Research Trends of Carbon Dioxide Capture using Ionic Liquids and Aqueous Amine-Ionic Liquids Mixtures

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ABSTRACT

Anthropogenic CO_2 emissions has led to global climate change and widely contributed to global warming since its concentration has been increasing over time. It has attracted vast attention worldwide. Currently, the different CO_2 capture technologies available include absorption, solid adsorption and membrane separation. Chemical absorption technology is regarded as the most mature technology and is commercially used in the industry. However, the key challenge is to find the most efficient solvent in capturing CO_2. This paper reviews several types of CO_2 capture technologies and the various factors influencing the CO_2 absorption process, resulting in the development of a novel solvent for CO_2 capture.

Keywords: *chemical absorption, amine, ionic liquids, CO_2 solubility*

INTRODUCTION

Generally, major sources of carbon dioxide (CO_2) emission are from industries such as natural gas treatments, fossil fuel power plant and petroleum industries. Flue gas from coal combustion contains hazardous pollutants such as CO_2, mercury (Hg) and sulfur dioxide (SO_2), at different compositions and percentages. Carbon dioxide has been considered a major contributor to greenhouse gases since its concentration in the atmosphere has been increasing over time. Excessive CO_2 emission has a negative impact that leads to global climate change and disruption of the ecosystem. In order to maintain a safe and secure environment, CO_2 emissions can alternatively be controlled by carbon capture and storage (CCS) [1], whereby CO_2 is separated from flue gas and permanently stored in large subsurface geologic reservoirs.

Various technologies that have been developed for CO_2 capture are gas membrane separation, solid adsorption, and absorption by chemical/physical solvents[1][2][3]. Table 1 shows a comparison of three different CO_2 capture methods. These methods utilize different approaches which can be adapted to the treatment of industrial effluents. In the chemical industry, the use of separation technologies correlate directly with operational confidence [4]. The absorption technology is the most common method for capturing CO_2, since it is applicable to industry demands and scale. Efficiency of the carbon dioxide absorption technology can be as high as 98%. The source of energy penalty in absorption-based methods is from thermal regeneration, h leads to high-energy consumption. This may be because a large amount of water is vaporized during solvent regeneration. Compared to other CO_2 capture methods, adsorption requires a slightly lower amount of energy for the regeneration process. This is because there is low steam loss during CO_2 desorption [5]. Energy penalty in the adsorption process comes from either thermal or vacuum regenerations. In the membrane separation methods, energy penalty is from feed compression or/and vacuum on the permeate. Membrane separation technology is seldom used as the operating flexibility of the membrane system is strongly affected by flue gas conditions, making it difficult to apply the technology [6].

Absorption processes are commonly employed in chemical industries, where they are significantly utilized for CCS applications, and in the treatment of natural gases. One of the most effective methods for CO_2 capturing is by the use of amine solvents in gas absorption. However, the use of alkanolamines for CO_2 capture has several drawbacks, such as high incidence of corrosion, volatility and proneness to thermal and oxidative degradation [7][8]. These limitations cause extra energy consumption and require more investment on equipment [9]. Therefore, the detriment of using amine solvents for CO_2 capture drives researchers to find alternative solvents with improved absorption performance.

Table I: Comparison of Post-Combustion CO_2 Capture Methods

	Absorption	Adsorption	Membrane	Ref
Commercial usage in CPI[a]	High	Moderate	Low	[4]
Operational Confidence	High	High but complex	Low to moderate	[4]
Operating Flexibility	Moderate	Moderate	High (CO_2 > 20%) Low (CO_2 < 20%)	[10]
Energy Requirement	4-6 MJ/ kg CO_2	2-3 MJ/kg CO_2	0.5-6 MJ/ kg CO_2	[10]
CO_2 Recovery	90-98%	80-95%	80-90%	[6] [10]
Scale	Industrial	Pilot	Experimental	[11]
Primary source of energy penalty	Solvent Regeneration (Thermal)	Sorbent Regeneration (Thermal/ Vacuum)	Compression on feed and/ or vacuum on permeate	[4]
Development trends	New chemistry, thermal integration	New chemistry, process configuration	New membrane, process configuration	[4]

[a] Chemical Process Industry

The use of amines and ionic liquid mixtures as improved solvents has been suggested to overcome these drawbacks[12]. Ionic liquids (ILs) are the organic salts that form stable liquids below 100°C, or even at room temperature. The recent concept of using ILs for CO_2 capture has became attractive when it was discovered that they had CO_2 absorption properties [3][13][14]. The advantages of ILs are that they have low vapor pressure and are thermally stable over a wide range of temperature. However, most ILs are not cost effective compared to commercial amines. Thus, developing economical and energy efficient CO_2 capture technologies is urgently required.

