Substituent Effect on Corrosion Inhibition of Schiff Bases Derived from Isatin
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The Effect of Oxalic Acid as a Doping Agent on the Conductivity of Polyaniline
Amirah Amalina Ahmad Tarmizi, Mohamad Kamal Harun, Saifollah Abdullah Hadariah Bahron, Muhd Zu Azhan Yahya, Sabrina M. Yahaya & Nurul Huda Abdul Halim

Climate Change and Freshwater Availability: Present and Future Challenges
Izzadin Ali, Dasimah Omar & Siti Mazwin Kamaruddin

Polyhydroxybutyrate for Improved Oil Recovery: A Literature Review
Norruhluda Mohd Taib, Norfarisha Achim & Zulkafli Hassan

Research Trends of Carbon Dioxide Capture using Ionic Liquids and Aqueous Amine-Ionic Liquids Mixtures
Siti Nabihah Jamaludin & Ruzitah Mohd Salleh

Dye-Sensitized Solar Cells Using Natural Dyes Extracted From Plumeria and Celosia Cristata Flowers
Siti Noraini Abu Bakar, Huda Abdullah, Kamisah Mohamad Mahbub & Shahida Hanum Kamarullah

Revised Normal Ratio Methods for Imputation of Missing Rainfall Data
Siti Nur Zahrah Amin Burhanuddin, Sayang Mohd Deni & Norazan Mohamed Ramlil

The Influence of Cr Doped TiO2 on the Optical Property and Photocatalytic Activity under Sunlight Irradiation
Siti Zulaikah Suhaili, Muhamad Kamil Yaakob, Siti Irma Yuana Saaid & Umi Sarah Jais

Developing Multi-Tier Network Design for Effective Energy Consumption of Cluster Head Selection in WSN
Wan Isni Sofiah Wan Din, Saadiah Yahya, Mohd Nasir Taib, Ahmad Insan Mohd Yassin & Razulaimi Razali

Effect of Culture Technique of Ganoderma australe Mycelia on Percentage Removal of Leachate Organics
1. Substituent Effect on Corrosion Inhibition of Schiff Bases Derived from Isatin
   Aliyin Abdul Ghani
   Hadariah Bahron
   Mohamad Kamal Hj Harun
   Karimah Kassim

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   Hadariah Bahron
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   Sabrina M. Yahaya
   Nurul Huda Abdul Halim

3. Climate Change and Freshwater Availability: Present and Future Challenges
   Izzadin Ali
   Dasimah Omar
   Siti Mazwin Kamaruddin

4. Polyhydroxybutyrate for Improved Oil Recovery: A Literature Review
   Norruilhuda Mohd Taib
   Norfarisha Achim
   Zulkafli Hassan
5. Research Trends of Carbon Dioxide Capture using Ionic Liquids and Aqueous Amine-Ionic Liquids Mixtures
   Siti Nabihah Jamaludin
   Ruzitah Mohd Salleh

6. Dye-Sensitized Solar Cells using Natural Dyes Extracted From Plumeria and Celosia Cristata Flowers
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   Huda Abdullah
   Kamisah Mohamad Mahbor
   Shahida Hanum Kamarullah

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   Ahmad Ihsan Mohd Yassin
   Razulaimi Razali

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    Wan Razarinah Wan Abdul Razak
    Noor Zalina Mahmood
    Noorlidah Abdullah
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ABSTRACT

Anthropogenic CO\textsubscript{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\textsubscript{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\textsubscript{2}. This paper reviews several types of CO\textsubscript{2} capture technologies and the various factors influencing the CO\textsubscript{2} absorption process, resulting in the development of a novel solvent for CO\textsubscript{2} capture.

Keywords: chemical absorption, amine, ionic liquids, CO\textsubscript{2} solubility

INTRODUCTION

Generally, major sources of carbon dioxide (CO\textsubscript{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\textsubscript{2}, mercury (Hg) and sulfur dioxide (SO\textsubscript{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 CO₂ 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, CO₂ emissions can alternatively be controlled by carbon capture and storage (CCS) [1], whereby CO₂ is separated from flue gas and permanently stored in large subsurface geologic reservoirs.

