



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.

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1. Introduction

The greenhouse effect and environmental problems caused by the emission of carbon dioxide (CO₂) from industrial processes and coal-fired boilers seriously impacted the sustainable development of the economy. Development of affordable yet technically feasible separation technologies for CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ capture from natural gas and synthesis gas due to its high physical solubility of CO₂ and low heat consumption in the solvent regeneration step. Fu et al. [27] experimentally investigated the absorption performance of

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CO₂ absorption into the hybrid solvent MEA–MeOH and they found the overall gas phase mass transfer coefficient of CO₂ absorption into the MEA–MeOH is higher than that into the MEA–H₂O. Phairat et al. [28] demonstrated the reason why the MeOH mixed into MEA solution can enhance the CO₂ absorption. They suggested that the hybrid solvent increases the CO₂ 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 CO₂ 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 CO₂ 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 CO₂ capture. The surface tension can significantly affect the absorption efficiency because both the penetration of CO₂ 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 $u(w_{\text{MeOH}}) = \pm 0.001$, $u(w_{\text{MEA}}) = \pm 0.002$ and $u(w_{\text{DEA}}) = \pm 0.002$.

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·m⁻¹. The uncertainty is ± 0.1 mN·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 (0.1–10⁵) mPa·s and $\pm 1\%$ (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

Table 2

Surface tension (γ) and viscosity (η) of pure water, pure MeOH and amine aqueous solution (the mass fraction of amine $w = 0.2, 0.3, 0.4, 1.0$) and the average relative deviation (ARD) between the experiments from this work and those from literatures [42,45–64]. Pressure (p) = 101 kPa.^a

Sample	w	Surface tension/(mN·m ⁻¹)			Viscosity/(mPa·s)		
		303.2 K	313.2 K	323.2 K	303.2 K	313.2 K	323.2 K
Water		71.1/71.21[42,46]	69.4/69.52[42,46]	67.7/67.92[42,46]	0.80/0.7975[47]/ 0.8007[48]	0.65/0.6531[47]/0.6559 [48]	0.55/0.5494[48]
		ARD = 0.2%			ARD = 0.4%		
MeOH		21.7/22.01[42]/21.75 [49]	20.9/21.13[42]/20.90 [49]	20.0/20.21[42]/20.04 [49]	0.51/0.508[45]/ 0.514[50]	0.44/0.436[51]/0.45[52]	0.39/0.383[51]/ 0.396[52]
		ARD = 0.7%			ARD = 1.3%		
MEA	0.200	62.1/61.84[46]/61.8 [53]/64.7[54]/66.7[55]	60.4/60.17[46]/60.2 [53]/63.3[54]/65.2[55]	58.9/58.89[46]/58.7 [53]/61.7[54]/63.6[55]	1.51/1.49[47]/1.53 [56]	1.18/1.165[47]/1.18 [56]/1.18[57]	0.97/0.98[56]/0.95 [57]
	0.300	59.7/59.61[46]/59.5 [53]/63.6[54]/63.7[55]	58.4/57.94[46]/58[53]/ 62.6[54]/62.4[55]	56.4/56.36[46]/56.5 [53]/61.2[54]/61[55]	2.11/2.099[47]/ 2.13[56]/2.1[58]	1.63/1.596[47]/1.59 [56]/1.67[57]/1.6[58]	1.31/1.31[56]/1.33 [57]/1.29[58]
	0.400	57.8/57.94[46]/61.7 [54]/61.5[55]	56.3/56.27[46]/60.3 [54]/59.8[55]	54.6/54.67[46]/58.8 [54]/58.4[55]	3.12/3.086[47]	2.29/2.276[47]/2.28[57]	1.79/1.75[57]
	1.000	47.7/48.14[46]/48.1 [54]/47.64[55]	45.9/46.43[46]/46.7 [54]/46.88[55]	44.2/44.81[46]/45.6[54] [61]/45.4[62]	14.47/14.05[47]/ 15.11[58]	9.87/9.95[47]/9.61[57]/ 10.02[58]	6.79/6.72[57]/ 6.972[58]
		ARD = 3.3%			ARD = 1.5%		
DEA	0.200	63.3/64.4[53]/62.46[59]	62.1/63.9[53]/60.74 [59]/63.94[60]	59.9/61.8[53]/59.1[59]/ 62.62[60]	1.64/1.67[56]/ 1.646[63]	1.29/1.32[56]/1.28[63]/ 1.25[64]	1.03/1.05[56]/ 1.034[63]
	0.300	60.5/61.3[53]/59.95[59]	58.8/60.4[53]/58.25 [59]/60.17[60]	57.4/58.2[53]/56.6[59]/ 58.57[60]	2.57/2.57[56]/2.57 [58]/2.542[63]	1.98/1.99[56]/1.97[58]/ 1.93[63]/1.884[64]	1.42/1.41[56]/1.41 [58]/1.469[63]
	0.400	58.3/58.03[59]	56.6/56.32[59]	54.7/54.68[59]	4.27	3.01/3.008[64]	2.27
	1.000	46.0/46.46[59]/46.4 [61]/46.5[62]	44.1/44.75[59]/45[61]/ 46[62]	42.8/43.12[59]/43.4 [61]/45.4[62]	381.21/379.3[48]/ 383.9[58]	188.64/186.4[48]/188.2 [58]/188.2[64]	105.98/98.85[48]/ 119.5[58]
	ARD = 1.9%			ARD = 1.9%			