With respect to this, the desirable properties of ILs and alkanolamines may be integrated, so that energy can be saved during the regeneration process. The aim of this short review is to summarize research trends in CO_2 capture, and the factors influencing the CO_2 capture when using ionic liquids and aqueous amine-ionic liquids mixtures.

RESEARCH PROGRESS ON CO_2 CAPTURE

Ionic Liquid

In the recent decade, ionic liquids have been considered as good absorbents for CO_2 capture. Zubeir *et al.,*[15] studied the solubility of CO_2 in low viscous ILs; 1-butyl-3-methylimidazolium tricyanomethanide [bmim][tcm] for the first time, using it as a solvent for capturing CO_2. They performed the experiment at 288.15K to 363.15K with pressure of 0.01 to 10 MPa using two different methods (gravimetric and volumetric). The authors discovered that [bmim][tcm] had good solubility performance compared to other non-fluorinated ionic liquids. This indicated that [bmim][tcm] was a suitable and promising solvent candidate for carbon capture.

In a subsequent research, three ionic liquids; Methyl Trioctyl Ammonium Bis (trifluoromethylsulfonyl)imide [MOA][Tf_2N], 1-butyl-3-Methylimidzolium Bis (trifluoromethylsulfonyl)imide [bmim][Tf_2N] and 1-butyl-3-Methylimidazolium Methyl Sulfate [bmim][$MeSO_4$] were used by Bahadur et al., [16] to capture CO_2. They used gravimetric analysis to measure CO_2 solubility and found that absorption increased with pressure and decreased with temperature. [MOA][Tf_2N] was noted to have the highest CO_2 solubility, followed by [bmim][Tf_2N] and [bmim][$MeSO_4$].

Most CO_2 absorption performance studies were carried out using imidazolium ionic liquids. Pinto et al., [17] investigated CO_2 solubility performance using pyridinium-based ionic liquid, 1-ethylpyridinium ethylsulfate [C2Py][$EtSO_4$]. The temperature was set at 298.2 K and pressure up to 1.6 MPa. They compared the CO_2 loading performance between [C2Py][$EtSO_4$] and an equivalent imidazolium-based ionic liquids, 1-ethylimidazoliumethylsulfate [C2mim][$EtSO_4$] [18]. The authors found that pyridinium ionic liquids exhibited slightly lower CO_2 absorption. This may due to greater steric effects on [C2mim][$EtSO_4$] that led to the generation of more free volume. Pyridinium-based ionic liquids were considered more biodegradable and cheaper compared to imidazolium ionic liquids, and much more promising when used in real scaled-up CO_2 absorption.

The CO_2 solubility performance using six hydroxyl ammonium ionic liquids; 2-hydroxyethanaminium acetate [hea], bis (2-hydroxyethyl) ammonium acetate [bheaa], 2-hydroxy-N-(2-hydroxyethyl)-N-methylethanaminium acetate [hhemea], 2-hydroxyethanaminium lactate [hel], bis (2-hydroxyethyl) ammonium lactate [bheal] and 2-hydroxy-N-(2-hydroxyethyl)-N-methylethanaminium lactate [hhemel] were studied by Kurniaet al., [19]. They observed the CO_2 solubility results in the sequences of [hea] > [bheaa] ≈ [hel] > [bheal] > [hhemel] >[hhemea]. Enthalphy and entrophy from estimated Henry's constant showed that solubility increased with pressure and decreased with temperature.

AQUEOUS AMINE-IONIC LIQUID MIXTURES

Recently, Lv *et al.*, [20] forwarded the idea of mixing Monoethanolamine (MEA) and hydrophilic amino acid ionic liquid [C_2OHmim][Gly] in order to study CO_2 capture performance. They found that the CO_2 absorption capacity of the mixed solution was higher compared to the total absorption of MEA single solution. The MEA/ [C_2OHmim] system tend reacts with CO_2 as zwitterions They act as a base to participate in the carbamate formation.

This is in agreement with the findings of Taib and Murugesan [21] which compared the CO_2 absorption performance of ionic liquids [bmim][BF_4] and [bheaa], to MEA solution. Both amine ionic liquid mixtures were found to have higher CO_2 loading compared with single amines or ionic liquid solutions. In subsequent research, Feng *et al.*, [22] found that the addition of ionic liquids ([N_{1111}][Gly]), ([N_{2222}][Gly]), ([N_{1111}][Lys]) and ([N_{2222}][Lys]) to amine mixtures greatly reinforced CO_2 absorption. .