Various technologies that have been developed for CO₂ capture are gas membrane separation, solid adsorption, and absorption by chemical/physical solvents [1][2][3]. Table 1 shows a comparison of three different CO₂ 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 CO₂, 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, leading to high-energy consumption. This may be because a large amount of water is vaporized during solvent regeneration. Compared to other CO₂ capture methods, adsorption requires a slightly lower amount of energy for the regeneration process. This is because there is low steam loss during CO₂ 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/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 CO₂ capturing is by the use of amine solvents in gas absorption. However, the use of alkanolamines for CO₂ 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 CO₂ capture drives researchers to find alternative solvents with improved absorption performance.
Table I: Comparison of Post-Combustion CO₂ Capture Methods

<table>
<thead>
<tr>
<th></th>
<th>Absorption</th>
<th>Adsorption</th>
<th>Membrane</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Commercial usage in CPI</strong></td>
<td>High</td>
<td>Moderate</td>
<td>Low</td>
<td>[4]</td>
</tr>
<tr>
<td><strong>Operational Confidence</strong></td>
<td>High</td>
<td>High but complex</td>
<td>Low to moderate</td>
<td>[4]</td>
</tr>
<tr>
<td><strong>Operating Flexibility</strong></td>
<td>Moderate</td>
<td>Moderate</td>
<td>High (CO₂ &gt; 20%)</td>
<td>[10]</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Low (CO₂ &lt; 20%)</td>
<td></td>
</tr>
<tr>
<td><strong>Energy Requirement</strong></td>
<td>4-6 MJ/kg CO₂</td>
<td>2-3 MJ/kg CO₂</td>
<td>0.5-6 MJ/kg CO₂</td>
<td>[10]</td>
</tr>
<tr>
<td><strong>CO₂ Recovery</strong></td>
<td>90-98%</td>
<td>80-95%</td>
<td>80-90%</td>
<td>[6]</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[10]</td>
</tr>
<tr>
<td><strong>Scale</strong></td>
<td>Industrial</td>
<td>Pilot</td>
<td>Experimental</td>
<td>[11]</td>
</tr>
<tr>
<td><strong>Primary source of energy</strong></td>
<td>Solvent</td>
<td>Sorbent</td>
<td>Compression on</td>
<td>[4]</td>
</tr>
<tr>
<td><strong>penalty</strong></td>
<td>Regeneration (Thermal)</td>
<td>Regeneration (Thermal/Vacuum)</td>
<td>feed and/or or vacuum on permeate</td>
<td></td>
</tr>
<tr>
<td><strong>Development trends</strong></td>
<td>New chemistry, thermal integration</td>
<td>New chemistry, process configuration</td>
<td>New membrane, process configuration</td>
<td>[4]</td>
</tr>
</tbody>
</table>

*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 CO₂ capture has became attractive when it was discovered that they had CO₂ 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 CO₂ 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 CO₂ capture, and the factors influencing the CO₂ capture when using ionic liquids and aqueous amine-ionic liquids mixtures.
RESEARCH PROGRESS ON CO₂ CAPTURE

Ionic Liquid

In the recent decade, ionic liquids have been considered as good absorbents for CO₂ capture. Zubeir et al., [15] studied the solubility of CO₂ in low viscous ILs; 1-butyl-3-methylimidazolium tricyanomethanide [bmim][tcm] for the first time, using it as a solvent for capturing CO₂. 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][Tf₂N], 1-butyl-3-Methylimidazolium Bis (trifluoromethylsulfonyl)imide [bmim][Tf₂N] and 1-butyl-3-Methylimidazolium Methyl Sulfate [bmim][MeSO₄] were used by Bahadur et al., [16] to capture CO₂. They used gravimetric analysis to measure CO₂ solubility and found that absorption increased with pressure and decreased with temperature. [MOA][Tf₂N] was noted to have the highest CO₂ solubility, followed by [bmim][Tf₂N] and [bmim][MeSO₄].

Most CO₂ absorption performance studies were carried out using imidazolium ionic liquids. Pinto et al., [17] investigated CO₂ solubility performance using pyridinium-based ionic liquid, 1-ethylpyridinium ethylsulfate [C₂Py][EtSO₄]. The temperature was set at 298.2 K and pressure up to 1.6 MPa. They compared the CO₂ loading performance between [C₂Py][EtSO₄] and an equivalent imidazolium-based ionic liquids, 1-ethylimidazoliumethylsulfate [C₂mim][EtSO₄] [18]. The authors found that pyridinium ionic liquids exhibited slightly lower CO₂ absorption. This may due to greater steric effects on [C₂mim][EtSO₄] 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 CO₂ absorption.
The \( \text{CO}_2 \) solubility performance using six hydroxyl ammonium ionic liquids; 2-hydroxyethanaminium acetate [hea], bis (2-hydroxyethyl) ammonium acetate [bheaa], 2-hydroxy-N-(2-hydroxyethyl)-N-methylethananminium acetate [hhemea], 2-hydroxyethananminium lactate [hel], bis (2-hydroxyethyl) ammonium lactate [bheal] and 2-hydroxy-N-(2-hydroxyethyl)-N-methylethananminium lactate [hhemel] were studied by Kurniaw et al., [19]. They observed the \( \text{CO}_2 \) solubility results in the sequences of [hea] > [bheaa] ≈ [hel] > [bheal] > [hhemel] >[hhemea]. Enthalphy and entropy 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\(_2\)OHmim] [Gly] in order to study \( \text{CO}_2 \) capture performance. They found that the \( \text{CO}_2 \) absorption capacity of the mixed solution was higher compared to the total absorption of MEA single solution. The MEA/ [C\(_2\)OHmim] system tend reacts with \( \text{CO}_2 \) 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 \( \text{CO}_2 \) absorption performance of ionic liquids [bmim] [BF\(_4\)] and [bheaa], to MEA solution. Both amine ionic liquid mixtures were found to have higher \( \text{CO}_2 \) loading compared with single amines or ionic liquid solutions. In subsequent research, Feng et al., [22] found that the addition of ionic liquids ([N\(_{111}\)] [Gly]), ([N\(_{2222}\)] [Gly]), ([N\(_{111}\)] [Lys]) and ([N\(_{2222}\)] [Lys]) to amine mixtures greatly reinforced \( \text{CO}_2 \) absorption.

