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Study on the Viscosities of MDEA-Amino Acid Ionic Liquid Aqueous Solutions

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Abstract. The viscosities of 1-butyl-3-methylimidazolium glycine ([Bmim][Gly])-N-methyldiethanolamine (MDEA), tetramethyl amine glycine ([N₁₁₁₁][Gly])-MDEA and 1-butyl-3-methyl-imidazolium lysine ([Bmim][Lys])-MDEA aqueous solutions were respectively measured by using the NDJ-5S digital rotational viscometer. The temperatures ranged from 303.2K to 323.2K. The mass fractions of MDEA and amino acid ionic liquids (AAILs) respectively ranged from 0.30 to 0.45 and 0.05 to 0.15. The Weiland equation was used to correlate and predict the viscosities of [Bmim][Gly]-MDEA, [N₁₁₁₁][Gly]-MDEA and [Bmim][Lys]-MDEA aqueous solutions. The temperature and mass fraction dependences of the viscosity of MDEA-AAIL aqueous solutions were demonstrated on the basis of experiments and calculations.

1. Introductions

The great amount of CO₂ emitted by coal-fired boilers has become the main cause of the greenhouse effect, and its contribution to the greenhouse effect has reached more than 60%. As the largest coal consumer in the world, the coal in China accounts for more than 2/3 of the primary energy structure. Moreover, more CO₂ has been emitted with per unit GDP than other countries ^[1]. In recent years, chemical absorption method is considered to be the common methods in the CO₂ emission reduction measures, which are actively promoted at home and abroad. The decarbonization essence of chemical absorption is the contact of absorbent solution with CO₂ in flue gas, and form unstable salts. Under certain conditions, the salt will decompose in reverse and release CO₂ to regenerate, thus achieving the separation of CO₂ ^[2]. It is well known that the alkanolamines absorption method ^[3-8] takes the advantages of mature technology, large absorption capacity and low operating cost. However, the equipment is easily to be corroded after absorption of CO₂ in the primary alkanolamine and the secondary alkanolamine. Due to the disadvantages of low mass fraction of alkanolamine, the absorption capacity per unit mass of CO₂ is small, and the amount of steam used for heating in desorption is increased due to the large amount of water in the solution ^[9]. Many alkanolamine reagents have been proposed for the removal of CO₂, among which the N-methyldiethanolamine (MDEA) has the advantages of large decarbonization capacity, low corrosiveness, low partial pressure of aqueous solution and low viscosity. MDEA is considered to be a good absorbent to capture CO₂ due to the large absorption cycle, low heat



consumption in regeneration and low operation cost, but it has low absorption rate. Therefore, it needs to be compounded with reagents for faster absorption rate^[10]. Xiang et al.^[11] determined the absorption rate and the amount of CO₂ in flue gas of power plant with two mixed solutions of MDEA-diethylenetriamine (DETA) and MDEA-triethylenetetramine (TETA).

In recent years, ionic liquids, especially amino acid ionic liquid (AAIL), have received extensive attention in CO₂ capture^[12-17]. AAIL is characterized by high absorption rate, large absorption capacity, stable performance, extremely low vapor pressure, low regeneration temperature and no corrosion when absorbing CO₂. At the same time, compared with traditional alkanolamine absorbents such as MEA and MDEA, AAIL has the advantages of easy availability of raw materials, stable preparation process and low product toxicity^[12]. The experimental results show that the absorption rate of CO₂ can be significantly increased by adding a small amount of AAIL to the MDEA aqueous solution, such as tetramethyl amine glycine [N₁₁₁₁][Gly]. Fu et al.^[16] added a small amount of 1-butyl-3-methylimidazolium glycine ([Bmim][Gly]) in MDEA aqueous solution, which could significantly improve the absorption rate of MDEA aqueous solution, and the absorption rate was higher than that of MDEA-MEA and MDEA-N, N-dimethyl-iso-propanolamine (DMA2P) composite solution. Considering the advantages and disadvantages of AAIL, a new type of absorbent can be obtained by combining AAIL with MDEA, which can not only overcome the slow absorption rate of MDEA, but also prevent the influence of high viscosity of single AAIL solution. Few experiments have been reported on the viscosity of AAIL-MDEA aqueous solution. Gao et al.^[18] determined the viscosity of MDEA-[N₁₁₁₁][Gly] aqueous solution in the range of 298K-343K. However, the determination and calculation of the viscosity of MDEA-AAIL aqueous solution in the wide range of concentrations is rarely reported. It is necessary to carry out systematic experimental determination and model calculation on the viscosity of MDEA-AAIL aqueous solution, thereby providing basic data and calculation for the engineering design involved mass transfer and CO₂ absorption in MDEA-AAIL aqueous solution.