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(p) = 2$ kPa; $u(w_{\text{MEA}}) = \pm 0.002$; $u(w_{\text{DEA}}) = \pm 0.002$; $u(w_{\text{MeOH}}) = \pm 0.001$; $u(\gamma) = 0.3$ mN·m⁻¹; $u(\eta) = 0.06$ mPa·s ($\eta \leq 10$ mPa·s), $u(\eta) = 0.22$ mPa·s ($10 < \eta \leq 20$ mPa·s), $u(\eta) = 2.58$ mPa·s ($100 < \eta \leq 200$ mPa·s), $u(\eta) = 8.56$ mPa·s ($200 < \eta \leq 500$ mPa·s). Relative uncertainties u_r are $u_r(\eta) = 5.1\%$ ($\eta \leq 10$ mPa·s), $u_r(\eta) = 1.5\%$ ($10 < \eta \leq 20$ mPa·s), $u_r(\eta) = 1.9\%$ ($100 < \eta \leq 200$ mPa·s), $u_r(\eta) = 2.3\%$ ($200 < \eta \leq 500$ mPa·s).

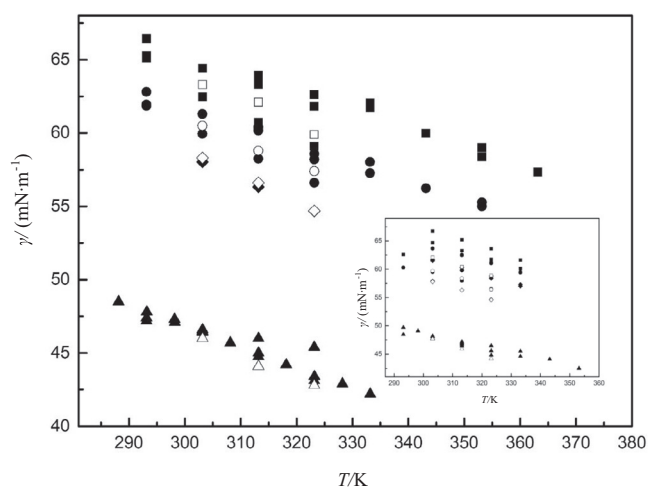


Fig. 1. Comparison of the surface tension of DEA and MEA (inset plot) aqueous solutions from different work. Solid symbols: from literatures [46,53–55,59–62,65–67]; hollow symbols: from this work. Main plot: \blacksquare $w_{\text{DEA}} = 0.2$ [53,59,60,66]; \bullet $w_{\text{DEA}} = 0.3$ [53,59,60,66]; \blacklozenge $w_{\text{DEA}} = 0.4$ [59]; \blacktriangle $w_{\text{DEA}} = 1.0$ [59,61,62,65]; \square $w_{\text{DEA}} = 0.2$; \circ $w_{\text{DEA}} = 0.3$; \diamond $w_{\text{DEA}} = 0.4$; \triangle $w_{\text{DEA}} = 1.0$. Inset plot: \blacksquare $w_{\text{MEA}} = 0.2$ [46,53–55]; \bullet $w_{\text{MEA}} = 0.3$ [46,53–55]; \blacklozenge $w_{\text{MEA}} = 0.4$ [46,54,55]; \blacktriangle $w_{\text{MEA}} = 1.0$ [46,54,55,65,67]; \square $w_{\text{MEA}} = 0.2$; \circ $w_{\text{MEA}} = 0.3$; \diamond $w_{\text{MEA}} = 0.4$; \triangle $w_{\text{MEA}} = 1.0$.

are reliable. Moreover, our data are graphically compared with literature data [46,47,53–69], as shown in Figs. 1 and 2. From Fig. 1, one finds that there is significant difference in the surface tension data from different work. Such deviation may be caused by the purity and the water content of the used chemical, e.g., the largest deviation between the surface tensions presented in our previous work [53] and the current manuscript reaches nearly 2 mN·m⁻¹. In our previous work [53], DEA (0.982 in mole fraction) was used, while in the current work, DEA (0.995 in mole fraction) was used and the water content was accounted for upon solution

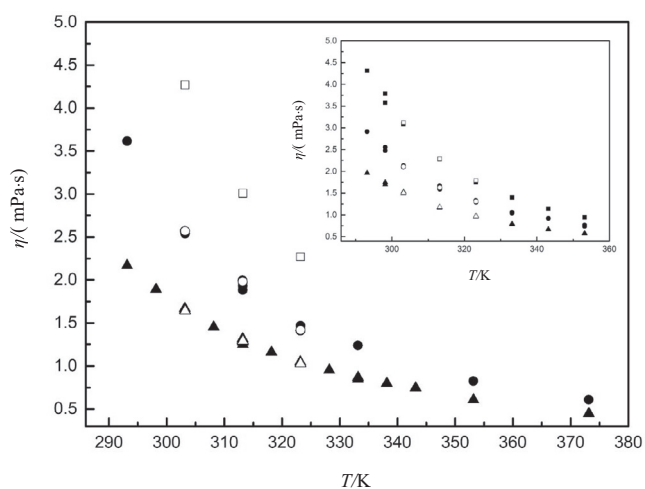


Fig. 2. Comparison of the viscosity of DEA and MEA (inset plot) aqueous solutions from different work. Solid symbols: from literatures [47,56–58,63,64,66,68,69]; hollow symbols: from this work. Main plot: \blacktriangle $w_{\text{DEA}} = 0.2$ [56,63,64,66,68]; \bullet $w_{\text{DEA}} = 0.3$ [56,58,63,64,66]; \blacklozenge $w_{\text{DEA}} = 0.4$ [64]; \triangle $w_{\text{DEA}} = 0.2$; \circ $w_{\text{DEA}} = 0.3$; \square $w_{\text{DEA}} = 0.4$. Inset plot: \blacktriangle $w_{\text{MEA}} = 0.2$ [47,56,57,69]; \bullet $w_{\text{MEA}} = 0.3$ [47,56–58,69]; \blacklozenge $w_{\text{MEA}} = 0.4$ [47,57,69]; \triangle $w_{\text{MEA}} = 0.2$; \circ $w_{\text{MEA}} = 0.3$; \square $w_{\text{MEA}} = 0.4$.

