

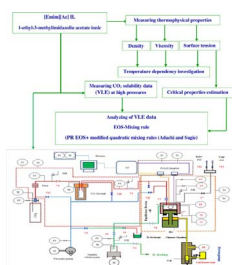
Experimental and theoretical study of CO₂ solubility under high pressure conditions in the ionic liquid 1-ethyl-3-methylimidazolium acetate



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GRAPHICAL ABSTRACT



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ABSTRACT

CO₂ solubility into [Emim][Ac] IL at high pressures and temperatures measured and predicted by using a combination of PR EOS and mixing rule that has not been previously applied for analyzing the CO₂ + [Emim][Ac] IL system. The vapor-liquid equilibria of CO₂ in [Emim][Ac] IL has been investigated experimentally within temperature range of 293.15–363.15 K and pressures of 0.03–60 bar. The bubble point pressures of the binary system were presented for CO₂ concentrations of 10.2–52.1 mol%. The density, viscosity and surface tension of pure [Emim][Ac] IL were measured and the temperature dependency of the properties investigated. The PR EOS with the quadratic mixing rules modified by Adachi and Sugie has been used to analyze the measured vapor-liquid equilibria. The given results were compared with previous data reported in the literature. Finally, the regeneration process of [Emim][Ac] IL showed that complete CO₂ desorption is possible.

1. Introduction

Global warming is one of the unequivocal and palpable consequences of climate change [1]. It is extremely likely that the impact of carbon dioxide (CO₂) emissions has been the dominant cause of climate change and global warming since the mid-20th century [2]. Because of the key role of growing world energy from fossil fuels in the upward trend of CO₂ emissions, further warming will inevitably occur in the decades to come [3]. It is a well-known fact that limiting the climate change and global warming would require substantial and sustained mitigations in the greenhouse gas emissions [4–8]. As a result, efficient

and environment friendly CO₂ capture and sequestration (CCS) technologies are necessary to reduce the impact of anthropogenic CO₂ emissions [9,10].

Industries like thermal fired power plants, petrochemicals, and cement manufacturing plants which use fossil fuels, are the major sources of CO₂ emissions into atmosphere [10]. The fossil fuel burning processes are classified as post-combustion, pre-combustion, and oxy-combustion [11]. The choice of the method mainly depends on the burning process, leading to development of a wide range of CO₂ capture technologies. There have been many attempts for post combustion capture of CO₂ from the flue gas streams of the mentioned industries

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Nomenclature

AARD	Absolute average relative deviation
[Ac] [−]	Acetate anion
a	Equation of state parameter
a _i	Equation of state parameter for pure component ‘i’
b	Equation of state parameter
b _i	Equation of state parameter for pure component ‘i’
[Emim] ⁺	1-ethyl-3-methylimidazolium cation
f	Fugacity [Pa]
IL	Ionic liquid
K _{ij}	Binary interaction parameter between two components i and j
l _{ij}	Binary interaction parameter between two components i and j
M	Molecular weight (kg kmol ^{−1})
N	Number of data points
P	Pressure [Pa]
R	Ideal gas constant [J mol ^{−1} K ^{−1}]
T	Temperature[K]
ϑ	Molar volume[m ³ gmol ^{−1}]
V	Volume[m ³]
VLE	Vapor–liquid equilibrium

x Mole fraction in the liquid phase
 C, D, A', B', C₁, C₂ Constant

Greek letters

λ _{ij}	Binary interaction parameter between two components i and j used in Eq. (5)
ρ	Density[Kg.m ^{−3}]
φ _i	Fugacity coefficient of component i
ω	Acentric factor
γ _i	Activity coefficient of component ‘i’
δ	Surface tension [mN m ^{−1}]
α	Equation of state parameter
η	Viscosity [Pa s]
φ	Fugacity coefficient symbol
χ	Chi. symbol
σ	Sample standard deviation

Subtitles

Exp.	Experiment
Calc.	Calculated
Ave.	Average

because of its easier retrofitting advantages and huge volumes of CO₂ emitted [12,13]. Post-combustion involves CO₂ capture from the gas streams produced after combustion of fossil fuels or other carbonaceous materials. In these methods, the thermodynamic driving force for CO₂ capture from the gas streams is low because of its relatively small partial pressure (5–15 vol% at atmospheric pressure) [13,14]. For this reason, a solvent with strong absorption capability is required to efficiently capture CO₂ from the flue gas stream.