In this paper, the viscosity of MDEA with three amino acid ionic liquids, including [Bmim][Gly], [N₁₁₁₁][Gly] and [Bmim][Lys], was determined in the temperature range of 303.2K-323.2K, and the mass fractions of MDEA and amino acid ionic liquids (AAILs) ranged from 0.30 to 0.45 and 0.05 to 0.15, respectively. In addition, the viscosity of three MDEA-AAIL aqueous solutions was correlated and predicted by the Weiland equation, and the relationship among the viscosity of MDEA-AAIL aqueous solution, temperature and the mass fraction of each component was clarified.

2. Experimental

The NDJ-5S digital rotary viscometer and the HWY-501 circulating thermostat with constant temperature used in the experiment were produced by Shanghai Changji Geological Instrument Factory. The rotor of the rotary viscometer ranges from 0 to 100mPa·s with an accuracy of 0.05mPa·s, and the accuracy of temperature control in constant temperature water tank is 0.1K. The accuracy of the electronic balance is ±0.1mg. During the measurement, 30g of measured liquid is injected into the outer sleeve after installing the rotor and fixing the sleeve, the sleeve is fixed and tightened, and the height in the super thermostatic tank is adjusted for heating. When the temperature is constant, turn on the switch, and the corresponding viscosity value can be obtained by reading the data when the viscosity data is stable. Each measurement was repeated three times and averaged.

The deionized water was prepared by the Environmental College of North China Electric Power University; MDEA was 99.5% of analytically pure, and provided by Shanghai Maclean Biochemical Co., Ltd.; amino acid ionic liquid with a purity of 99% was provided by Shanghai Chengjie Chemical Co., Ltd.

3. Results and discussion

In this paper, the viscosity of MDEA-AAIL aqueous solution was determined in the temperature range of 303.2K-323.2K, and the sum of mass fractions of MDEA and AAILs ranged from 0.30 to 0.60. The results are shown in Table 1.

Table 1. Viscosities of MDEA-AAILs aqueous solutions.

w_{MDEA}	w_{AAILs}	$w_{[\text{Bmim}][\text{Gly}]}$			$w_{[\text{N1111}][\text{Gly}]}$			$w_{[\text{Bmim}][\text{Lys}]}$		
		$\eta / (\text{mPa}\cdot\text{s})$			$\eta / (\text{mPa}\cdot\text{s})$			$\eta / (\text{mPa}\cdot\text{s})$		
		303.2K	313.2K	323.2K	303.2K	313.2K	323.2K	303.2K	313.2K	323.2K
0.3001	0.0499	3.41	2.55	2.04	3.45	2.60	2.06	4.43	3.25	2.54
	0.1001	4.40	3.03	2.49	4.13	3.15	2.55	5.23	3.36	2.62
	0.1499	5.29	3.69	2.79	5.32	3.77	2.92	6.69	5.03	3.85
0.3501	0.0501	4.33	3.21	2.44	4.47	3.30	2.59	4.88	3.62	2.74
	0.1001	5.52	3.76	2.85	5.77	4.09	3.10	5.95	4.34	3.47
	0.1501	6.59	4.67	3.40	6.56	4.68	3.50	8.36	5.82	4.48
0.4001	0.0501	5.62	4.27	3.14	5.65	4.10	3.14	6.43	4.79	3.70
	0.0999	6.91	4.85	3.52	7.12	5.00	3.71	8.06	5.53	4.32
	0.1501	8.60	5.83	4.24	8.80	6.02	4.42	11.65	7.72	5.33
0.4501	0.0501	7.31	5.05	3.68	7.44	5.19	3.81	8.41	5.85	4.58
	0.1001	9.21	6.20	4.46	9.34	6.37	4.58	10.90	7.14	5.09
	0.1499	11.7	7.74	5.47	12.35	8.31	5.94	15.10	9.74	6.81

