.

Nomenclature

C_i, C_o	CO_2 mole fraction in the inlet and outlet gas [mol/mol]
C_{MEA}	MEA concentration [mol/L]
E	CO_2 absorption efficiency, %
K_Ga	Overall volumetric gas-side mass transfer coefficient [1/s]
N	Rotational speed [rpm]
P	power [W]
Q	Flow rate [L/h]
R_i	The inner radius of the RPB [m]
R_o	The outer radius of the RPB [m]
Z_b	The axial height of the packings [m]
Greek Symbols	
σ_{K_Ga}	Relative volumetric gas-side mass transfer coefficient (dimensionless)
Subscripts	
0	Without ultrasound
G	Gas stream
i	inlet
L	Liquid stream
o	Outlet
us	With ultrasound
Abbreviations	
NUAR	Non-ultrasonic assisted RPB reactor
UAR	Ultrasonic assisted RPB reactor

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