In contrast, Fu and Zhang [23] found that CO_2 loading decreased significantly when 1-butyl-3-methylimidazolium glycinate [bmim][Gly] was added to the methyldiethanolamine MDEA aqueous solution. However, the absorption rate increased with the addition of ILs into MDEA aqueous solution. Results from the study of Xu *et al.*, [24] supported the findings, where the addition of ionic liquids in amine solution was shown to decrease CO_2 absorption efficiency. They conducted the experiment using two low viscous ionic liquids; [C_2OHmim][DCA] and [bmim][DCA], mixed with an aqueous MEA solution. The authors reported that both [C_2OHmim]

[DCA] and [bmim][DCA] gave slightly reduced solubility, due to salting out effects that inhibited CO_2 absorption.

Ahmady *et al.*, [25] also found that the amount of CO_2 loading declined with the addition of [bmim][DCA], [bmim][BF4] and [bmim][Ac], into MDEA solution. Similarly, results from Sairi *et al.*, [26] demonstrated that the use of MDEA and $[gua]^+[OTf]^-$ mixtures resulted in less CO_2 absorption, as $[gua]^+[OTf]^-$ hindered the tertiary amine of MDEA.

Table 2 shows summarizes recent research findings in CO_2 capture using amine and ionic liquid mixtures.

Table 2: Summary of the Effects of Various Aqueous Amine-Ionic Liquid Mixtures on CO_2 Loading

Amine	Ionic Liquids	CO_2 Loading	Ref
MEA	• hydrophilic amino acid ionic liquid $[C_2OHmim][Gly]$	Increase	[20]
MEA	• bis(2 hydroxyethyl)ammonium acetate (bheea) • 1-butyl-3-methylimidazolium tetrafluoroborate $[bmim][BF_4]$	Increase	[21]
MDEA	• tetramethylammonium glycinate $[N_{1111}][Gly]$, • tetraethylammonium glycinate $[N_{2222}][Gly]$ • tetramethylammonium lysinate $[N_{1111}][Lys]$ • tetraethylammonium lysinate $[N_{2222}][Lys]$	Enhance	[22]
MDEA	• 1-butyl-3-methylimidazolium glycinate [bmim][Gly]	Decrease	[23]
MEA	• 1-(2-hydroxyethyl)-3-methyl-imidazolium dicyanamide $[C_2OHmim][DCA]$ • 1-butyl-3-methylimidazolium [Bmim][DCA]	Decrease	[24]
MDEA	• 1-butyl-3-methyl-imidazolium tetrafluoroborate [bmim][BF4] • 1-butyl-3-methyl-imidazolium acetate [bmim][Ac] • 1-butyl-3-methyl-imidazolium dicyanamide [bmim][DCA]	Decrease	[25]

Amine	Ionic Liquids	CO_2 Loading	Ref
MDEA	• Guanidium trifluoromethanesulfonate $[gua]^+[OTf]^-$	Decrease	[26]
MDEA	• Guanidium tris(pentafluoroethyl) trifluorophosphate $[gua]^+[FAP]^-$	Decrease	[27]the solubility of CO 2 in aqueous blends of N-methyldiethanolamine (MDEA
AMP	• N-butyl-3-methylpyridinium tetrafluoroborate, $[B_3MPYR][BF_4]$	Decrease	[28]

FACTORS AFFECTING ABSORPTION PERFORMANCE

In recent years, several experimental works have been carried out to investigate the efficiency of CO_2 capture. Details on the effect of temperature, pressure, absorbent/solvent concentration, anion types, and alkyl chain length in cations, on the performance of CO_2 capture is further discussed in following subsections.

Effects of Temperature and Pressure

Gas absorption theories by chemical reaction have assumed in isothermal conditions to facilitate the experimental procedure. Generally, liquid phase temperature can increase due to the heat given off by the solution, or even by the reaction itself. In CO_2 removal with alkanolamine solution, high thermal effects have been recorded.

The study of CO_2 solubility using aqueous mixtures of MEA + Ionic liquids; bis (2-hydroxylethyl) ammonium acetate [bheaa] and 1-butyl-3-methylimidazolium tetrafluoroborate [bmim] $[BF_4]$ was conducted by Taib and Murugesan [21] .They discovered that the solubility of CO_2 increased linearly with pressure, for both aqueous bheaa and in addition of MEA solution. A similar trend was also reported when they used aqueous [bmim] $[BF_4]$. The addition of ionic liquid was expected predominated the process by physical solubility of CO_2. In terms of temperature effects, they found that there was not much change in CO_2 absorption when temperature was increased in the range of 298.15K-313.15K.