preparation, thus the data presented in the current work are more accurate.

3. Results and discussion

3.1. Surface tension and the model

Surface tension values of MEA–MeOH and DEA–MeOH aqueous solutions at different temperatures and different mass fraction of MeOH/amines are shown in Tables 3 and 4.

Table 3
Surface tensions (γ) of MEA–MeOH aqueous solutions under different mass fractions of MEA (w_{MEA}) and MeOH (w_{MeOH}) and temperatures. Pressure (p) = 101 kPa.^a

w_{MEA}	w_{MeOH}	$\gamma/(\text{mN}\cdot\text{m}^{-1})$		
		303.2 K	313.2 K	323.2 K
0.200	0.025	60.1	58.5	56.4
	0.050	58.3	56.1	54
	0.075	56.3	54.2	52.3
0.300	0.025	57.5	56.2	55.1
	0.050	55.3	54.3	53.5
	0.075	53.3	52.1	51.6
0.400	0.025	56.5	55.3	53.1
	0.050	54.6	53.3	51.3
	0.075	52.4	51.2	49.9

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w_{\text{MEA}}) = \pm 0.002$; $u(w_{\text{MeOH}}) = \pm 0.001$; $u(p) = 2$ kPa; $u(\gamma) = 0.3$ mN·m⁻¹

Table 4
Surface tensions (γ) of DEA–MeOH aqueous solutions under different mass fractions of DEA (w_{DEA}) and MeOH (w_{MeOH}) and temperatures. Pressure (p) = 101 kPa.^a

w_{DEA}	w_{MeOH}	$\gamma/(\text{mN}\cdot\text{m}^{-1})$		
		303.2 K	313.2 K	323.2 K
0.200	0.025	60.2	58.5	56.2
	0.050	57.6	56.1	54.1
	0.075	55.7	53.8	52.2
0.300	0.025	57.6	56.1	54
	0.050	55.8	54.2	52.3
	0.075	53.9	52.3	51.2
0.400	0.025	55.3	53.8	51.5
	0.050	53.7	51.6	49.1
	0.075	51.7	49.6	47.7

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w_{\text{DEA}}) = \pm 0.002$; $u(w_{\text{MeOH}}) = \pm 0.001$; $u(p) = 2$ kPa; $u(\gamma) = 0.3$ mN·m⁻¹

Besides experimental measurements, models that can correctly calculate the surface tensions of concerned systems are also important. In this work, the surface tensions of MEA–MeOH and DEA–MeOH aqueous solutions are formulated as follows:

$$\gamma^{\text{aq}} = \gamma^0 + \gamma' \quad (1)$$

in which γ^0 and γ' are expressed as:

$$\gamma^0 = x_1\gamma_1 + x_2\gamma_2 + x_3\gamma_3 \quad (2)$$

$$\gamma' = x_1x_2G_{12} + x_1x_3G_{13} + x_2x_3G_{23} \quad (3)$$

where the subscripts 1, 2 and 3 stand for MEA/DEA, MeOH and water, respectively; x_i is the mole fraction of component i in the aqueous solution, γ_i is the surface tension of pure component i , which can be expressed as a function of the temperature by fitting to the experimental data. G_{ij} is expressed as a function of temperature and mass fraction:

$$G_{13} = (a_{13} + b_{13}w_1 + c_{13}w_1^2)T \quad (4)$$

$$G_{23} = (a_{23} + b_{23}w_2 + c_{23}w_2^2)T \quad (5)$$

$$G_{12} = (a_{12} + b_{12}[(w_1 + w_2)/2] + c_{12}[(w_1 + w_2)/2]^2)T \quad (6)$$

The model parameters a_{ij} , b_{ij} and c_{ij} can be obtained by fitting to the experimental data. The objective function (ARD) is defined as:

$$\text{ARD} = \sum_{i=1}^n (1 - \gamma^{\text{cal}}/\gamma^{\text{exp}}) \times 100\%/n \quad (7)$$

where the superscripts 'exp' and 'cal' respectively stand for the experimental and calculated data, n is the number of experimental points.