In order to clarify the relationship among the viscosity of MDEA-AAIL aqueous solution, temperature and the mass fraction of each component, it is necessary to correlate and predict the experimental data with a suitable calculation model of viscosity. Pure AAIL belongs to Newtonian fluid, and the viscosity of AAIL can be expressed by the Arrhenius equation and the improved equation ^[19] but the equations are not suitable for the MDEA-AAIL aqueous solution. The empirical equation proposed by Weiland et al. ^[20] can correctly describe the quantitative relationship among viscosity, temperature and mass fraction of components in mixed solution:

The Weiland equation is as follows:

$$\eta_{\text{mix}} = \frac{w_1}{w_1 + w_2} \eta_1 + \frac{w_2}{w_1 + w_2} \eta_2 \quad (1)$$

Where η_{mix} is the viscosity of the MDEA-AAIL aqueous solution, w_1 is the mass fraction of MDEA, and w_2 is the mass fraction of AAIL.

η_1 and η_2 are expressed as follows:

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

$$w = w_1 + w_2 \quad (3)$$

When using the Weiland equation to describe the viscosity of the MDEA-AAIL aqueous solution, in addition to the parameters of MDEA-H₂O ^[21], each mixed solution requires four tunable parameters, which can be correlated with the experimental data listed in Tables 1. The objective function in the associated process is as follows:

$$fs = \sum_{i=1}^n \left[1 - \frac{\eta^{\text{cal}}}{\eta^{\text{exp}}} \right] \times 100\% / n \quad (4)$$

where the superscripts ‘exp’ and ‘cal’ are experimental data and the calculated values of the model, respectively, and n is the number of experimental points. When the objective function is the smallest, the optimal tunable value is obtained, and the average relative deviation (ARD) is used to indicate the

degree of agreement between the experimental value and the calculated value. The optimized model parameters and corresponding ARD are shown in Table 2:

Table 2. Model parameters in Weiland equation.

Parameters	MDEA-H ₂ O	MDEA-[N ₁₁₁₁][Gly]	MDEA-[Bmim][Gly]	MDEA-[Bmim][Lys]
<i>a</i>	-0.1944	-0.0200	-0.0232	-0.2051
<i>b</i>	0.4315	11.2629	10.3236	19.1786
<i>c</i>	80.648	-0.0551	-0.0989	41.0444
<i>d</i>	2889.1	-1.6346	-0.0564	-68.5842
ARD		2.44%	1.83%	5.11%