This CO_2 solubility pattern is in agreement with findings of other researchers. Feng *et al.*, [22] used four amino acid based ILs: tetramethylammonium glysinate $[N_{1111}]$[Gly], tetraethylammonium glysinate $[N_{2222}]$[Gly], tetramethylammonium lysinate $[N_{1111}]$[Lys], and tetraethylammonium lysinate $[N_{2222}]$[Lys] to investigate the effect of temperature ranging 298K to 318K towards the absorption of CO_2 in ILs + MDEA aqueous solutions. They found that raising the temperature increased the absorption rate only during the first 20 minutes. This is due to the larger reaction rate and might be attributed to the solution becoming less viscous at higher temperatures, which led to higher diffusibility. However, a further increase of temperature resulted in less CO_2 amount being absorbed.

This similar trend was also seen by Xu *et al.*, [24] in their work where CO_2 solubility decreased with an increase in temperature. The authors used two low viscous ionic liquids; [C_2OHmim][DCA] and [Bmim][DCA] mixed with aqueous 30 wt% MEA. In terms of pressure effects toward CO_2 absorption, results showed that CO_2 solubility was directly proportional to the increase in CO_2 partial pressure.

The study of the effects of temperature and pressure on CO_2 absorption in aqueous N-methyldiethanolamine (MDEA) and guanidinium trifluoromethanesulfonate, $[gua]^+$ $[OTf]^-$ ionic liquid system at elevated pressures and various temperatures were conducted by Sairi *et al.*, [26]. Their finding is in agreement with the expected general trend. Equilibrium loading was found to decrease with temperature and increase with pressure. This indicated that more gas was present in the solution with a lower temperature, compared to a solution of higher temperature. This trend can be explained by the fact that vapor pressure increases with temperature. Based on Henry's law, the solubility of a gas in a liquid is proportional to the partial pressure of the gas above the surface of the liquid [29]. The same phenomenon was also reported by Anthony *et al.*, [30] and Husson-Borg *et al.*, [31].

In 2012, Aziz *et al.*, [27] demonstrated the effects of temperature and pressure on absorptionby conducting an experiment,usingaqueous mixtures of MDEA and [gua]+[FAP]−. They concluded that the solubility of carbon dioxide was inversely proportional to an increase in temperature and pressure.

Effects of Anions and Cations of Ionic Liquids

Anions and cations also provide significant impact on absorption systems and CO_2 solubility.

With respect to the influence of anion and cation, the effect of length of the alkyl chain was evaluated by Gonzalez-Miquel *et al.,*[32]. They compared three different imidazolium based ionic liquids; 1-hexyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [hxmim][Tf_2N], 1-octyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [omim][Tf_2N] and 1-decyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [dcmim][Tf_2N] in CO_2 capture. They found that CO_2 absorption in ILs slightly increased with increasing length of cation alkyl chain. This solubility pattern was supported by evaluating the Henry's law constant (K_H) of $CO_{2,}$ where K_H values decreased with increasing length of the alkyl chain, indicating higher CO_2 solubility.

Yunus *et al.*, [33] investigated the effect of alkyl chain length in four of pyridinium based ionic liquids toward CO_2 capture where the CO_2 solubility in [C4py][Tf2N], [C8py] [Tf2N], [C10py][Tf2N], and [C12py][Tf2N] were measured at 3 different temperatures of 298.15 K, 313.15 K and 333.15 K. They found that [C12py] [Tf2N] had the greatest total of CO_2 absorbed compared to others, since the absorption performance depends on the length of chain. Aki *et al.*, [34] reported that increasing the cation alkyl chain length of the ionic liquid will decrease the density of the ionic liquid and subsequently increase free volume. Space filling allows for greater absorption of CO_2. Thus, it was concluded that an increase in the cation alkyl chain length, slightly increases the CO_2 solubility in the ionic liquid [35].