For MEA–MeOH aqueous solutions, with the experimental surface tension of MEA aqueous solutions as input, the model param-

eters for MEA aqueous solutions were optimized as $a_{13} = -0.566$, $b_{13} = 1.05$ and $c_{13} = 0.549$ with ARD = 0.45%. Similarly, a_{23} , b_{23} and c_{23} can be regressed from the experimental data of MeOH aqueous solutions. Once a_{13} , b_{13} , c_{13} , a_{23} , b_{23} and c_{23} are obtained, a_{12} , b_{12} and c_{12} , can be regressed from the experimental data of MEA–MeOH aqueous solutions. However, it is worth noting that the mass fraction dependence of the surface tension of MeOH aqueous solutions is similar to that of the surfactant aqueous solutions, i.e., when the values of the mass fraction of MeOH (w_{MeOH}) are small, the surface tension decreases monotonically and rapidly with increasing w_{MeOH} . Exceeding a certain value, the surface tension tends to change slightly. The present model is unable to describe accurately such a tendency of surface tension. For example, we correlated the surface tension of MeOH aqueous solutions under w_{MeOH} (from 0.05 to 0.9). The optimized model parameters are $a_{23} = -0.787$, $b_{23} = 1.35$ and $c_{23} = -0.723$. The ARD is 2.35%. With the optimized 6 parameters (a_{13} , b_{13} , c_{13} , a_{23} , b_{23} and c_{23}) as input, we correlated the surface tensions of MEA–MeOH aqueous solutions, the ARD is 5.18%. It seems when there are only 3 adjustable parameters (a_{12} , b_{12} and c_{12}), the correlation is not satisfactory. Hence, the parameters a_{23} , b_{23} , c_{23} , a_{12} , b_{12} and c_{12} are simultaneously regressed from the experimental results of MEA–MeOH aqueous solutions.

Using the experimental values presented in Table 3, we optimized the residual 6 model parameters of MEA–MeOH aqueous solutions as: $a_{23} = -0.556$, $b_{23} = -3.34$, $c_{23} = 33.0$, $a_{12} = 6.85$, $b_{12} = -17.9$ and $c_{12} = -6.45$. The ARD is 0.76%. The binary parameters and the corresponding ARD are presented in Table 5. Fig. 3 shows the influence of w_{MeOH} on the surface tension of MEA–MeOH aqueous solutions. One finds that at given temperature and given mass fraction of MEA (w_{MEA}), the surface tension decreases with the increase of the w_{MeOH} . Fig. 4 shows the temperature dependence of the surface tension of MEA–MeOH aqueous solutions. One finds that at given w_{MEA} and w_{MeOH} , the surface

Table 5
Model parameters of surface tension and the corresponding average relative deviation (ARD).

Model parameters	MEA-MeOH	DEA-MeOH
a_{13}	-0.566	-1.62
b_{13}	1.05	7.29
c_{13}	0.549	-10.4
ARD	0.45%	1.18%
a_{23}	-0.556	-0.885
b_{23}	-3.34	4.55
c_{23}	33.0	-10.8
a_{12}	6.85	8.42
b_{12}	-17.9	-21.5
c_{12}	-6.45	-8.01
ARD	0.76%	1.07%

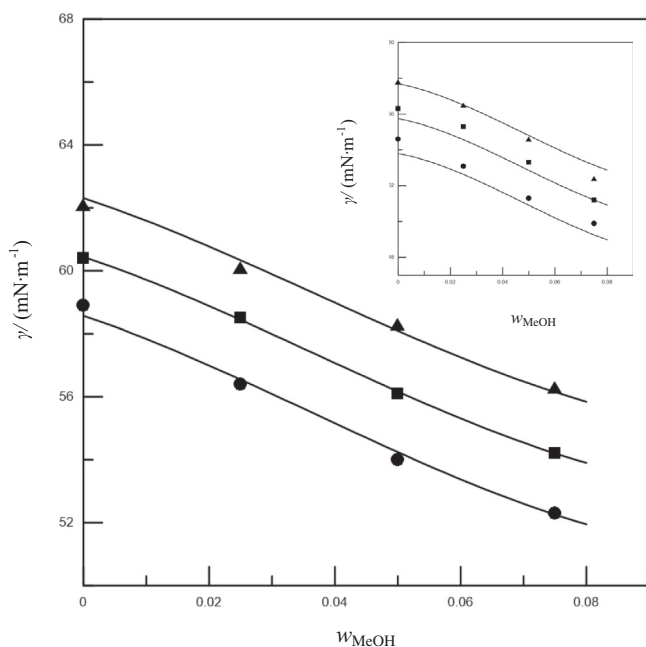


Fig. 3. Effect of w_{MeOH} on the surface tension of MEA-MeOH aqueous solutions. $w_{\text{MEA}} = 0.20$ and $w_{\text{MEA}} = 0.40$ (inset). Symbols: experiments from this work, $\blacktriangle T = 303.2$ K; $\blacksquare T = 313.2$ K; $\bullet T = 323.2$ K. Lines: calculated values.

tension decreases with the increase of temperature. The agreement between the experiments and calculations is satisfactory in both Figs. 3 and 4. One may also find from Figs. 3 and 4 that the predicted surface tension decreases almost linearly with the increase of temperature, however, the relationship between the surface tension and w_{MeOH} cannot be described using linear functions.