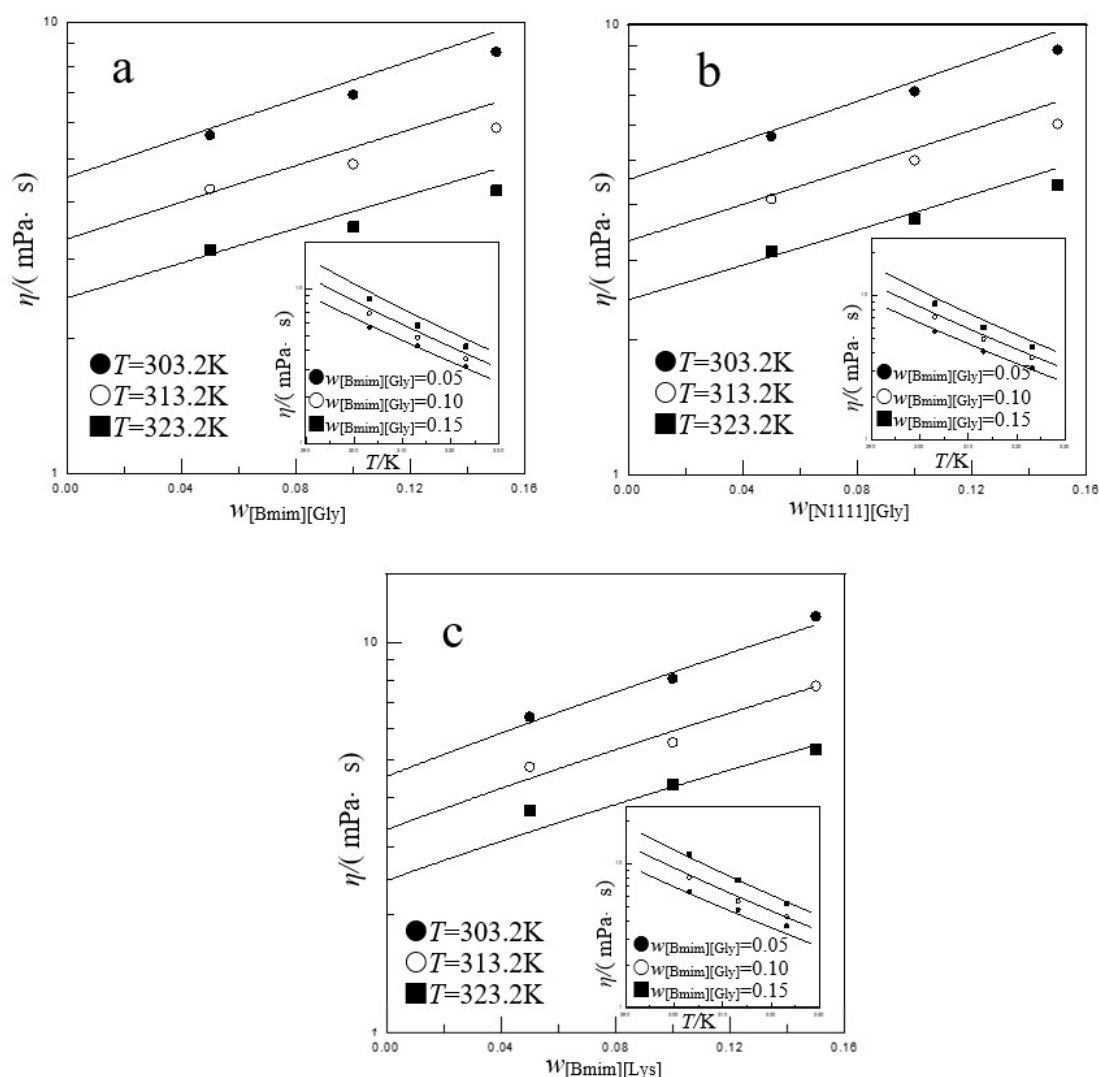


Figure 1. Effects of mass fraction and temperature (insert plot) on the viscosities of MDEA-[Bmim][Gly](a), MDEA-[N₁₁₁₁][Gly](b), MDEA-[Bmim][Lys](c) aqueous solutions. $w_{\text{MDEA}}=0.40$.

Based on the obtained model parameters, the viscosity of MDEA-AAIL aqueous solution was predicted by the Weiland equation. The comparison between the calculated results and the experimental values is shown in Figure 1. The experimental results show that when the concentration of MDEA and AAIL were constant, the viscosity of the compound aqueous solution decreased monotonously with the

increase of temperature; when the temperature was constant, the viscosity of the compound aqueous solution increased monotonously with the increased concentration of AAIL. When the total mass fraction of mixed solution is high, the effect of temperature on viscosity is greater than that of low mass fraction. For example, when $w_{\text{MDEA}}=0.30$, $w_{[\text{N1111}][\text{Gly}]}=0.05$, the viscosity decreased from 3.45mPa·s at 303.2K to 2.06mPa·s at 323.2K; when $w_{\text{MDEA}}=0.45$, $w_{[\text{N1111}][\text{Gly}]}=0.15$, the viscosity decreased from 12.35mPa·s at 303.2K to 5.94mPa·s at 323.2K. In addition, when the total mass fraction was less than 0.40, the viscosity of the mixed solution was similar to that of MEA with the mass fraction of 0.15 at 303.2K-323.2K^[22]. When the total mass fraction of MDEA and AAIL was less than 0.60, the Weiland equation of viscosity could accurately predict the relationship among the viscosity of MDEA-AAIL aqueous solution, temperature and mass fraction of components.

