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Investigation of the absorption performance and viscosity for CO_2 capture process using [Bmim][Gly] promoted MDEA (N-methyldiethanolamine) aqueous solution

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The absorption performance of CO_2 in 1-butyl-3-methylimidazolium glycinate ([Bmim][Gly]) promoted MDEA (N-methyldiethanolamine) aqueous solution was investigated. The influences of temperature and mass fraction on CO_2 solubility were determined. The time dependence of CO_2 solubility demonstrated that the absorption rate of CO_2 significantly increased when MDEA aqueous solution was promoted by small amount of [Bmim][Gly]. The viscosities of both CO_2-unloaded and CO_2-loaded MDEA-[Bmim][Gly] aqueous solutions were measured at temperatures ranging from 303.2 K to 323.2 K and calculated by using the Weiland equation. The effects of temperature, mass fraction of MDEA and [Bmim][Gly], and CO_2 loading on viscosities were demonstrated on the basis of experiments and calculations. Our work showed that when the mass fraction of [Bmim][Gly] ranged from 0.10 to 0.15 and the total mass fraction of MDEA and [Bmim][Gly] was around 0.5, high absorption rate, large absorption capacity and appropriate viscosity can be simultaneously achieved.

1. Introduction

In recent decades, climate change and carbon dioxide (CO_2) emission have attracted increasing attentions worldwide and the reduction of CO_2 has become a hot issue [1]. The widely used technology for CO_2 capture is chemical absorption using alkanolamine aqueous solutions as absorbents [2–5]. However, the major disadvantage of traditional alkanolamine aqueous solutions is the high energy cost of regeneration. In particular, when using the popularly applied MEA (monoethanolamine)aqueous solution as absorbent, the mass fraction of MEA must be no greater than 30% in order to lower the corrosion, thus large quantities of superheated steam are needed in the regeneration tower to heat the 70% residual water. To lower the energy cost, many new absorbents have been developed in recent years [6,7], e.g., MEA promoted MDEA (N-methyldiethanolamine) aqueous solution. MDEA has the advantages of high resistance to thermal and chemical degradation, and low corrosivity [8]. However, the absorption of CO_2 in MDEA aqueous solution is quite slow. It has been well documented that adding activators such as MEA, DEA (diethanolamine) or PZ (piperazine) into MDEA aqueous solution can overcome this shortcoming, because the blending amines preserve high rate of the reaction of activators with CO_2 and low enthalpy of the reaction of MDEA with CO_2 [9,10].

Besides blending amines, FILs (functionalized ionic liquids) promoted MDEA aqueous solution is also considered to be a good absorbent for the removal of CO_2. FILs are organic salts composed of cations and anions that have unique characteristics, including wider liquid range, thermal stability, negligible vapor pressure, tunable physicochemical character and high CO_2 solubility [11–15]. The process of CO_2 uptake is reversible because CO_2 can be extruded from FILs upon heating (353 K–373 K) for several hours under vacuum. Bates et al. [16] reported a new "task-specific" ionic liquid named [NH_2p-bim][BF_4] for CO_2 capture. When exposed to a stream of bone dry CO_2 for 3 h at 1 atm and 295 K, about 7.4% CO_2 was captured by this ionic liquid. Jacauemin et al. [13] showed that CO_2 was highly soluble in 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF_4]) at pressures close to atmospheric. Xue et al. [17] synthesized a dual amine room temperature ionic liquid, [aemmim][Tau], and found that its absorption capacity reached about 0.9 mol CO_2 per mole ionic liquid at ambient pressure. Moreover, the mixtures of FILs and amines can preserve the desired property of FILs for CO_2 capture, e.g., Ahmaday et al. [18] showed

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Table 1
Sample description.

Chemical	CAS No.	Purity (in mass fraction %)	Molecular mass (Da)	Density (g cm^{-3})at 293.15 K
MDEA	105-59-9	99.5	119.16	1.0337
[Bmim][Gly]	1028361-04-7	99	213.28	1.26026

that under certain conditions, the presence of FILs increased the initial rate of CO_2 absorption in MDEA aqueous solution.

