



Experiment and model for the surface tension of amine–ionic liquids aqueous solutions



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ABSTRACT

The surface tension (γ) of 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF₄])–monoethanolamine (MEA), 1-butyl-3-methylimidazolium bromide ([Bmim][Br])–MEA, [Bmim][BF₄]-diethanolamine (DEA) and [Bmim][Br]-DEA aqueous solutions was measured by using the BZY-1 surface tension meter. The temperature ranged from (293.2 to 323.2) K. The mass fraction of amines and ionic liquids (ILS) respectively ranged from 0.15 to 0.30 and 0.05 to 0.10. A thermodynamic equation was proposed to model the surface tension of amines–ILS aqueous solutions and the calculated results agreed well with the experiments. The effects of temperature, mass fraction of amines and ILS on the surface tension were demonstrated on the basis of experiments and calculations.

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1. Introduction

In recent decades, atmospheric levels of CO₂ have increased rapidly due to the utilization of large quantities of fossil fuels. Development of affordable yet technically feasible separation technologies for reducing CO₂ emissions has attracted global attention. Among the available separation technologies including absorption [1–10], adsorption [11,12], membrane [13–15] and hydration [16–18], chemical absorption using aqueous solution of alkanolamines as an absorbent has been widely used for the removal of CO₂ from a variety of gas streams. However, the major disadvantage of traditional aqueous solutions of alkanolamines is the high energy cost for regeneration. In particular, when using the monoethanolamine (MEA) aqueous solution as an absorbent, the mass fraction of MEA must be no greater than 30% 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, new absorbents using amine blends have been developed in recent years. For example, the blends of MEA and *N*-methyldiethanolamine (MDEA) preserve the high rate of the reaction of MEA with CO₂. The low enthalpy of the reaction of MDEA with CO₂, hence leads to high absorption rates in the absorber column, yet a lower heat of regeneration in the stripper section. Moreover, as the corrosivity of MDEA is low, the total mass fraction of amines in the MEA–MDEA aqueous solution can be significantly

increased. Besides the amine blends, adding a physical solvent to the aqueous solutions of alkanolamines is also considered to be an effective method to lower the regeneration energy cost [19,20]. For example, Archane *et al.* [19] showed that when using poly(ethylene oxide)400 (PEG400)-diethanolamine (DEA) aqueous solution as the absorbent, a lower energy cost can be achieved in regeneration because at a given CO₂ loading and constant concentration of DEA, the CO₂ molecular concentration increases with the increase of PEG400 concentration, whereas the ion repartition is not significantly influenced by the solvent composition.

Recently, interest has increased rapidly in CO₂ capture using the ionic liquids (ILS) as the absorbents. The ILS have unique characteristics including a wide liquid range, thermal stability, negligible vapor pressure, tuneable physicochemical character and high CO₂ solubility [21–26]. The mixtures of ILS and amines preserve the desired property of ILS for CO₂ capture, but without many of their inherent drawbacks such as high viscosity because of their corresponding CO₂ adducts. For example, Jacquemin *et al.* [23] showed that CO₂ was highly soluble in 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF₄]) at pressures close to atmospheric. Ahmady *et al.* [24,25] showed that under certain conditions, the presence of ILS increased the initial rate of CO₂ absorption in MDEA aqueous solution. Bidart *et al.* [26] showed that although 1-butyl-3-methylimidazolium bromide ([Bmim][Br]) did not present a greater absorption capacity than the amine solutions, the mixture of [Bmim][Br] and MEA solution was explored as an option to enhance CO₂ absorption due to the advantages of low volatility,

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TABLE 1
Provenance and mass fraction purity of the materials studied.

Chemical	Supplier	CAS No.	Mass fraction purity	Molecular mass/(Dalton)
[Bmim]BF ₄	Shanghai Chengjie Chem. Co., Ltd [China]	174501-65-6	0.990	226.02
[Bmim]Br		85100-77-2	0.990	219.20
MEA	Tianjin Kermel Chem. Reagent Co., Ltd [China]	141-43-5	0.990	61.09
DEA		111-42-2	0.997	105.14

TABLE 2
Surface tension γ^a /(mN · m⁻¹) of MEA–[Bmim][BF₄] aqueous solutions under different mass fractions of [Bmim][BF₄]($w_{[Bmim][BF_4]}$) and MEA(w_{MEA}). The pressure (p) is 100 kPa.

$w_{MEA}/w_{[Bmim][BF_4]}$	γ^a /(mN · m ⁻¹)			
	$T = 293.2$ K	$T = 303.2$ K	$T = 313.2$ K	$T = 323.2$ K
0.15/0.05	53.2	52.5	51.8	51.2
0.15/0.10	49.4	49.2	49.0	48.8
0.20/0.05	53.5	52.8	52.4	51.8
0.20/0.10	49.8	49.5	49.4	49.2
0.25/0.05	54.3	53.7	53.3	52.9
0.25/0.10	50.2	50.1	50.0	49.8
0.30/0.05	54.5	53.9	53.6	53.1
0.30/0.10	50.6	50.5	50.4	50.3

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w) = \pm 0.0001$; $u(\gamma) = 0.1$ mN · m⁻¹; $u(p) = 5$ kPa.

