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CO_2 capture with the absorbent of a mixed ionic liquid and amine solution considering the effects of SO_2 and O_2

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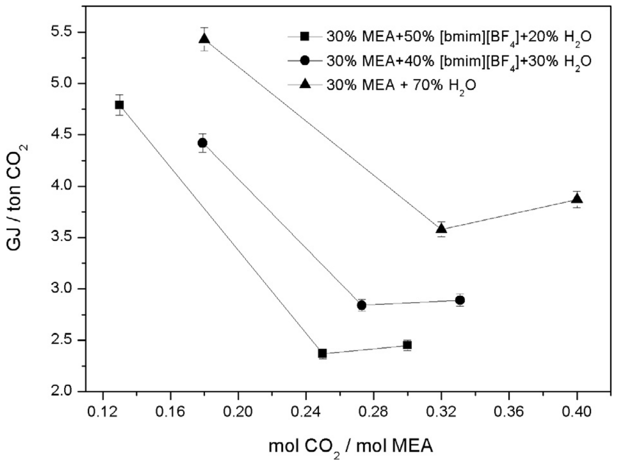
HIGHLIGHTS

- The addition of IL to MEA aqueous solution reduced the losses of MEA and water.
- O_2 and SO_2 played less important roles in the losses of MEA and water than evaporation.
- The thermal energy at stripper of IL mixed absorbent is 33.8% lower than that of aqueous MEA solution.

GRAPHICAL ABSTRACT



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ABSTRACT

Room-temperature ionic liquids (ILs) have recently been proposed as a potential candidate for CO_2 capture. In this study, experiments were conducted in an absorption-desorption loop system to investigate the effects of SO_2 and O_2 on CO_2 capture using an aqueous amine solution mixed with IL. The gas mixture containing CO_2, O_2, SO_2 and N_2 in the composition range of flue gas from a coal-fired power plant after flue gas desulfurization was selected as the feed gas. It was found that the addition of hydrophilic IL of 1-butyl-3-methylimidazolium tetrafluoroborate ([bmim][BF_4]) to a monoethanolamine (MEA) aqueous solution reduced the losses of MEA and water by lowering the saturated vapour pressure of the mixed absorbent. For hydrophobic IL of 1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([hmim][Tf_2N]), the MEA loss for 30 wt% MEA + 70 wt% [hmim][Tf_2N] increased dramatically with the system running because carbonate, which was formed by MEA reacting with CO_2, was insoluble in [hmim][Tf_2N] at the absorber operation temperature of 323 K. The effects of O_2 and SO_2 were relatively insignificant for the MEA and water losses. The aqueous amine solution mixed with [bmim][BF_4] showed good performances with a CO_2 removal efficiency of above 90% and the SO_2 concentration at the absorber

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Nomenclature

C_{in}	CO_2 volumetric fractions at the gas inlet, %
C_{out}	CO_2 volumetric fractions at the gas outlet, %
Q_{in}	gas flow rates at the inlet, $m^3\ s^{-1}$
Q_{out}	gas flow rates at the outlet, $m^3\ s^{-1}$
E_{loss}	heat loss, J
E_{reg}	heat energy requirement of regeneration of the absorbent solutions, J
U_{oil}	voltage provided to the heating rod for the heat-treatment silicone oil, V
U_{samp}	voltage provided to the heating rod for the sample, V
I_{oil}	electric current provided to the heating rod for the heat-treatment silicone oil, A
I_{samp}	electric current provided to the heating rod the sample, A
t	heating time, s
C_{oil}	heat capacity of the heat-treatment silicone oil, $J\ kg^{-1}\ K^{-1}$
m	mass of the oil used for calibration, kg
ΔT	temperature difference of the oil during heating, K
Greek letters	
η	CO_2 removal efficiency, %
μ	viscosity of solution, Pa·s

outlet below 20 ppb. No obvious IL loss was detected. For the absorbent of 30 wt% MEA + 50 wt% [bmim][BF_4] + 20 wt% H_2O, the thermal energy consumption for absorbent regeneration is 33.8% lower than that of the aqueous MEA solution.

1. Introduction

Global CO_2 emissions from fossil-fuel burning and industrial processes still keep increasing at present [1–3]. Amine-based chemical absorbents, which have been applied to remove acid gas from natural gas for several decades, are also available for CO_2 capture from power plant emissions. However, conventional CO_2 absorption by aqueous amine solutions is highly energy intensive. In addition, the volatility of amine and its degradation products from amine-based solutions are also concerning because of their potent carcinogenicity [4,5].