Figure 2 and Figure 3 showed the relationship between the three MDEA-AAIL aqueous solutions with the MDEA mass fraction and the AAIL mass fraction, respectively. The results showed that the viscosity of the compound solution increased monotonously with the increased concentration of MDEA and AAIL. Under the same conditions, the viscosity of the three MDEA-AAILs aqueous solutions was $\text{MDEA-}[\text{Bmim}][\text{Lys}]>\text{MDEA-}[\text{N1111}][\text{Gly}]>\text{MDEA-}[\text{Bmim}][\text{Gly}]$ in the same concentrations and temperatures. In general, there is little difference among the viscosity of the three mixed solutions, especially the viscosity of MDEA- $[\text{N1111}][\text{Gly}]$ and MDEA- $[\text{Bmim}][\text{Gly}]$ aqueous solutions at higher temperatures and at lower concentrations. For example, when the mass fraction of MDEA and AAIL were 0.30 and 0.05, respectively, and the temperature was 323.2K, the viscosity of MDEA- $[\text{Bmim}][\text{Gly}]$ aqueous solution was 2.04mPa·s, which was close to 2.06mPa·s of the viscosity of MDEA- $[\text{N1111}][\text{Gly}]$ aqueous solution. When $w_{\text{MDEA}}=0.45$, $w_{\text{AAIL}}=0.15$, and the temperature was 303.2K, the difference between the viscosity of MDEA- $[\text{Bmim}][\text{Gly}]$ and MDEA- $[\text{Bmim}][\text{Lys}]$ aqueous solution was the largest, which was 3.4mPa·s.