TABLE 3
Surface tension γ^a /(mN · m⁻¹) of MEA–[Bmim]Br aqueous solutions under different mass fractions of [Bmim]Br($w_{[Bmim]Br}$) and MEA(w_{MEA}). The pressure (p) is 100 kPa.

$w_{MEA}/w_{[Bmim]Br}$	γ^a /(mN · m ⁻¹)			
	$T = 293.2$ K	$T = 303.2$ K	$T = 313.2$ K	$T = 323.2$ K
0.15/0.05	64.6	62.8	61.3	59.5
0.15/0.10	62.5	60.4	58.9	56.8
0.20/0.05	63.9	62.0	59.9	58.3
0.20/0.10	62.1	59.9	58.2	56.0
0.25/0.05	63.0	61.1	59.4	57.6
0.25/0.10	61.3	59.6	57.5	55.5
0.30/0.05	62.5	60.8	59.2	57.3
0.30/0.10	60.8	59.1	56.9	55.0

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w) = \pm 0.0001$; $u(\gamma) = 0.2$ mN · m⁻¹; $u(p) = 5$ kPa.

TABLE 4
Surface tensions γ^a /(mN · m⁻¹) of DEA–[Bmim][BF₄] aqueous solutions under different mass fractions of [Bmim][BF₄]($w_{[Bmim][BF_4]}$) and DEA(w_{DEA}). The pressure (p) is 100 kPa.

$w_{DEA}/w_{[Bmim][BF_4]}$	γ^a /(mN · m ⁻¹)			
	$T = 293.2$ K	$T = 303.2$ K	$T = 313.2$ K	$T = 323.2$ K
0.15/0.05	52.5	51.6	51.0	50.1
0.15/0.10	47.1	46.7	46.4	46.0
0.20/0.05	52.8	51.9	51.2	50.4
0.20/0.10	47.8	47.3	47.0	46.7
0.25/0.05	52.9	52.2	51.4	50.7
0.25/0.10	48.1	47.8	47.5	47.3
0.30/0.05	53.3	52.6	51.9	51.2
0.30/0.10	48.2	47.9	47.7	47.5

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w) = \pm 0.0001$; $u(\gamma) = 0.1$ mN · m⁻¹; $u(p) = 5$ kPa.

chemical stability and stability of the complex formed when CO₂ saturation is achieved.

A knowledge of surface tension is required when designing or simulating an absorption column for CO₂ capture. It can significantly affect the absorption efficiency because both the penetration

TABLE 5
Surface tension γ^a /(mN · m⁻¹) of DEA–[Bmim]Br aqueous solutions under different mass fractions of [Bmim]Br($w_{[Bmim]Br}$) and DEA(w_{DEA}). The pressure (p) is 100 kPa.

$w_{DEA}/w_{[Bmim]Br}$	γ^a /(mN · m ⁻¹)			
	$T = 293.2$ K	$T = 303.2$ K	$T = 313.2$ K	$T = 323.2$ K
0.15/0.05	64.4	62.4	60.3	58.4
0.15/0.10	62.1	59.9	57.5	55.1
0.20/0.05	63.4	61.5	59.6	57.6
0.20/0.10	61.4	59.2	57.0	54.8
0.25/0.05	62.1	60.5	58.7	57.3
0.25/0.10	60.3	58.2	56.5	54.3
0.30/0.05	61.0	59.1	57.8	56.5
0.30/0.10	59.5	57.8	56.0	53.8

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w) = \pm 0.0001$; $u(\gamma) = 0.1$ mN · m⁻¹; $u(p) = 5$ kPa.

of CO₂ molecules from gas phase to the liquid phase and the enhancement of the absorption closely relate to the surface tension. In recent years, there are some experimental and theoretical works concerning the surface tension of aqueous solutions containing amines [27–35] and ILS [36–41]. However, experimental and theoretical works concerning the surface tension of ILS–MEA and ILS–DEA aqueous solutions are relatively rare.

The main purpose of this work is to investigate the surface tension of MEA–[Bmim][BF₄], MEA–[Bmim][Br], DEA–[Bmim][BF₄] and DEA–[Bmim][Br] aqueous solutions experimentally and theoretically, and then demonstrate the effects of temperature, mass fractions of amines and ILS on the surface tension. To this end, the surface tension was measured at the temperatures from (293.2 to 323.2) K. The mass fraction of amines and ILS respectively ranged from 0.15 to 0.30 and 0.05 to 0.10. Besides experimental work, a thermodynamic equation was also proposed in this work to model the surface tension.

2. Experimental

2.1. Materials

The amines and ILS were purchased from HuaXin chemical Co. The sample description is shown in table 1. They were used without further purification. Aqueous solutions of MEA–[Bmim][BF₄], MEA–[Bmim][Br], DEA–[Bmim][BF₄] and DEA–[Bmim][Br] were prepared by adding doubly distilled water. The uncertainty of the electronic balance is ± 0.1 mg.

2.2. Apparatus and procedure

The surface tension was measured by using the BZY-1 surface tension meter produced by Shanghai Hengping Instrument Factory. The BZY-1 meter employs the Wilhemy plate principle, *i.e.*, the maximum tensile force competing with the surface tension is measured when the bottom edge is parallel to the interface and just touches it. The measurement ranges for temperature and surface tension are respectively (268.15 to 383.15) K and (0.1–400.0) mN · m⁻¹. The uncertainty is ± 0.1 mN · m⁻¹. The size-volume of the different

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