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High performance promoter-free CO_2 absorption using potassium carbonate solution in an ultrasonic irradiation system

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ABSTRACT

CO_2 capture for power plant and natural gas purification using absorption process suffers two major drawbacks: large absorption body and high regeneration energy. The high energy penalty of absorption technology can be addressed using low heat of absorption solvent. However, the challenge of using low heat of absorption solvent is to develop a practical approach to intensify absorption rate. In recent years, high frequency ultrasonic system emerges as a potential technology for mass transfer process. In this study, the potential of using high frequency ultrasonic system have been investigated using the slow kinetic solvent: (potassium carbonate) without utilizing any chemical promoter. The ultrasonic-assisted absorption system with 20 wt% potassium carbonate (without promoter) has provides 1.75 times higher volumetric mass transfer coefficient than the Piperazine (PZ) promoted potassium carbonate using stirring method. The absorption rate has been increased up to 32 times as compared to the case without ultrasonic irradiation. Besides, the required absorption time to achieve 0.9 loading (CO_2 mole/K_2CO_3 mole) has been significantly reduced to approximate 400 s. Furthermore, an ultrasonic-assisted absorption model has been developed by including the atomization, ultrasonic streaming, and ultrasonic chemical effect in order to investigate the mechanism involved in high frequency ultrasonic-assisted absorption. Based on the model validation study, the ultrasonic chemical effect is essential to be considered in order to match the simulated and the experimental results. The results of current study prove that, high frequency ultrasonic system possesses high potential to be utilized to enhance the absorption using promoter-free potassium carbonate.

1. Introduction

CO_2 removal technology plays an important role in fossil fuel power plant and natural gas purification. There are three major approaches for CO_2 capture for power plant, including: pre-combustion, post-combustion, and oxyfuel process [1]. The pre-combustion and post-combustion are required to be integrated with the CO_2 capture process. For post-combustion, CO_2 removal is required for the treatment of flue gas from combustion [2]. Meanwhile, for pre-combustion treatment, integrated gasification combined cycle [3] or integrated reforming combined cycle [4] process is applied in order to generate the syngas from the fuel source. CO_2 is then captured from the product of the syngas. The idea strategic is to capture CO_2 from the power plant for CO_2 utilization in industry [5]. Furthermore, CO_2 capture is also an essential process in natural gas purification. Nowadays, most of the high quality natural gas reservoirs are depleted. The remained natural gas reservoirs content large amount of CO_2 up to 87% [6]. The CO_2 is required to be removed from the natural gas and injected back to underground for enhanced oil recovery. The critical process condition for power plant and natural gas purification remain challenging for efficient CO_2 capture process due to the high temperature and high pressure of condition.

To date, several technologies have been widely reported for CO_2 capture, including absorption [6–8], adsorption, membrane [9,10], and cryogenic. Among these technologies, chemical absorption is the most established technology with the high CO_2 absorption capacity [11,12]. For the absorption process, amine-based solvent, such as monoethanolamide (MEA), has been widely used for industrial application as it provides high chemical reaction rate in capturing CO_2 [11,13]. However, the usage of MEA causes several drawbacks, such as high energy consumption for regeneration and degradation of the solvent. In order to overcome the high regeneration energy, the low heat of absorption solvent is recommended such as ammonia and potassium carbonate. Potassium carbonate (K_2CO_3) solution is an attractive solvent for CO_2 capture due to the advantages of low toxicity, low volatility, and not prone to degradation [14,15]. The solvent is commonly used to capture CO_2 using pressure swing method at operating temperature higher than 100 °C [16]. The approach is believed to be more energy efficiency, particularly at the high pressure and high

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Nomenclature

u	Axis velocity [m s^{-1}]
c_0	Sound velocity [m s^{-1}]
I	Ultrasonic power intensity [W m^{-2}]
P_{us}	Ultrasonic power [W]
P_0	Acoustic pressure [Pa]
f_T	Effective fountain flow rate [m^3 s^{-1}]
d	Droplet diameter [m]
L_{CO_2}	CO_2 loading density [kmol m^{-3}]
k_{OH}	Second order rate constant [m^3 kmol^{-1} s^{-1}]
k_l^o	Liquid mass transfer coefficient [m s^{-1}]
K_g	Overall mass transfer coefficient [mol h^{-1} kPa^{-1}]
$\dot{n}_{co2}$	CO_2 absorption rate [kmol s^{-1}]
a_e	Effective surface area with ultrasonic irradiation [m^2]
$E_{K_2CO_3}$	Chemical enhancement factor [−]
Ha	Hatt number [−]
$C^*_{CO_2}$	Concentration of CO_2 in the liquid interfacial [kmol m^{-3}]
C_{CO_2}	Concentration of CO_2 in the liquid bulk [kmol m^{-3}]
H_{CO_2}	Henry's constant of CO_2 gas in solvent [kPa m^3 $kmol^{-1}$]
b	Stoichiometric factor [−]
D_{CO_2}	Diffusion coefficient of CO_2 [m^2 s^{-1}]
V	Gas volume [m^3]
p	Pressure [Pa]
t	Time [s]
T	Temperature [K]
T_A	Contact time of droplet a
T_B	Contact time of droplet B
R	Ideal gas constant [J mol^{-1} K^{-1}]
Z	Gas compressibility [−]
y	Vapor mole fraction [−]
Greek	
ρ	Solution density [kg m^{-3}]
ω	Mass fraction [−]
μ	Viscosity [k gm^{-1} s^{-1}]
η	Shear viscosity [Pa s]
η'	Bulk viscosity [Pa s]

temperature feed condition, such as natural gas purification [13] and pre-combustion process [4,17,18].