Among FILs, AAILs (amino acid ionic liquids) are considered to be promising activators to enhance the absorption rate of CO_2 in MDEA aqueous solution. By far, several AAILs have been proposed for CO_2 capture. For example, Zhang et al. [19] synthesized a new type of AAIL, terabutylphosphonium amino acid ([P(C_4)$_4$][AA]), and found that 0.5 mol CO_2 absorbed in 1 mol [P(C_4)$_4$][AA] at equilibrium. In presence of water, this AAIL could absorb equimolar amounts of CO_2. Guo et al. [20] compared the CO_2 absorption performance in 1-hexyl-3-methylimidazolium glycine ([Hmim][Gly]), MEA, DEA and AMP (2-amino-2-methyl-1-propanol) aqueous solutions. Their results showed that the absorption of CO_2 in these four absorbents were quite fast but the order of the absorption rate may be [Hmim][Gly]>MEA>DEA>AMP. Recently, trihexyl (tetradecyl) ammonium lysinate ([N_{66614}][Lys]) was examined as a high capacity absorbent for CO_2 capture. Experiments showed that 1 mol [N_{66614}][Lys] could absorb 2.1 mol CO_2 at ambient temperature and about 1 mol CO_2 at 353.15 K [21]. Zhang et al. [22] investigated the influences of temperature, solution composition and pressure on CO_2 absorption in aqueous solutions containing MDEA and AAILs including tetramethylammonium glycinate ([N_{1111}][Gly]), tetraethylammonium glycinate ([N_{2222}][Gly]), tetramethylammonium lysinate ([N_{1111}][Lys]) and tetraethylammonium lysinate ([N_{2222}][Lys]). They studied the effect of the concentration of [N_{1111}][Gly] on the absorption rate [23]. They also performed thermal regeneration of [N_{1111}][Gly] and MDEA aqueous solutions [24]. Zhou et al. [25] used [N_{1111}][Gly] promoted AMP aqueous solution to capture CO_2 and found that adding [N_{1111}][Gly] into AMP aqueous solution accelerated the absorption rate, e.g., the average absorption rates respectively enhanced 11.80%, 37.45%, 52.78% and 60.87% by adding 0.10, 0.20, 0.30 and 0.40 mol/L [N_{1111}][Gly], indicating that the higher was the concentration of AAIL, the higher was the absorption rate. However, due to the large viscosity of AAIL, the viscosity of AAIL-amine aqueous solution increased very significantly with increasing concentration of AAIL, thus significantly affected the mass transfer process and sometimes showed conflicting influence on the absorption rate [23]. Gao et al. [26] showed that the viscosity of AAIL-amine aqueous solution was closely related to the mass fractions of both AAIL and amine. For example, the viscosity was about 1.81 mPa s when the aqueous solution was composed of 10% [N_{1111}][Gly] and 40% MDEA, however, when the [N_{1111}][Gly] concentration increased to 15%, the viscosity very rapidly increased to 5.39 mPa s. The solution viscosity is important in mass transfer rate modeling of absorbers and regenerators because these properties significantly affect the liquid film coefficient for mass transfer [27–29]. The viscosities of both CO_2-unloaded and CO_2-loaded aqueous solutions are required when designing or simulating an absorption column for CO_2 absorption. It seems when using AAIL promoted amine aqueous solutions as absorbents for the removal of CO_2, both absorption performance and viscosities should be well considered.

Besides [N_{1111}][Gly], [N_{2222}][Gly], [N_{1111}][Lys] and [N_{2222}][Lys], 1-butyl-3-methylimidazolium glycinate ([Bmim][Gly]) is also a good absorbent for the removal of CO_2. However, the absorption performance of CO_2 in [Bmim][Gly] promoted MDEA aqueous solutions has not been documented. Moreover, although some properties of [Bmim][Gly] promoted alkylolamine aqueous solutions are available [30–32], the studies concerning the viscosities of CO_2-unloaded and CO_2-loaded MDEA-[Bmim][Gly] aqueous solutions are rare so far.

The main purposes of this work are to (1) experimentally determine the absorption performance of CO_2 in [Bmim][Gly] promoted MDEA aqueous solutions and illustrate the effect of the addition of [Bmim][Gly]; (2) experimentally determine the viscosity of carbonated MDEA-[Bmim][Gly] aqueous solution and then calculate it with the Weiland equation [33], so as to demonstrate the effects of temperature, CO_2 loading, mass fractions of [Bmim][Gly] and MDEA on viscosity; (3) determine an appropriate composition of [Bmim][Gly] and MDEA under which satisfactory absorption performance and viscosity can be simultaneously achieved.

2. Experimental

2.1. Materials

MDEA and [Bmim][Gly] were used without further purification. The sample description is shown in Table 1. Aqueous solutions of MDEA-[Bmim][Gly] were prepared by adding doubly distilled water. The uncertainly of the electronic balance is ±0.1 mg.

2.2. Apparatus and procedure

The absorption performance of aqueous solutions was measured by the equipment composed of one high-pressure CO_2 tank, one MFC (mass flow controller), one MFM (mass flow meter), one absorption bottle, one constant temperature water bath, one desiccator and one CO_2 analyzer (Advanced Gasmitter by Germany Sensors Europe GmbH, the accuracy is ±2%). The schematic diagram of the experiment is shown in Fig. 1. The flask was immersed into a thermostatic bath and the temperature of the solution can be regulated within 0.1 K. During the experiment, CO_2 from the high-pressure tank was inlet into the MFC to maintain a constant flow rate and then into the absorption bottle and absorbed by the solution. The residual and unabsorbed gas firstly flowed into the desiccator and then into the CO_2 analyzer. The gas concentration

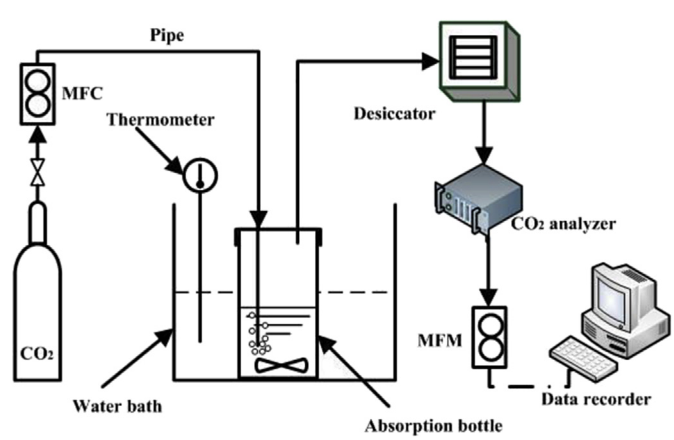

Fig. 1. The schematic diagram of the experiment.