For DEA-MeOH aqueous solutions, with the experimental surface tension of DEA aqueous solutions as input, the model parameters for DEA aqueous solutions were optimized as $a_{13} = -1.62$, $b_{13} = 7.29$ and $c_{13} = -10.4$ with $\text{ARD} = 1.18\%$. With the model parameters of DEA as input, the 6 residual parameters, a_{23} , b_{23} , c_{23} , a_{12} , b_{12} and c_{12} can be regressed from the experimental data of DEA-MeOH aqueous solutions shown in Table 4. The optimized results are $a_{23} = -0.885$, $b_{23} = 4.55$, $c_{23} = -10.8$, $a_{12} = 8.42$, $b_{12} = -21.5$ and $c_{12} = -8.01$. The ARD is 1.07%. The binary parameters and the corresponding ARD are also presented in Table 5. Fig. 5 shows the influence of w_{MeOH} on the surface tension of DEA-MeOH aqueous solutions. Similar to the case shown in Fig. 3, the surface tension decreases monotonically with the increase of w_{MeOH} at given temperature and given mass fraction

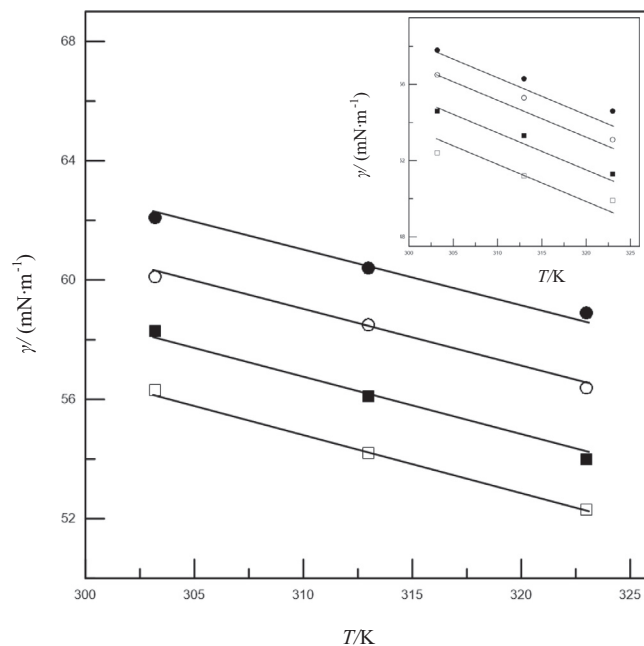


Fig. 4. Effect of temperature on the surface tension of MEA-MeOH aqueous solutions. $w_{\text{MEA}} = 0.20$ and $w_{\text{MEA}} = 0.40$ (inset). Symbols: experiments from this work, $\square w_{\text{MeOH}} = 0.00$; $\blacksquare w_{\text{MeOH}} = 0.025$; $\circ w_{\text{MeOH}} = 0.05$; $\bullet w_{\text{MeOH}} = 0.075$. Lines: calculated values.

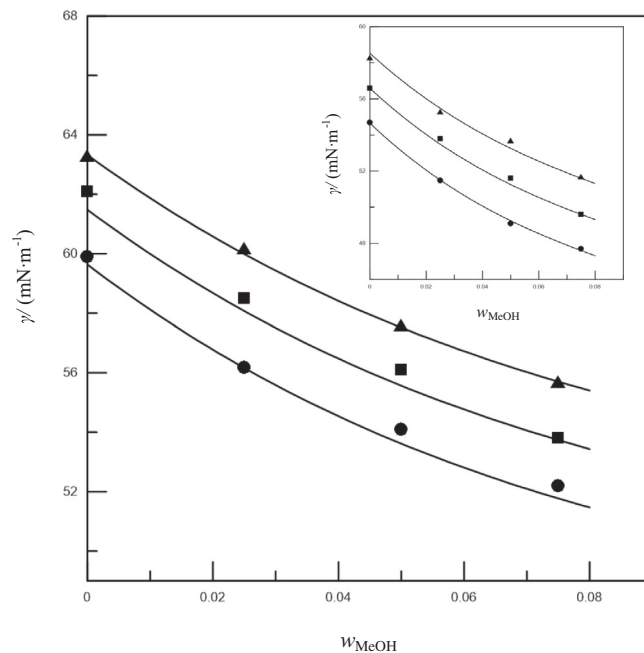


Fig. 5. Effect of w_{MeOH} on the surface tension of DEA-MeOH aqueous solutions. $w_{\text{DEA}} = 0.20$ and $w_{\text{DEA}} = 0.40$ (inset). Symbols: experiments from this work, $\blacktriangle T = 303.2$ K; $\blacksquare T = 313.2$ K; $\bullet T = 323.2$ K. Lines: calculated values.

of DEA (w_{DEA}). Moreover, the relationship between the surface tension and w_{MeOH} cannot be described using a linear function. Fig. 6 shows the temperature dependence of the surface tension of DEA-MeOH aqueous solutions. One finds that at given w_{DEA} and w_{MeOH} , the surface tension decreases linearly with the increase of temperature. The agreement between the experiments and calculations is also satisfactory in both Figs. 5 and 6.