Nonetheless, the heat of absorption of K_2CO_3 solution is much lower if compared with MEA solution [15,19,20]. Thus, the efficiency penalty for CO_2 capture using K_2CO_3 during the pre-combustion is less than 13.1% [4,17,21]. Meanwhile, the efficiency penalty for CO_2 capture using conventional MEA absorption is in the range of 20 to 30% [17,22]. The lower regeneration energy of the solvent is however always associated with slow absorption rate. The absorption rate of promoter-free K_2CO_3 is relatively slow [23]. Therefore, the major issue of using promoter-free K_2CO_3 for CO_2 absorption is the large size of absorption column required [14].

Therefore, addition of promoter is one of the approaches to enhance the absorption rate of K_2CO_3, which included salts [24], amines [25–27], acids solution [28], polymers [29], and enzyme [30]. Tetramethylammonium glycinate promoted K_2CO_3 solution was also demonstrated in recent years [23]. Table 1 shows the list of promoters used for K_2CO_3. However, most of the promoters cause several drawbacks towards the CO_2 capture performance, such as the increasing of heat of absorption [31], diffusion limiting under high pressure operating condition due to small concentration of promoter [31,32], the degradation problem, high volatility problem under high temperature operating condition, and etc. [14].

It is noteworthy that the conventional absorption system using fast kinetic solvent, such as PZ and MEA, has also suffered the oversized absorption column for CO_2 capture. For example, in a large scale natural power plant using conventional MEA absorption process [35] at 600 MW, a four-train process system is required to treat flue gas at flow rate of 600 m^3/h. Each of absorption column units has the diameter and height of 4.7 and 44 m, respectively. Therefore, the overall size of the absorption system is enormous, which requires high capital and maintenance cost. In addition, capital and maintenance cost required for the CO_2 absorption (per CO_2 mole) are approximated to be half from the total operating cost [36]. Therefore, the size of the absorption or desorption column is required to be minimized in order to reduce the capital cost.

It is reported in the literature that the size of the absorption column can be reduced using rotated packed bed (RPB) [37–40] and hollow fiber membrane contactor (HFMC) [41,42]. The advantage of the RPB is their high volumetric mass transfer, which is up to 50 times higher than conventional packed bed column [38–40]. However, the RPB also requires high power consumption in order to accelerate the CO_2 absorption process [38,43]. Thiels reported the total power required for standard-alone RPB is more than 0.1 kWhr per kg CO_2 using rotation speed of 700 rpm [44]. In relation to it, Thiels suggested a strategic of coupling RPB with conventional packed bed in order to reduce the size and power consumption of RPB. In general, RBP suffers the high power consumption and contains moving part. Meanwhile, the advantage of HFMC is their large surface area for CO_2 absorption. However, this technology also suffers several drawbacks, such as the constraint of operating condition (pressure and temperature), and requirement of pre-treatment process. Therefore, it is worth to explore a new alternative for CO_2 absorption system. In this paper, high frequency ultrasonic system is proposed for CO_2 absorption system using promoter-free K_2CO_3 solvent.

Fig. 1 shows the enhancement effect using high frequency ultrasonic irradiation in the formation of ultrasonic streaming force and ultrasonic fountain. There are several reasons for using high frequency ultrasonic irradiation in the enhancement of absorption process. Firstly, the streaming turbulence effect can be magnified using higher frequency of ultrasonic irradiation [45]. This is because, under ultrasonic irradiation, the streaming turbulence can be induced to create a convective dynamic flow of liquid phase [45–47]. Secondly, ultrasonic fountain effect is created under high frequency of ultrasonic irradiation [48]. At higher frequency, the size of the droplets that pinched out from the liquid surface through vibration is smaller, and thus, the surface area

Table 1
List of promoters used for K_2CO_3.

Promoter		CO_2 pressure, kPa	K_2CO_3 wt%	T, K	Acceleration	Absorption rate $\times 10^7$, mol/cm^2 s	Refs.
Piperazine (PZ)	0.6 M	3.5	3.6 M	333.15	10	1.68	[31]
Ethylaminoethanol (EAE)	2.0 M	4	30 wt%	303	12	6.0	[33]
Arginine	5 wt%	13	35 wt%	342.3	> 2	4.0	[34]
Boric acids	1.5 M	0.1	30 wt%	80 °C	1.29	–	[28]
Boric acids	3 wt%	–	30 wt%	50–75 °C	1	–	[18]