



Feasible ionic liquid-amine hybrid solvents for carbon dioxide capture



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ABSTRACT

Ionic liquid (IL)-amine hybrid solvents have been proved to be an energy-saving system for CO₂ capture. Lack of experimental information on the diffusivity and heat of CO₂ absorption in IL-amine hybrid solvents hampers the wide application of IL in CO₂ capture. In this work, the effects of IL on absorption rate, solubility, diffusivity and heat of CO₂ absorption in feasible hybrids of 1-butyl-3-methyl-imidazolium nitrate ([Bmim][NO₃]) and monoethanolamine (MEA) have been elaborated using weight method, vapor liquid equilibrium and calorimetric measurement. The results showed that the addition of IL slightly decreases CO₂ loading under atmospheric pressure. Based on the measured solubility and calorimeter results, IL is beneficial to improve the physical absorption of CO₂ and reduce the heat of absorption in IL-amine hybrids. The diffusivity of CO₂ can be maintained at an acceptable level by controlling the concentration of IL. Considering CO₂ capacity, kinetics and heat of absorption, the hybrids of 30% MEA + 10% [Bmim][NO₃] + 60% H₂O were recommended. Our work gives the meaningful insights on designing novel IL-amine hybrids for CO₂ capture.

1. Introduction

CO₂ capture is believed to be the most efficient strategy for mitigating greenhouse gas (GHG) emissions in recent decades (Sun et al., 2015; Zhang et al., 2014a). Chemical absorption using aqueous amine solution has been the commercialized benchmark in post-combustion CO₂ treating process (Haszeldine, 2009; Jun et al., 2015; Rochelle, 2009). To date, a variety of amines have been developed to remove CO₂ from flue gas mixtures. However, there are still several drawbacks concerning amine candidates. On one hand, the single amine cannot satisfy the requirements of fast absorption rate, high capacity, and low reaction enthalpy (Cao et al., 2015a; Zhu et al., 2011). On the other hand, the amine solvents suffer from inherent problems such as equipment corrosion, evaporative losses and high energy demand of regeneration (Gao et al., 2015b; Wang et al., 2011). Therefore, the interests of more efficient solvents for CO₂ capture are still expanding.

In these decades, ionic liquid (IL) have attracted considerable attention for CO₂ capture due to their nonvolatility, thermal stability, tunable properties, etc (Bates et al., 2002; Cadena et al., 2004; Huang and R  ther, 2009; Isik et al., 2016; Zhang et al., 2009). For example,

the negligible vapor pressure of IL can reduce solvent loss and make IL more environmentally benign than other volatile organic solvents. Also, the nature of tunable structure of IL offers unparalleled opportunity for improving the absorption rate and capacity of CO₂. However, it has been proved that CO₂ capture using IL encounters some inherent defects both for conventional and task-specific ionic liquid (TSIL). The conventional IL usually shows low CO₂ capacity due to their physical absorption (Blanchard et al., 2001; Cadena et al., 2004). Although the absorption capacity of CO₂ in TSIL can be greatly improved by tethering specific functional groups (Bates et al., 2002; Zhang et al., 2013), the synthesis of TSIL needs several steps and the following purification is generally troublesome. Another thorny challenge is the high viscosity of TSIL that causes mass transfer limitation.

Alternatively, the IL-amine hybrid solvents for CO₂ capture are proved to be competitive in recent years (Ahmady et al., 2011b; Gal  n S  nchez et al., 2011; Taib and Murugesan, 2012; Zhang et al., 2012a). Bara et al. first put forward the idea of IL-amine hybrids and their results showed that the hybrid solvents are prone to industrial applications (Bara et al., 2010; Camper et al., 2008). Until now, several IL-amine hybrid solvents have been formulated. The significant advantage

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Nomenclature

List of symbols

B , B_0 and B_1	Virial coefficient
C_p	Heat capacity, $J g^{-1} K^{-1}$
C_{MEA}	Concentration of MEA, $kmol m^{-3}$
C_{N_2O}	Concentration of N_2O in the liquid phase, $mol L^{-1}$
$H_{CO_2, hybrid}$	Physical solubility of CO_2 in solvent, $kPa m^3 kmol^{-1}$
$H_{N_2O, hybrid}$	Physical solubility of N_2O in the solvent, $kPa m^3 kmol^{-1}$
H_{CO_2, H_2O}	Physical solubility of CO_2 in water, $kPa m^3 kmol^{-1}$
H_{N_2O, H_2O}	Physical solubility of N_2O in water, $kPa m^3 kmol^{-1}$
$n_{CO_2}^g$	Total mole of CO_2 released from the gas reservoir, mol
$n_{CO_2}^l$	Mole of CO_2 in gas phase in equilibrium cell, mol
$n_{CO_2}^l$	Mole of CO_2 absorbed in liquid phase, mol
$n_{N_2O}^g$	Mole of N_2O in gas phase in equilibrium cell, mol
$n_{N_2O}^l$	Mole of N_2O dissolved in liquid in equilibrium cell, mol
n_{N_2O}	Mole of N_2O introduced into the equilibrium cell, mol
n_s	Mole of solvent in reactor, mol
P	Equilibrium pressure, kPa
P_c	Critical pressure, kPa
P_{Ea}	Equilibrium pressure of the equilibrium cell, kPa
P_{Eb}	Initial pressure of the equilibrium cell, kPa
P_{Gb}	Equilibrium pressure of the gas buffer vessel, kPa
P_{Ga}	Initial and pressure of the gas buffer vessel, kPa
R	Gas constant, $J mol^{-1} K^{-1}$
T	Absolute temperature, K
T_c	Critical temperature, K
T_r	Reduced temperature
V	Volume, cm^{-3}
V_{EC}	Volume of the equilibrium cell, l
V_{GR}	Volume of the gas buffer vessel, l
V_s	Volume of solvent, l
w	Mass fraction
X_0 , X_1 and X_2	Regression parameters of density
Y_0 , Y_1 and Y_2	Regression parameters of viscosity
z	Compressibility factor

