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Investigation of surface tension and viscosity for aqueous solutions of MEA-MeOH and DEA-MeOH

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ABSTRACT

The surface tension (γ) and viscosity (η) of monoethanolamine (MEA)-methanol (MeOH) and diethanolamine (DEA)-MeOH aqueous solutions were measured by using the BZY-1 surface tension meter and the NDJ-5S digital rotational viscometer. The temperature ranged from 303.2 K to 323.2 K. The mass fractions of MeOH, MEA and DEA ranged from 0.025 to 0.075, 0.2 to 0.4 and 0.2 to 0.4 respectively. Equations were proposed to model the surface tension and viscosity respectively and both the calculated results agreed well with the experiments. The effects of temperature and mass fraction of MeOH/amines on the surface tension and viscosity were demonstrated on the basis of experiments and calculations.

1. Introduction

The greenhouse effect and environmental problems caused by the emission of carbon dioxide (CO_2) from industrial processes and coal-fired boilers seriously impacted the sustainable development of the economy. Development of affordable yet technically feasible separation technologies for CO_2 capture has become a global issue [1–3]. Among the available separation technologies including absorption [4,5], adsorption [6], membrane [7] and hydration [8], chemical absorption is one of the most effective approaches because CO_2 can be satisfactorily removed and the absorbents can be well regenerated by heating [9]. Currently, chemical absorption technologies using aqueous solutions of alkanolamines such as primary amine - monoethanolamine (MEA), secondary amine - diethanolamine (DEA) and tertiary amine - N-methyldiethanolamine (MDEA) are extensively used for the CO_2 capture from various gas streams. However, the major disadvantage of primary and secondary amines aqueous solution is the high energy consumption in regeneration process [10,11]. Tertiary amines have high absorption capacity and low energy cost for regeneration, but the absorption rate is relatively low. Since these traditional amine solvents have their own weaknesses, the development of some new potential solvents is very important to improve the CO_2 capture performance and reduce the energy cost.

Nowadays, there are a variety of studies concerning the blended amines such as MEA–MDEA, MEA–2-diethylaminoethanol (DEAE) and piperazine (PZ)–DEAE [12–14] and novel amines such as 2-(1-piperazinyl)-ethylamine (PZEA), diethylenetriamine (DETA), 4-diethylamino-2-butanol (DEAB) [15–17]. In general, the blended amines combine the benefits of the high reaction rate of the primary or secondary amines and the high equilibrium capacity and low energy consumption in regeneration of the tertiary or sterically hindered amines [18,19]. Meanwhile, newly developed amines can also be considered as promising alternative solvents for CO_2 capture due to their excellent performance such as higher absorption rate and lower heat duty than that of traditional amines [17,20]. However, the regeneration of these amines-based solvents is still executed at elevated temperatures. These solvents significantly improve the absorption and regeneration performance of CO_2 capture, however, the energy cost in regeneration is still high.

Besides the amine blends and novel amines, adding physical solvents to the aqueous solutions of alkanolamines is also considered to be an effective method to lower the regeneration energy requirement [21,22]. Several hybrid solvents have been proposed for CO_2 capture, *e.g.*, ethylenediamine (EDA)–methanol/ethanol, triethanolamine (TEA)–methanol/ethanol/n-propanol/n-butanol/ethylene glycol, PZ–diethylene glycol (DEG) and MEA–triethylene glycol (TEG) [23–26]. Methanol (MeOH) is amongst the most widely used physical solvents for CO_2 capture from natural gas and synthesis gas due to its high physical solubility of CO_2 and low heat consumption in the solvent regeneration step. Fu et al. [27] experimentally investigated the absorption performance of

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CO_2 absorption into the hybrid solvent MEA–MeOH and they found the overall gas phase mass transfer coefficient of CO_2 absorption into the MEA–MeOH is higher than that into the MEA–H_2O. Phairat et al. [28] demonstrated the reason why the MeOH mixed into MEA solution can enhance the CO_2 absorption. They suggested that the hybrid solvent increases the CO_2 absorption rate because it enhances the diffusivity and solubility characteristics of the solvent. Hamborg et al. [29] evaluated the effect of aqueous MeOH on desorption process of MDEA and their results indicatively showed a maximum (7.5%) decrease in the reboiler duty of the desorber at MeOH fractions of about 0.06 compared to purely aqueous solutions, and the reboiler temperature decreased with increasing MeOH fractions. Henni et al. [30] measured the solubility of CO_2 in an aqueous mixed solvent consisting of MeOH (40 mass%), MDEA (40 mass%), and water (20 mass%) at 40 and 100 °C and partial pressures of the acid gas up to 7.04 MPa. Tounsi et al. [31] studied the CO_2 solubility in water-DEA-MeOH solutions with temperature ranging from 323.15 to 393.15 K and pressure up to 3.6 MPa.