In contrast, Fu and Zhang [23] found that \( \text{CO}_2 \) loading decreased significantly when 1-butyl-3-methylimidazolium glycinate [bmim][Gly] was added to the methylidiethanolamine 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 \( \text{CO}_2 \) absorption efficiency. They conducted the experiment using two low viscous ionic liquids; [C\(_2\)OHmim][DCA] and [bmim][DCA], mixed with an aqueous MEA solution. The authors reported that both [C\(_2\)OHmim]
[DCA] and [bmim][DCA] gave slightly reduced solubility, due to salting out effects that inhibited CO₂ absorption.

Ahmady et al., [25] also found that the amount of CO₂ 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 CO₂ absorption, as [gua]+[OTf] hindered the tertiary amine of MDEA.

Table 2 shows summarizes recent research findings in CO₂ capture using amine and ionic liquid mixtures.

<table>
<thead>
<tr>
<th>Amine</th>
<th>Ionic Liquids</th>
<th>CO₂ Loading</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>MEA</td>
<td>• hydrophilic amino acid ionic liquid [C₂OHmim][Gly]</td>
<td>Increase</td>
<td>[20]</td>
</tr>
<tr>
<td>MEA</td>
<td>• bis(2 hydroxyethyl)ammonium acetate (bhea) • 1-butyl-3-methylimidazolium tetrafluoroborate [bmim][BF₄]</td>
<td>Increase</td>
<td>[21]</td>
</tr>
<tr>
<td>MDEA</td>
<td>• tetramethylammonium glycinate [N₁₁₁₁][Gly], • tetraethylammonium glycinate [N₂₂₂₂][Gly] • tetramethylammonium lysinate [N₁₁₁₁][Lys] • tetraethylammonium lysinate [N₂₂₂₂][Lys]</td>
<td>Enhance</td>
<td>[22]</td>
</tr>
<tr>
<td>MDEA</td>
<td>• 1-butyl-3-methylimidazolium glycinate [bmim][Gly]</td>
<td>Decrease</td>
<td>[23]</td>
</tr>
<tr>
<td>MEA</td>
<td>• 1-(2-hydroxyethyl)-3-methylimidazolium dicyanamide [C₂OHmim][DCA] • 1-butyl-3-methylimidazolium [Bmim][DCA]</td>
<td>Decrease</td>
<td>[24]</td>
</tr>
<tr>
<td>MDEA</td>
<td>• 1-butyl-3-methyl-imidazolium tetrafluoroborate [bmim][BF4] • 1-butyl-3-methyl-imidazolium acetate [bmim][Ac] • 1-butyl-3-methyl-imidazolium dicyanamide [bmim][DCA]</td>
<td>Decrease</td>
<td>[25]</td>
</tr>
</tbody>
</table>
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-hydroxyethyl) 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 CO₂ solubility pattern is in agreement with findings of other researchers. Feng et al., [22] used four amino acid based ILs: tetramethylammonium glysinate [N₁₁₁₁][Gly], tetraethylammonium glysinate [N₂₂₂₂][Gly], tetramethylammonium lysinate [N₁₁₁₁][Lys], and tetraethylammonium lysinate [N₂₂₂₂][Lys] to investigate the effect of temperature ranging 298K to 318K towards the absorption of CO₂ 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 CO₂ amount being absorbed.