Abbreviation

[Bmim][Acetate] 1-Butyl-3-methyl-imidazolium acetate

[Bmim][NO ₃]	1-Butyl-3-methyl-imidazolium nitrate
[Bmim][BF ₄]	1-Butyl-3-methyl-imidazolium tetrafluoroborate
[Bmim][PF ₆]	1-Ethyl-3-methylimidazolium hexafluorophosphate
[Emim][NTf ₂]	1-Ethyl-3-methyl-imidazolium bis(trifluoromethyl) sulfonylimide
[Emim][EtSO ₄]	1-Ethyl-3-methylimidazolium ethylsulfate
[Emim][TfO]	1-Ethyl-3-methylimidazolium tri-fluoromethanesulfonate
CO ₂	Carbon dioxide
CH ₄	Methane
EC	Equilibrium cell
GC-EOS	Group contribute equation of state
GR	Gas reservoir
IL	Ionic liquids
MDEA	<i>N</i> -methyldiethanolamine
MEA	Monoethanolamine
[N ₁₁₁₁][Gly]	Tetramethylammonium glycinate
N ₂	Nitrogen
N ₂ O	Nitrous oxide
[Omim][NTf ₂]	1-Octyl-3-methylimidazolium bis(tri-fluoromethylsulfonyl)imide
PZ	Piperazine
TSIL	Task-specific ionic liquids

Greek letters

α	CO ₂ loading, mol CO ₂ per mol solvent
ρ	Density, $g cm^{-3}$
η	Viscosity, Pa s
ω	Centric factor

Superscripts & subscripts

a	After CO ₂ absorption
b	Equilibrium
G	Gas
g	Gas phase
i	the number of component
l	Liquid phase

of the hybrid solvents are their potentials in eliminating the energy requirements compared with conventional amine scrubbing process. Huang et al. has proved that the IL-amine hybrid solvents can save 15% regeneration heat duty compared with the commercialized MEA technology (Huang et al., 2014). Very recently, Ma et al. studied CO₂ capture using process simulation and found that the energy consumption in 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF₄]) and [Bmim][PF₆] based processes could be lowered up to 26.7% and 24.8% respectively than that of MEA-based process (Ma et al., 2017). In addition, from a kinetic point of view, the IL-amine hybrid solvents have advantages in CO₂ mass transfer because the viscosity of the IL tailored solvents can be hugely decreased compared with the pure ILs, which facilitates CO₂ diffusion in liquid phase. Kinetic characteristics of CO₂ absorption in [Bmim][BF₄] based IL-amine hybrids were studied by some authors (Ahmady et al., 2012; Lu et al., 2013). However, the fluorine contained ILs are reported to be unstable and are harmful because of the potentials releasing hazardous hydrogen fluoride (HF) (Villagran et al., 2004). Recently, 1-butyl-3-methyl-imidazolium nitrate ([Bmim][NO₃]) have aroused public attention in CO₂ capture (Bermejo et al., 2009; Hong et al., 2014; Zhang et al., 2014b) because it is more stable compared with other ILs like 1-butyl-3-methyl-imidazolium acetate ([Bmim][Ac]) (Shiflett et al., 2010). Also, [Bmim][NO₃] is

cheaper than some ILs such as tetramethylammonium glycinate ([N₁₁₁₁][Gly]) (Zhang et al., 2012b). Zhang et al. (2014b) suggested [Bmim][NO₃] tailored system for CO₂ capture considering the kinetics and stability of the solvents. However, the liquid-side mass-transfer coefficient of the CO₂ in this system was estimated using a recommended correlation coefficient, where the Henry's constants of CO₂ in these hybrids are not directly measured.

To our knowledge, the experimental information on the CO₂ absorption in MEA-[Bmim][NO₃] is still insufficient. Especially, direct measurements of the heat of CO₂ absorption in MEA-[Bmim][NO₃] are not reported until now. Also, physical absorption of CO₂ in this reactive solvent is rarely determined. Based on our previous work (Cao et al., 2015b; Gao et al., 2015a; Huang et al., 2014; Zhang et al., 2014b), the absorption performance of CO₂ in [Bmim][NO₃]-MEA hybrids systematically studied.

2. Experimental section

2.1. Materials

MEA was purchased from Sinopharm Chemical Reagent Co., Ltd., and used without further purification. [Bmim][NO₃] was obtained from

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