



Study on the use of an imidazolium-based acetate ionic liquid for CO₂ capture from flue gas in absorber/stripper packed columns: Experimental and modeling

Fazlollah Zareiekordshouli, Asghar Lashanizadehgan*, Parviz Darvishi

Department of Chemical Engineering, School of Engineering, Yasouj University, Yasouj, Iran

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ABSTRACT

In the present work, 1-Ethyl-3-methylimidazolium acetate ([Emim][Ac]) ionic liquid (IL) has been considered for experimental and theoretical investigation of post-combustion carbon dioxide (CO₂) capture from flue gas. The absorption and stripping of CO₂ into [Emim][Ac] IL has been studied in a typical absorber/stripper system that randomly packed with Raschig ring at absorption pressures of 5–8 bar, absorption temperatures of 298.15–338.15 K and stripping conditions of 1.5 bar in temperature range 363.15–398.15 K. A mathematical model was developed for absorption and stripping processes based on mass transfer concepts and Peng–Robinson equation of state (PR EOS). The validity of the model was verified via comparison of the results achieved by the model with data taken from the experiments performed in this work and VLE data given in the literature. The impacts of parameters such as absorption/stripping pressure and temperature on the performance of CO₂ capture, the sorbent flow rate and energy demand at selected operating conditions and specified CO₂ capture rates were examined. The experimental tests showed that the recovered CO₂ from the stripper column was pure. The results demonstrated that the energy requirement for the CO₂ capture IL-based process is about 4890 kW or 2.75 GJ/t CO₂. It was also found that the degradation rate of ion liquid is 3.78 wt.% of circulated IL. Using the enhancement factor obtained based on experimental results, the pseudo-first order reaction constant of the CO₂ + [Emim][Ac] IL system was estimated. By fitting the kinetics data into Arrhenius equation, the activation energy and frequency factor of the reaction rate constant were found to be 10.317 kJ/mol and 1545 s⁻¹, respectively.

1. Introduction

Carbon dioxide as a major greenhouse gas, which is mainly emitted from the burning of fossil fuels, causes palpable global warming and climate change that the world faces today (Yu et al., 2012). Therefore, it is a highly clear fact that reduction of anthropogenic CO₂ emissions is vital for human beings and all other lives on the earth (Finkenrath, 2011). The most efficient way to reduce carbon dioxide emissions may be the post-combustion CO₂ capture (IPCC, 2013). The post-combustion capture route is ideally applicable for conventional power stations and energy conversion systems and can be applied to retrofit the existing power plants (Energy Technology Perspectives, 2008). Post-combustion capture is the separation of CO₂ from the flue gas that is produced from combustion of fossil fuels and is mainly diluted with nitrogen (Mac Dowell et al., 2010). Today, amine-based absorption is one of the most popular technologies in the post-combustion CO₂ capture due to the high tendency of amines (MEA, MDEA and DEA) to react with CO₂

(Kohl and Nielsen, 1997). It is well-known that amine solvents suffer from high energy penalty, corrosion, thermal degradation and solvent loss that may limit their usage (Zhao et al., 2011; Wang et al., 2015). Considerable scientific efforts have been made with the emphasis on identifying new solvents to develop more efficient processes (Hu et al., 2016). The liquid organic salts (ion liquids) are regarded as attractive kinds of novel solvents to overcome the disadvantages of amine solvents in CO₂ capture. Ion liquids (ILs) compose of big heterocyclic organic cations and various small anions. ILs have remarkable properties such as extremely low vapor pressure, tunable structure, high thermal and chemical stability, low demand energy for regeneration and excellent solvent power (Wasserscheid and Welton, 2008; D'Alessandro et al., 2010; Blanchard et al., 2001; Ramdin et al., 2012; Feng et al., 2011).

In post-combustion CO₂ capture from the flue gas flows, the concentration of CO₂ in the inlet stream is less than 15% (v/v). In this respect, the thermodynamic driving force for physical absorption of

* Corresponding author.

E-mail address: lashani@yu.ac.ir (A. Lashanizadehgan).

Nomenclature

| | |
|---------------------------------------|--|
| A | Column cross section area (m^2) |
| a | Packing specific area ($\text{m}^2 \text{m}^{-3}$) |
| a_i, b_i | Component specific parameters in PREOS |
| B | Constant parameter in PR EOS |
| C | Concentration, (kmol m^{-3}) |
| D | Column diameter (m) |
| D | Differential symbol |
| D_{CO_2} | Molecular diffusivity of CO_2 ($\text{m}^2 \text{s}^{-1}$) |
| $D_{\text{CO}_2-\text{w}}$ | Molecular diffusivity of CO_2 in water ($\text{m}^2 \text{s}^{-1}$) |
| $D_{\text{CO}_2-\text{IL}}$ | Molecular diffusivity of CO_2 in ionic liquid ($\text{m}^2 \text{s}^{-1}$) |
| E_A | Enhancement factor |
| E_{act} | Activation energy (kJ mol^{-1}) |
| F | Molar flow rate (kmol s^{-1}) |
| G | Superficial molar gas velocity ($\text{kmol m}^{-2} \text{s}^{-1}$) |
| G' | Non-diffusing super facial molar flow ($\text{kmol m}^{-2} \text{s}^{-1}$) |
| g | Gas |
| $H_{\text{CO}_2-\text{liquid}}$ | Henry's constant of CO_2 in liquid (kPa) |
| H | Henry's constant |
| Ha | Hatta number |
| IL | Ionic liquid |
| in | Inlet of the column |
| k_F | Forward rate constants of Reaction (1) ($\text{kmol}^{-1} \text{m}^3 \text{s}^{-1}$) |
| k_{-F} | Backward rate constants of Reaction (2) ($\text{kmol}^{-1} \text{m}^3 \text{s}^{-1}$) |
| k | Apparent first order rate constant (s^{-1}) |
| k_0 | Pseudo-first order rate constant (s^{-1}) |
| k_1, k_3 | Rate constant ($\text{kmol}^{-1} \text{m}^3 \text{s}^{-1}$) |
| k_2, k_4 | Rate constant (s^{-1}) |
| k_{IL} | Rate constant for IL ($\text{kmol}^{-2} \text{m}^6 \text{s}^{-1}$) |
| $k_{\text{IL}-\text{CO}_2\text{BIL}}$ | Rate constant of CO_2 binded-IL ($\text{kmol}^{-2} \text{m}^6 \text{s}^{-1}$) |
| k_{00} | Frequency factor (s^{-1}) |
| k_g | Mass transfer coefficient in gas phase (m s^{-1}) |
| K_g | Overall mass transfer coefficient based on gas phase (m s^{-1}) |
| k_{ij} | Binary interaction coefficients |