Most studies have proved that the solubility of CO_2 was far more influenced by the nature of anions, compared to cations of the ILs [36][37][38]. Bahadur *et al.*, studied the effect of anions and cations of ionic liquids on CO_2 solubility. They used Methyl Trioctyl Ammonium Bis (trifluoromethylsulfonyl) imide [MOA][Tf_2N], 1-butyl-3-Methylimidzolium Bis (trifluoromethylsulfonyl) imide [bmim][Tf_2N] and 1-butyl-3-Methylimidazolium Methyl Sulfate [bmim][$MeSO_4$] as the solvent to absorb CO_2. The authors reported that CO_2 solubility is in the sequence of [MOA]

[Tf_2N > [bmim] [Tf_2N] > [bmim] [$MeSO_4$]. The increasing fluorination of the ionic liquids resulted in higher absorption. Solubility was influenced by the strong interaction between anion gas and the molar volume of the ionic liquid. When the molar volume of the ILs increases, the void space eventually increases, allowing for more gas to be dissolved.

Similar solubility trends was also reported by Ramdin *et al.,* [39] using inexpensive solvents; tributylmethylammonium methylsulfate [TBMN] [$MeSO_4$] and tributylmethylphosphonium methylsulfate [TBMP][$MeSO_4$] for capturing $CO_{2.}$ The authors compared the solvents with with commonly used industrial ILs. Their findings revealed that both [TBMN][$MeSO_4$] and [TBMP][$MeSO_4$], had slightly higher solubility compared to 1-butyl-3-methylimidazolium methylsulfate [bmim][$MeSO_4$] [40]. However, by comparing with 1-butyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [bmim][Tf_2N] [41], obviously fluorinated anion has a much larger effect on solubility. This explanation was strongly supported by the sequences of CO_2 solubility reported by Sharma *et al*., [42] where; BF^-_4 < DCA^- < PF^-_6 < TfO^-< Tf_2N. The anion effect on CO_2 solubility was attributed to the presence of the fluoroalkyl group, which led to highest CO_2 solubility [43][44][45][46], and due to the weaker ILs cation-anion interactions. Table 3 lists anion classification and CO_2 solubility.

Table 3: Influence of Anions in Different Ionic Liquids

Anion	Nomenclature	Classification	CO_2 solubility
Tetrafluoroborate	BF_4	Fluorinated	High
Hexafluorophosphate	PF_6		
Trifluoromethanesulfonate	TfO		
Bis (trifluoromethylsulfonyl) imide	Tf_2N		
Tris (trifluoromethylsulfonyl) methide	methide		
Dicyanamide	DCA	Non fluorinated	Low
Nitrate	NO_3		

Effects of Ionic Liquids / Amines Concentrations

Recently, Lv *et al.*, [7] compared the CO_2 capture into different mole ratios of MEA / [C_2OHmim] [Gly] mixtures.They found that the absorption capacity of aqueous MEA solution was lower than pure ionic liquid. The absorption capacity increased with increasing [C_2OHmim] [Gly] concentration for mixed solutions of MEA and [C_2OHmim] [Gly].

Nordin *et al.,* [28] evaluated the interaction of 2- amino-2-methyl-1 propanol (AMP) with N-butyl-3-methylpyridinuim tetrafluoroborate, [B_3MPYR][BF_4]. The amount of CO_2 absorbed was found to decrease with increasing of [B_3MPYR][BF_4] concentrations. The acidity of the ionic liquid reduced the alkalinity of aqueous AMP in the mixtures, causing the driving force for the mass transfer to decrease and inhibit CO_2 absorption [27]. Blends of methyldietanolamine (MDEA) with three different types of ionic liquids, 1-butyl-3-methyl-imidazolium tetrafluoroborate [bmim][BF_4], 1-butyl-3-methyl-imidazolium acetate [bmim][Ac] and 1-butyl-3-methyl-imidazolium dicyanamide [bmim][DCA] have been extensively studied by Ahmady *et al.*, [25] who observed similar trends.

CONCLUSION

Generally, the addition of ionic liquids into aqueous amine solution enhanced CO_2 absorption and CO_2 loading performance. However, in some cases there is still contrasting trends found by researchers. Absorption was seen to decrease with increasing temperatures, and linearly increased with increasing pressure. Carbon dioxide solubility was found to decrease with increased concentrations of amine or ionic liquids, due to the increase in viscosity and decresed presence of water molecules in the solution. Anions and cations significantly affects CO_2 absorption. Solubility is more affected by the nature of anions, compared to cations. Ionic liquids with fluoroalkyl groups absorb a high amount of CO_2 compared to non-fluorinated ionic liquids. Cation alkyl chains impart a slight effect on CO_2 solubility. Increasing the chain length will slightly increase CO_2 solubility. Selecting the right combination of amine-ionic liquid mixtures can improve the performance of CO_2 absorption.

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