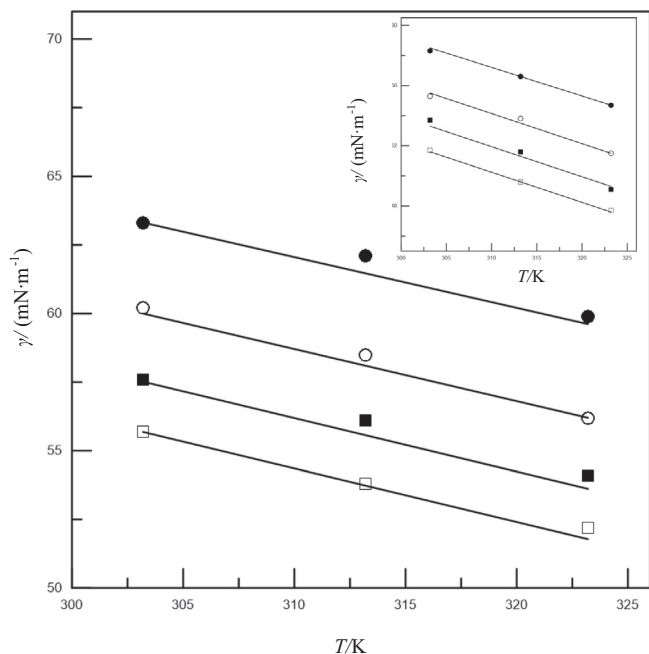


Fig. 6. Effect of temperature on the surface tension of DEA-MeOH aqueous solutions. $w_{\text{DEA}} = 0.20$ and $w_{\text{DEA}} = 0.40$ (inset). Symbols: experiments from this work, $\square w_{\text{MeOH}} = 0.00$; $\blacksquare w_{\text{MeOH}} = 0.025$; $\circ w_{\text{MeOH}} = 0.05$; $\bullet w_{\text{MeOH}} = 0.075$. Lines: calculated values.

3.2. Viscosity and the model

The experimental results of the viscosities of MEA-MeOH and DEA-MeOH aqueous solutions are respectively shown in Tables 6 and 7. Besides experiments, equations that can correctly correlate the viscosities are also important. As summarized in the work of Zhang and Yu [70], a variety of semi-empirical equations have been proposed to describe the viscosities of fluids in recent years [71–73]. In this work, the Weiland equation [74] was used to fit the viscosities of MEA-MeOH and DEA-MeOH aqueous solutions because of its simplicity and accuracy.

When applied to amine-MeOH aqueous solutions, the Weiland equation can be expressed as:

$$\eta_{\text{mix}} = \frac{w_{\text{amine}}}{w_{\text{amine}} + w_{\text{MeOH}}} \eta_1 + \frac{w_{\text{MeOH}}}{w_{\text{MeOH}} + w_{\text{amine}}} \eta_2 \quad (8)$$

where η_{mix} is the viscosity of the aqueous solution, w_{amine} and w_{MeOH} respectively stand for the mass fractions of amine and

Table 6
Viscosities (η) of MEA-MeOH aqueous solutions under different mass fractions of MEA (w_{MEA}) and MeOH (w_{MeOH}) and temperatures. Pressure (p) = 101 kPa.^a

w_{MEA}	w_{MeOH}	$\eta/(\text{mPa}\cdot\text{s})$		
		303.2 K	313.2 K	323.2 K
0.200	0.025	1.59	1.29	1.07
	0.050	1.71	1.41	1.19
	0.075	1.83	1.54	1.33
0.300	0.025	2.21	1.72	1.39
	0.050	2.35	1.86	1.52
	0.075	2.51	2.01	1.66
0.400	0.025	3.27	2.43	1.91
	0.050	3.44	2.59	2.05
	0.075	3.63	2.8	2.22

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w_{\text{MEA}}) = \pm 0.002$; $u(w_{\text{MeOH}}) = \pm 0.001$; $u(p) = 2$ kPa; $u(\eta) = 0.06$ mPa·s.

Table 7

Viscosities (η) of DEA-MeOH aqueous solutions under different mass fractions of DEA (w_{MEA}) and MeOH (w_{MeOH}) and temperatures. Pressure (p) = 101 kPa.^a

w_{DEA}	w_{MeOH}	$\eta/(\text{mPa}\cdot\text{s})$		
		303.2 K	313.2 K	323.2 K
0.200	0.025	1.75	1.42	1.18
	0.050	1.87	1.53	1.33
	0.075	2.03	1.65	1.44
0.300	0.025	2.72	2.14	1.61
	0.050	2.85	2.31	1.83
	0.075	3.02	2.49	2.08
0.400	0.025	4.51	3.28	2.47
	0.050	4.85	3.62	2.72
	0.075	5.51	4.06	3.07

^a Standard uncertainties u are $u(T) = 0.1$ K; $u(w_{\text{DEA}}) = \pm 0.002$; $u(w_{\text{MeOH}}) = \pm 0.001$; $u(p) = 2$ kPa; $u(\eta) = 0.08$ mPa·s.

MeOH. η_1 and η_2 are respectively the viscosities of amine aqueous solutions and MeOH aqueous solutions, which can be expressed as:

$$\eta_i/\eta_{\text{water}} = \exp\left\{[(a_i w + b_i)T + (c_i w + d_i)]w/T^2\right\} \quad (9)$$

where η_{water} is the viscosity of pure water, $w = w_{\text{amine}} + w_{\text{MeOH}}$ is the total mass fraction of amine and MeOH. The parameters a_i , b_i , c_i and d_i can be regressed from the experimental results of their binary aqueous solutions. The objective function (ARD) was expressed as:

$$fs = \sum_{i=1}^n [1 - \eta^{\text{cal}}/\eta^{\text{exp}}] \times 100\%/n \quad (10)$$

where the superscripts ‘exp’ and ‘cal’ respectively stand for the experimental and calculated data, n is the numbers of the data.

The model parameters of MEA, $a_1 = 0$, $b_1 = 0$, $c_1 = 21.186$ and $d_1 = 2373$, were taken directly from the previous work of Weiland et al.[58]. The model parameters of MeOH can also be regressed from the experimental data of their binary aqueous solutions. However, when the model parameters of both MEA and MeOH are given, there are no adjustable parameters in the Weiland equation and the deviation between the experiments and calculations would be very significant. Thus in this work, the model parameters of MeOH were regressed by fitting to the experiments of MEA-MeOH aqueous solutions. The optimized values are $a_2 = 2.789$, $b_2 = 27.968$, $c_2 = -1132.551$ and $d_2 = -66.608$. The average relative deviation is 5.87%. The binary parameters and the corresponding ARD are presented in Table 8.