Besides mass transfer and heat transfer of MeOH–amines, knowledge of surface tension and viscosity is also required when designing or simulating an absorption column for CO_2 capture. The surface tension can significantly affect the absorption efficiency because both the penetration of CO_2 molecules from gas phase to the liquid phase and the enhancement of the absorption closely relate to the surface tension. Meanwhile, solution viscosity is important to the mass transfer rate modeling of absorbers and regenerators because the properties significantly affect the liquid film coefficient for mass transfer [32–36]. Recently, there are many experimental and theoretical works concerning the surface tension and viscosity of aqueous solutions containing amines and physical solvents [37–45]. However, experimental and theoretical studies concerning the surface tension and viscosity of aqueous solutions of physical solvent–alkanolamines (*e.g.*, DEA-MeOH and MEA-MeOH) are rare, and the effects of temperature and mass fractions of amine and MeOH on surface tension and viscosity have not been well documented so far.

The main purpose of this work is to investigate the surface tension and viscosity of MEA-MeOH and DEA-MeOH aqueous solutions experimentally and theoretically, so as to demonstrate the effects of temperature, mass fractions of MeOH, MEA and DEA on the surface tension and viscosity. To this end, the surface tension and viscosity were measured at the temperatures ranging from 303.2 K to 323.2 K. The mass fraction of MeOH, MEA, and DEA respectively ranged from 0.025 to 0.075, 0.2 to 0.4 and 0.2 to 0.4. Besides experimental work, thermodynamic equations were used to model the surface tension and viscosity.

2. Experimental

2.1. Materials

The samples used in this work are detailed in Table 1. Purities are as stated by the supplier, and no further purification was carried out. An analytical balance (Jingtian FA1604A) with an accuracy of 0.1 mg was used to weigh all required chemicals. The water contents (in mass percent) of MeOH, MEA and DEA are respectively 0.1%, 0.06% and 0.05% (determined by using the Karl Fischer method, as stated by the supplier) and they were accounted for upon solution preparation. Aqueous solutions of MEA-MeOH and DEA-MeOH were prepared by adding deionized water (Electrical resistivity >15 MΩ·cm at 298 K) obtained from the Heal Force ROE (Reverse Osmosis Electrodeionization)-100 apparatus to the weighed quantities of amines. Taking the purities and water content into account, the uncertainties of the mass fractions of MeOH, MEA and DEA are respectively $u(w_{MeOH}) = \pm 0.001$, $u(w_{MEA}) = \pm 0.002$ and $u(w_{DEA}) = \pm 0.002$.

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 the liquid. The measurement ranges for temperature and surface tension are respectively (268.15–383.15) K and (0.1–400.0) $mN \cdot m^{-1}$. The uncertainty is ±0.1 $mN \cdot m^{-1}$. The size-volume of the different samples used in the BZY-1 m is 20 mL. During the experiments, the copper pan in the host of the BZY-1 m is connected with the thermostatic bath (CH-1006, uncertainty is ±0.1 K). Via the circulation of the water, the temperature of the water in the copper pan is kept the same as that in the thermostatic bath. The aqueous solution is put into the solution container immersed in the copper pan and its temperature can be measured by a thermocouple. The scale reading of the thermocouple has been well calibrated by a mercury thermometer.

The viscosities of MEA-MeOH and DEA-MeOH aqueous solutions were measured by using the NDJ-5S digital rotational viscometer produced by the Shanghai Changji Geological Instrument company. The measurement ranges and measurement error are respectively $(0.1–10^5)$ mPa·s and ±1% (for a Newtonian fluid). The instrument was designed and manufactured with 5 rotors (0#, 1#, 2#, 3# and 4#) and 4 different velocities (6, 12, 30 and 60) rpm. An automatic switch was set for free selection of proper rotating speed or rotor number, which enables the instrument to measure any viscosity value in the given range. In our experiments, five combinations of rotor and rotating speed were selected. The combinations (0#-60, 0#-30, 1#-60, 1#-30 and 1#-12) rpm, respectively, correspond to the upper limits of (10, 20, 100, 200 and 500) mPa·s. The uncertainty of temperature is ±0.1 K.

To verify the reliability of the equipments, surface tension and viscosity of pure water, pure MeOH and amine aqueous solution (the mass fraction of amine, $w = 0.2, 0.3, 0.4, 1.0$) were measured at 303.2 K, 313.2 K and 323.2 K. The measured surface tensions and viscosities were compared with those from different literatures [42,45–64], as shown in Table 2. The average relative deviations (ARD) of surface tensions and of viscosities were also shown in Table 2, indicating that the experimental equipment in this work

Table 1
Sample description.

Chemical name	CAS	Purity (mole fraction, as stated by the supplier)	Water content (mass percent, as stated by the supplier)	Source
MeOH	67-56-1	$x \geq 0.995$	0.1%	Kermel Chemical Reagent
MEA	141-43-5	$x \geq 0.995$	0.06%	Aladdin Reagent
DEA	111-42-2	$x \geq 0.995$	0.05%	Aladdin Reagent
water	7732-18-5	Electrical resistivity > 15 MΩ cm at $T = 298$ K		Heal force ROE-100 apparatus