One of the most common and cost-effective technologies in the post-combustion method is the liquid-gas chemical contact. Among the chemicals used, the selective CO₂ absorption by aqueous amine solvents such as monoethanolamine (MEA) is still the most popular process [15–19]. The major drawback of this process is its high energy penalty in solvent regeneration step. Amine processes also suffer a number of other disadvantages that make them unattractive for large scale applications [20–24]. Therefore, much research focused on the improvement of absorbent formulation and process efficiency to efficiently capture CO₂ from the huge post combustion flue gases emit from these plants [25]. To cope MEA disadvantageous, liquid organic salts which do not easily form an ordered crystal and remain liquid near room temperature, have emerged as a promising alternative to the amines in recent decades. These salts composed of big heterocyclic organic cations and small anions, namely, ionic liquids (ILs). ILs have remarkable properties like negligible vapor pressure, lower specific heat capacity than water, favorable CO₂ solubilities, high thermal stability, non-corrosivity, reusability, and ability to remain liquid over a large temperature ranges [15,26–30]. Compared to conventional amine-based methods, CO₂ capture by ILs needs low energy for regeneration of solvent [15,16]. Another attractive property of ionic liquids is the fact that their physical and chemical properties are tuneable. This depends on the cation and anion present in their structure, making them to be considered as designer solvents [29,30].

Due to the high degree of CO₂ solubility in selected ILs, interest has recently increased in ILs for applications involving CO₂ separation [4–6]. Among the broad diversity of ILs, those based on an imidazolium cation typically present a higher degree of CO₂ solubility, which could be even higher with the appropriate IL anion [26]. The catalyzer of this research field was a work from Blanchard et al., reporting a high solubility of CO₂ in imidazolium-based ionic liquids [32]. The solubility of CO₂ has been mostly studied in imidazolium-based ionic liquids

pairing with different anions. Other cations like pyridinium, pyrrolidinium, phosphonium and ammonium-based cation have also been investigated [31]. Many researches showed that solubility of CO₂ in imidazolium based ionic liquids is very high [30–33].

The absorption of CO₂ in non-functionalized IL occurs based on physical absorption mechanism and high solubility is only achieved at high pressures [28,30–32]. It is a well-known fact that CO₂ possesses relatively high solubility in ILs, but most of the literature provides only examples of physical absorption [15,32]. According to solution-diffusion theory, the most effective ILs dissolve large amounts of CO₂, but they do not bind gas molecules too strongly. Chemical absorption occurs in ILs which contain a significantly basic group such as an amino group, a super basic moiety or a carboxylate anion [34–36]. On the other hand, through adding either the fluorinated group or amines and acetates on the IL ions, they are functionalized to chemically react with CO₂ [28,30,33–35].

Recently, certain classes of ILs with improved hydrogen bonding acceptor capability such as acetate-based fluids have shown to be promising solvents for CO₂ capture. Acetate-based fluids are able to strongly coordinate with CO₂ and hydrogen bond donor groups, and favorable results have thus been published. Moreover, acetate-based ILs present low toxicity, low corrosiveness, and favorable biodegradability. Despite their undeniable interest, the thermophysical properties of acetate-based ILs are still poorly characterized [24,27,33]. ILs contain the acetate anion [CH₃–COO[−]] exhibit a strong absorption for CO₂ in a wide range of temperatures and pressures [32–35]. Shiflett et al., Gurau et al., Gomez-Coma et al., Krupiczka et al. reported that imidazolium-based ionic liquids contain a carboxylic anion such as [Emim][Ac] and [Bmim][Ac] are able to chemically react with CO₂ [36–40]. Gurau et al. with experimental evidence confirmed formation of the reversible imidazolium carboxylate complex and acetic acid in CO₂ reaction with [Emim][Ac], and proposed a mechanism of forming reversible chemical complexes for the chemisorption of [Emim][Ac] IL with CO₂ [40]. Shiflett et al., Gomez-Coma et al. and Jongpitisub et al. suggested that these ILs can candidate as an IL solvent for post combustion capturing [37,38,41]. However, the tunability of ILs has made possible the substitution of common anions for others with a metal ion, which confers them a magnetic response. These magnetic ionic liquids (MILs) are an interesting approach in selective separation processes [29,30]. Except to MILs, [Emim][Ac] IL has the highest CO₂ solubility among the ILs

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