Fig. 7 shows the effect of w_{MeOH} on the viscosity of MEA-MeOH aqueous solution. One may find from the figure that at given temperature and given w_{MEA} , the viscosity increases with increasing w_{MeOH} . Fig. 8 shows the influence of temperature on the viscosity of MEA-MeOH aqueous solution, indicating that the viscosity decreases with the increase of temperature at given w_{MeOH} and w_{MEA} . The effects of w_{MEA} , w_{MeOH} and temperature on viscosities

Table 8

Model parameters of viscosity and the corresponding average relative deviation (ARD).

Model parameters	MEA-MeOH	DEA-MeOH
a_1	0	-0.0724
b_1	0	-3.436
c_1	21.186	54.319
d_1	2373	3628
a_2	2.789	6.034
b_2	27.968	26.715
c_2	-1132.551	-2135.450
d_2	-66.608	-68.127
ARD	5.87%	4.61%

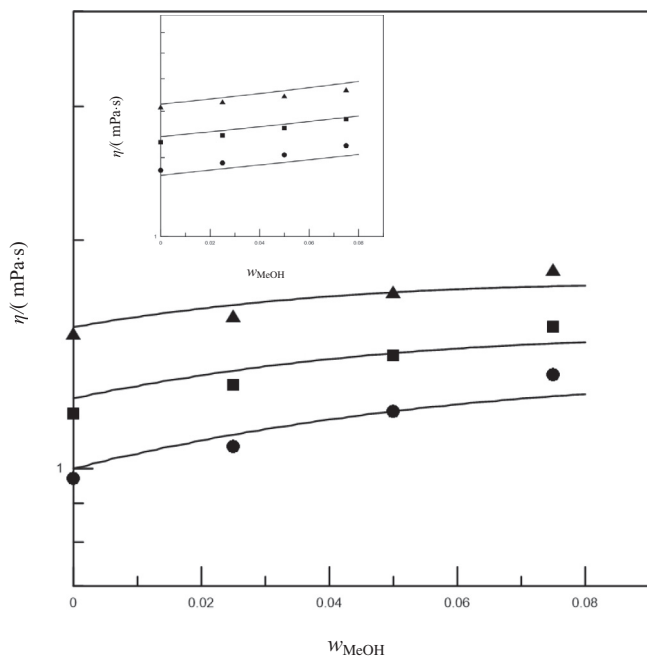


Fig. 7. Effect of w_{MeOH} on the viscosity of MEA-MeOH aqueous solutions. $w_{\text{MEA}} = 0.20$ and $w_{\text{MEA}} = 0.40$ (inset). Symbols: experiments from this work, $\blacktriangle T = 303.2$ K; $\blacksquare T = 313.2$ K; $\bullet T = 323.2$ K. Lines: calculated values.

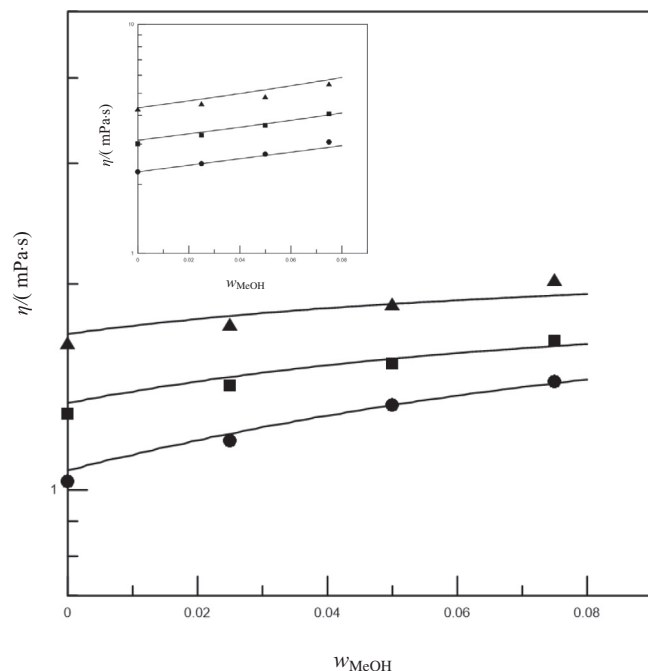


Fig. 9. Effect of w_{MeOH} on the viscosity of DEA-MeOH aqueous solutions. $w_{\text{DEA}} = 0.20$ and $w_{\text{DEA}} = 0.40$ (inset). Symbols: experiments from this work, $\blacktriangle T = 303.2$ K; $\blacksquare T = 313.2$ K; $\bullet T = 323.2$ K. Lines: calculated values.