Recently, room-temperature ionic liquids (ILs) have been proposed as a potential candidate for CO_2 absorbent [6]. The application of ILs as CO_2 absorbents has given rise to a large collection of experimental and theoretical studies. ILs with imidazolium-based cations exhibited their affinity toward CO_2 and easy desorption. They have been measured and tested for CO_2 solubility to explore the phase behaviour of CO_2-IL pairs, as well as to develop methods to obtain higher solubility [7]. To overcome the limitations of ILs as CO_2 absorbents on CO_2 capture and to still take full advantage of their existing physical properties, mixed IL-amine solutions are employed to be co-capture agents [8,9]. However, MEA-carbamate, as the product of MEA reacting with CO_2, is too viscous to pump or precipitate from IL solvent. The separation of MEA-carbamate can also result in severe corrosion of the stripper during absorbent regeneration.

To solve the problem caused by MEA-carbamate in mixed IL-amine solution, absorbents of amine + IL + H_2O were proposed by Ahmady et al. [10]. They experimentally investigated the solubility and initial rate of CO_2 absorption in the mixture of aqueous 4 mol L^{-1} methyldiethanolamine (MDEA) solution with 1-butyl-3-methylimidazolium tetrafluoroborate ([bmim][BF_4]). Zhao et al. [11] measured densities, viscosities and CO_2 capture performances of 16 mixtures of amine, IL and H_2O. The experimental results suggested that an amine + IL + H_2O system might be an attractive CO_2 capture media with the advantages of low viscosity and a high CO_2 absorption rate.

The CO_2 capture performances of the MEA + [bmim][BF_4] + H_2O system in continuous CO_2 ab-desorption were investigated in our previous work [5]. The experimental results suggested that the mixed absorbent could reduce MEA loss to 1.16 kg per tonne of captured CO_2. The addition of 40 wt% [bmim][BF_4] to a 30% aqueous MEA solution decreased the thermal energy at the stripper from 8.18 to 5.14 GJ per tonne captured CO_2. However, it was also found that the value of 8.18 GJ per tonne captured CO_2 of 30% aqueous MEA solution is much higher than those of 4.0 and 3.9 GJ per tonne captured CO_2, as reported by Alie et al. [12] and Abu-Zahra et al. [13]. The main reason for these values could be attributed to the lean solvent CO_2 loading of the MEA solution which represents the degree of MEA regeneration that was not optimized in our previous case [5]. The lean solvent CO_2 loading prominently determines the thermal energy at the stripper. In the case of 4.0 GJ per tonne captured CO_2 by Alie et al., the lean solvent loading was between 0.25 and 0.30 mol CO_2 per mol MEA [12]. In the case of 3.9 GJ per tonne captured CO_2 by Abu-Zahra et al., the lean solvent loading was 0.24 mol CO_2 per mol MEA [13]. While in the case of 8.18 GJ per tonne captured CO_2 in our previous study, the lean solvent loading was lower than 0.10 mol CO_2 per mol of MEA. Therefore, to determine to what extent the mixed IL and amine solutions can decrease the thermal energy at the striper, it is essential to optimize the CO_2 ab-desorption loop system by varying the lean solvent CO_2 loading.

Meanwhile, it should be noted that for the calculation of thermal energy of the MEA + [bmim][BF_4] + H_2O solution at the stripper in our previous work, the absorbent was divided into two parts: aqueous MEA solution and pure [bmim][BF_4] in accordance with their individual concentrations. The total thermal energies at the stripper were obtained by adding together the aqueous MEA solution and the pure [bmim][BF_4]. Thus, the interaction between the aqueous MEA solution and the [bmim][BF_4] was ignored in this method. However, [bmim][BF_4] acts with water by means of replacing the C—H···F with C—H···O hydrogen bonds [14]. Hence, it is necessary to investigate the accuracy and uncertainty of the thermal energy at the stripper calculated by ignoring the interaction between the IL and the aqueous MEA solution.

On the other hand, besides CO_2 and N_2, the flue gas from a coal fired power plant contains SO_2, O_2, NO_x and fly ash. Among them, O_2 and SO_2 play important roles in the degradation of amine-based absorbents. Uyanga and Supap et al. [15,16] evaluated the contributions of SO_2 and O_2 to the degradation of MEA during