This similar trend was also seen by Xu et al., [24] in their work where CO₂ solubility decreased with an increase in temperature. The authors used two low viscous ionic liquids; [C₂OHmim][DCA] and [Bmim][DCA] mixed with aqueous 30 wt% MEA. In terms of pressure effects toward CO₂ absorption, results showed that CO₂ solubility was directly proportional to the increase in CO₂ partial pressure.

The study of the effects of temperature and pressure on CO₂ 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 absorption by conducting an experiment, using aqueous 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$_2$N], 1-octyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [omim][Tf$_2$N] and 1-decyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [dcmim][Tf$_2$N] 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$_2$N], 1-butyl-3-Methylimidizolium Bis (trifluoromethylsulfonyl) imide [bmim][Tf$_2$N] 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]
[\text{Tf}_2\text{N}] > [\text{bmim}] [\text{Tf}_2\text{N}] > [\text{bmim}] [\text{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] [\text{MeSO}_4] and tributylmethylphosphonium methylsulfate [TBMP][\text{MeSO}_4] for capturing \text{CO}_2. The authors compared the solvents with with commonly used industrial ILs. Their findings revealed that both [TBMN][\text{MeSO}_4] and [TBMP][\text{MeSO}_4], had slightly higher solubility compared to 1-butyl-3-methylimidazolium methylsulfate [bmim][\text{MeSO}_4] [40]. However, by comparing with 1-butyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [bmim][\text{Tf}_2\text{N}] [41], obviously fluorinated anion has a much larger effect on solubility. This explanation was strongly supported by the sequences of \text{CO}_2 solubility reported by Sharma et al., [42] where; \text{BF}_4^- < \text{DCA}^- < \text{PF}_6^- < \text{TfO}^- < \text{Tf}_2\text{N}. The anion effect on \text{CO}_2 solubility was attributed to the presence of the fluoroalkyl group, which led to highest \text{CO}_2 solubility [43][44][45][46], and due to the weaker ILs cation-anion interactions. Table 3 lists anion classification and \text{CO}_2 solubility.

**Table 3: Influence of Anions in Different Ionic Liquids**

<table>
<thead>
<tr>
<th>Anion</th>
<th>Nomenclature</th>
<th>Classification</th>
<th>\text{CO}_2 solubility</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tetrafluoroborate</td>
<td>\text{BF}_4</td>
<td>Fluorinated</td>
<td>High</td>
</tr>
<tr>
<td>Hexafluorophosphate</td>
<td>\text{PF}_6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Trifluoromethanesulfonate</td>
<td>\text{TfO}</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bis (trifluoromethylsulfonyl) imide</td>
<td>\text{Tf}_2\text{N}</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tris (trifluoromethylsulfonyl) methide</td>
<td>methide</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dicyanamide</td>
<td>\text{DCA}</td>
<td>Non fluorinated</td>
<td>Low</td>
</tr>
<tr>
<td>Nitrate</td>
<td>\text{NO}_3</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Effects of Ionic Liquids / Amines Concentrations

Recently, Lv et al., [7] compared the CO₂ capture into different mole ratios of MEA / [C₂OHmim] [Gly] mixtures. They found that the absorption capacity of aqueous MEA solution was lower than pure ionic liquid. The absorption capacity increased with increasing [C₂OHmim] [Gly] concentration for mixed solutions of MEA and [C₂OHmim] [Gly].

Nordin et al., [28] evaluated the interaction of 2- amino-2-methyl-1 propanol (AMP) with N-butyl-3-methylpyridinium tetrafluoroborate, [B₃MPYR][BF₄]. The amount of CO₂ absorbed was found to decrease with increasing of [B₃MPYR][BF₄] 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 CO₂ absorption [27]. Blends of methyldeanolamine (MDEA) with three different types of ionic liquids, 1-butyl-3-methylimidazolium tetrafluoroborate [bmim] [BF₄], 1-butyl-3-methylimidazolium acetate [bmim][Ac] and 1-butyl-3-methylimidazolium 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 CO₂ absorption and CO₂ 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 decreased presence of water molecules in the solution. Anions and cations significantly affects CO₂ absorption. Solubility is more affected by the nature of anions, compared to cations. Ionic liquids with fluoroalkyl groups absorb a high amount of CO₂ compared to non-fluorinated ionic liquids. Cation alkyl chains impart a slight effect on CO₂ solubility. Increasing the chain length will slightly increase CO₂ solubility. Selecting the right combination of amine-ionic liquid mixtures can improve the performance of CO₂ absorption.
ACKNOWLEDGEMENT

Financial Support from Fundamental Research Grant Scheme (600-RMI/FRGS 5/3 (91/2014)) for this project is gratefully acknowledged. The authors also would like to thank Universiti Teknologi MARA (UiTM) for supporting the completion of this research work.

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