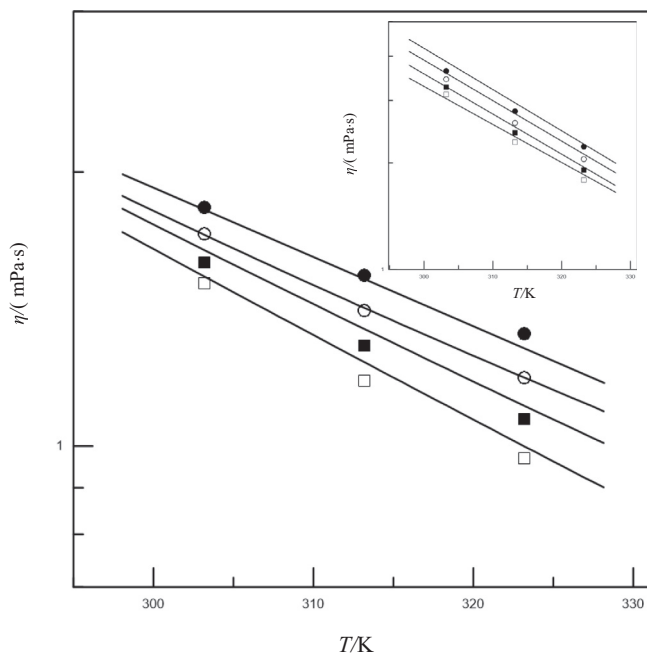


Fig. 8. Effect of temperature on the viscosity of MEA-MeOH aqueous solutions. $w_{\text{MEA}} = 0.20$ and $w_{\text{MEA}} = 0.40$ (inset). Symbols: experiments from this work, $\square w_{\text{MeOH}} = 0.00$; $\blacksquare w_{\text{MeOH}} = 0.025$; $\circ w_{\text{MeOH}} = 0.05$; $\bullet w_{\text{MeOH}} = 0.075$. Lines: calculated values.

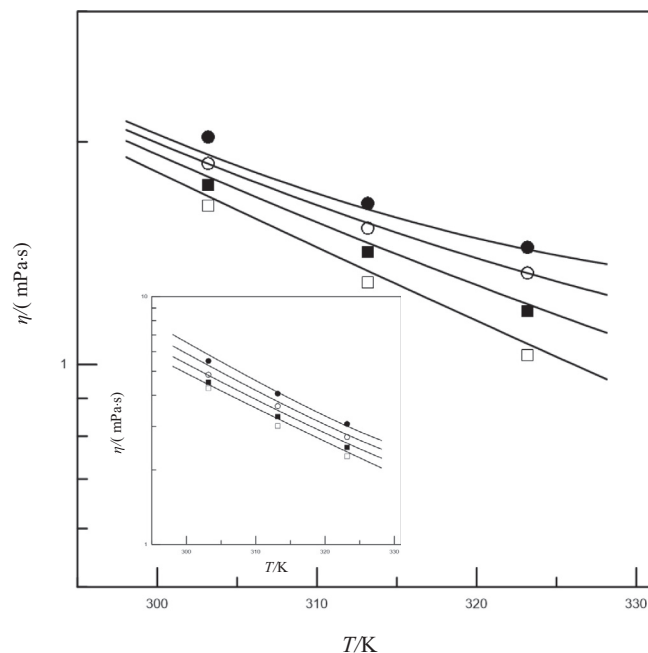


Fig. 10. Effect of temperature on the viscosity of DEA-MeOH aqueous solutions. $w_{\text{DEA}} = 0.20$ and $w_{\text{DEA}} = 0.40$ (inset). Symbols: experiments from this work, $\square w_{\text{MeOH}} = 0.00$; $\blacksquare w_{\text{MeOH}} = 0.025$; $\circ w_{\text{MeOH}} = 0.05$; $\bullet w_{\text{MeOH}} = 0.075$. Lines: calculated values.

can be well captured by the Weiland equation, and the agreement between the experiments and calculations is satisfactory.

For DEA-MeOH aqueous solutions, the model parameters of DEA, $a_1 = -0.0724$, $b_1 = -3.436$, $c_1 = 54.319$ and $d_1 = 3628$, were also taken from the previous work of Weiland et al. When regressed the model parameters of MeOH by fitting the experiments of DEA-MeOH aqueous solutions, the optimized values are

$a_2 = 6.034$, $b_2 = 26.715$, $c_2 = -2135.450$ and $d_2 = -68.127$. The average relative deviation is 4.61%. The binary parameters and the corresponding ARD are also presented in Table 8.

Fig. 9 shows the effect of w_{MeOH} on the viscosity of DEA-MeOH aqueous solution. Similar to the case shown in Fig. 7, the viscosity increases with increasing w_{MeOH} at given temperature and given w_{DEA} . Fig. 10 shows the influence of temperature on the viscosity

of DEA-MeOH aqueous solution, indicating that the viscosity decreases with the increase of temperature at given w_{MeOH} and w_{DEA} . The calculated results agree well with the experimental results.

4. Conclusions

In this work, the surface tension and viscosity of MEA-MeOH and DEA-MeOH aqueous solutions were measured and satisfactorily modelled. The effects of temperature, mass fractions of amines and MeOH on the surface tension and viscosity were demonstrated. Our results show that:

- (1) The surface tensions of MEA-MeOH and DEA-MeOH aqueous solutions decrease almost linearly with increasing temperature within the experimental ranges of temperature and mass fraction. However, the relationship between the surface tension and w_{MeOH} cannot be described using linear functions;
- (2) The viscosities of MEA-MeOH and DEA-MeOH aqueous solutions increase with the increase of w_{MeOH} at given temperature and given mass fraction of amines, and decreases with increasing temperature at a given w_{MeOH} and mass fraction of amines;
- (3) The proposed equations correctly capture the effects of temperature, mass fraction of amines and MeOH on the surface tension and viscosity, and the calculated results agree well with the experimental results.

Acknowledgments

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