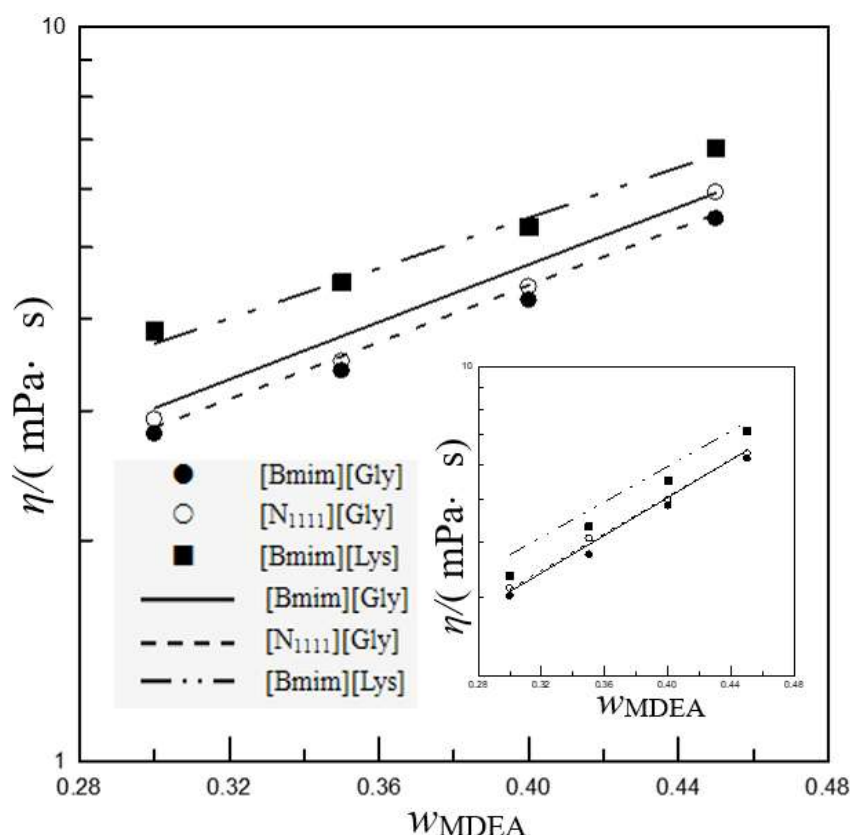


Figure 2. Effects of w_{MDEA} on the viscosity of MDEA-AAIL aqueous solutions. Main plot: $w_{\text{AAIL}}=0.10$, 313.2K; Insert plot: $w_{\text{AAIL}}=0.15$, 323.2K.

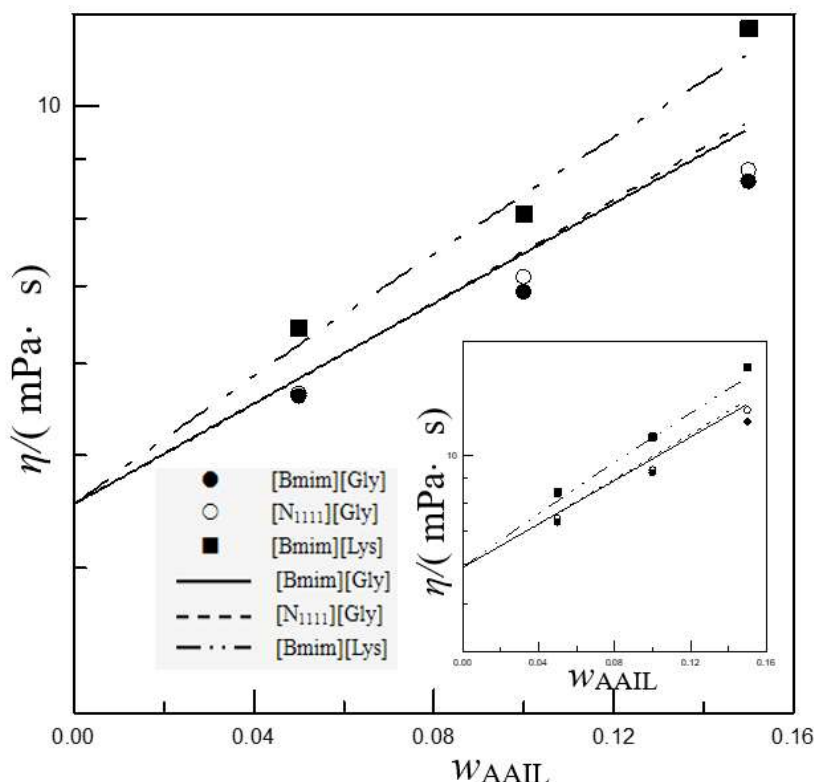


Figure 3. The concentration of AAIL dependence of the viscosity of MDEA-AAIL aqueous solution in the case of $w_{\text{MDEA}}=0.40$, 303.2K and $w_{\text{AAIL}}=0.45$, 323.2K (insert plot).

4. Conclusion

In this work, the viscosity of MDEA-AAIL aqueous solution was measured by using the NDJ-5S digital rotational viscometer in the mass fraction of MDEA and AAIL range of 0.30-0.45 and 0.05-0.15, respectively, and the temperature range from 303.2K to 323.2K. The Weiland equation was used to correlate and predict the viscosities of [Bmim][Gly]-MDEA, [N₁₁₁₁][Gly]-MDEA and [Bmim][Lys]-MDEA aqueous solutions. The results show that the viscosity of MDEA-AAIL aqueous solution decreased with the increase of temperature when the mass fraction of MDEA was constant; the viscosity of MDEA-AAIL aqueous solution increased with the increase of AAIL concentration when the temperature was constant. The viscosity of the three MDEA-AAIL aqueous solutions was MDEA-[Bmim][Lys]>MDEA-[N₁₁₁₁][Gly]>MDEA-[Bmim][Gly] when the concentration and temperature were constant, and the difference was not significant.

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