Greek letters

| | |
|----------------|--|
| $\dot{\phi}_G$ | Gas flow rate (kmol s^{-1}) |
| η | Viscosity (Pa s) |
| μ | Viscosity (Pa s) |
| β | Fraction of vaporized |
| λ_{ij} | Binary interaction coefficients |
| ω | Acentric factor |
| κ_i | Equilibrium constant |

Notation for anions/cations

| | |
|---|---|
| $[\text{Emin}]^+$ or $[\text{C2mim}]^+$ | 1-Ethyl-3-methylimidazolium cation |
| $\text{C}_2\text{mim}^+ - \text{COO}^-$ | 1-Ethyl-3-methylimidazolium-2-carboxylate |

Subscripts

| | |
|---------------|-------------------|
| A | Desired component |
| a | Absorber |
| C | Critical state |
| CO_2 | Carbon dioxide |

| | |
|---|---|
| i | Component |
| l, L | Liquid |
| m | Mixture |
| g | Gas |
| LMTD | Log mean temperature difference, °C |
| ij | Pairs of i and j |
| K_1 | Overall mass transfer coefficient based on liquid phase (m s^{-1}) |
| k_1 | Mass transfer coefficient in liquid phase (m s^{-1}) |
| k_L^* | Liquid mass transfer coefficient without reaction (m s^{-1}) |
| l | Liquid |
| L | Superficial molar liquid velocity ($\text{kmol m}^{-2} \text{s}^{-1}$) |
| m' | Packing factor |
| MEA | Monoethanolamine |
| n' | Packing factor |
| N_{CO_2} | Molar flux of CO_2 ($\text{kmol m}^2 \text{s}^{-1}$) |
| out | Outlet of the column |
| P | Total pressure (kPa) |
| $P_{\text{CO}_2}^b$ | Partial pressure of CO_2 in bulk (kPa) |
| $P_{\text{CO}_2}^i$ | Partial pressure of CO_2 in interface (kPa) |
| $P_{\text{CO}_2}^*$ | Equilibrium partial pressure of CO_2 in liquid phase (kPa) |
| ΔP | Mass transfer driving force (kPa) |
| R | Ideal gas constant ($\text{J mol}^{-1} \text{K}^{-1}$) |
| R_{liquid} | Mass transfer resistance in liquid phase |
| Re | Reynolds number |
| r | Reaction rate ($\text{kmol m}^{-3} \text{s}^{-1}$) |
| Sc | Schmidt number |
| Sh | Sherwood number |
| T | Temperature (K) |
| t | Time (s) |
| V | Velocity (m s^{-1}) |
| x | Liquid molar fraction |
| X | Diffusing mole to non-diffusing moles in the liquid phase |
| y | Gas molar fraction |
| Y | Diffusing mole to non-diffusing moles in the gas phase |
| Z | Height (m) |
| z | Compressibility factor |
| w | Water |
| α | Temperature-dependent parameter in the EOS |
| ρ | Density (kg m^{-3}) |
| ε | Bed porosity |
| δ | Film thickness (m) |
| f | Fugacity (kPa) |
| ν | Molar volume ($\text{m}^{-3} \text{mol}^{-1}$) |
| z_i | Mole fraction in feed to stripper |
| $[\text{Ac}]^{-1}$ or $[\text{CH}_3\text{COO}]^{-}$ | Acetate anion |

Superscripts

| | |
|---|-------------|
| m | Mixture |
| s | Stripper |
| g | Gas |
| V | Vapor |
| e | Equilibrium |
| i | Interface |
| b | Bulk |

CO_2 is low and its removal cannot be efficiently accomplished. In order to achieve an efficient CO_2 capture, a solvent with strong absorption capacity is required (Torralba-Calleja et al., 2013; Bates et al., 2002; Wang et al., 2013). It is noteworthy that this strong absorption capacity is achieved when chemical absorption (which must be reversible) is

occurred in addition to simple physical absorption.

Shiflett and Yokozeki (Shiflett and Yokozeki, 2009), Yokozeki et al. (Yokozeki et al., 2008) and Gomez-Coma et al. (Gomez-Coma et al., 2014) showed that ILs containing the acetate anion, $[\text{CH}_3 - \text{COO}]^{-}$, exhibit a reactive absorption for CO_2 capture. Carvalho et al. (Carvalho

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