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SHORT ARTICLE

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Density, Viscosity, and Surface Tension of Aqueous Solutions of Potassium Glycinate + Piperazine in the Range of (288.15 to 323.15) K

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ABSTRACT: Densities and viscosities of aqueous solutions of potassium glycinate (PG) + piperazine (PZ) composed of x_{PG}/x_{PZ} (mol/mol) = 1.0/0, 0.9/0.1, 0.8/0.2, 0.7/0.3, and 0.6/0.4 have been measured at (288.15, 303.15, 313.15, and 323.15) K. Surface tensions of aqueous solutions of (PG + PZ) have been determined at 288.15 K. Correlations by means of mathematical fitting were developed for the predictions of density, viscosity, and surface tension of the aqueous solutions of (PG + PZ) using the experimental data of this work. Results show that the densities and viscosities decrease as the temperature increases. The densities of the studied solutions decrease with the increase of PZ mole fraction in the solutions, while viscosities of the solutions increase. The surface tensions of the solutions almost decrease linearly as the PZ mole fraction in the solution increases from 0 to 0.4. The prediction values from correlation equations for density, viscosity, and surface tension are in good agreement with the experimental values. The correlation equations for each property can offer additional data for aqueous solutions of (PG + PZ).

INTRODUCTION

Excessive CO_2, which is caused by intensive human activities, is considered as one of the contributions to global warming that results in serious environmental problems.[1] Most commercial processes for the removal of bulk CO_2 from industrial gas streams involve the use of absorbents. The absorbents are usually amine-based aqueous solutions, for example, monoethanolamine (MEA), diethanolamine (DEA), *N*-methyldiethanolamine (MDEA), and 2-amino-2-methyl-1-propanol (AMP).[2] Recently, researchers have focused their attention to aqueous amino acid salts as the CO_2 absorbent because amino acid salts are nontoxic, thermally stable, able to be regenerated, and commercially available and have low vapor pressure.[3] Piperazine (PZ), a symmetrical diamino and cyclic compound, has become a key component for CO_2 absorption.[4] Piperazine is more effective than conventional accelerators. The reaction rate constant of piperazine with CO_2 is higher than that of conventional alkanolamines such as MEA.[5] Consequently, new blended absorbents based on aqueous amino acid salts and PZ as an activator were developed for CO_2 removal.[6] They could maintain high absorption rates and increase absorption capacity, while decreasing generation energy consumption simultaneously. Therefore, knowledge of the physical properties of aqueous amino acid salt-based solutions is very important for the design of gas treatment processes, simulation, and operation of the treatment equipment.

Experimental data of the physical properties of PZ or aqueous amino acid salts have been studied. The density, viscosity, and surface tension of aqueous PZ,[7] aqueous (AMP + PZ),[8] and (MDEA + PZ)[9] are available in the published literature. The density, viscosity, and surface tension of aqueous amino acid salts can also be obtained from the literature.[10,11] However, the density, viscosity, and surface tension of aqueous blends of amino acid salt + PZ are scarce in the open literature.

In this work, the density and viscosity of the aqueous solution of potassium glycinate + piperazine (PG + PZ) were measured over the temperature range (288.15 to 323.15) K. The surface tension of the aqueous solution of (PG + PZ) was determined at 288.15 K. The density and viscosity as a function of temperature and surface tension as a function of concentration were correlated.

EXPERIMENTAL SECTION

Materials. Reagent grade glycine (≥ 99.5 %) was obtained from the Nanjing Chemical Reagent Co., Ltd., China. Reagent grade PZ (≥ 99.5 %) and KOH (85 %) were obtained from the Shanghai Ling Feng Chemical Reagent Co., Ltd., China. Reagent grade MDEA (≥ 99.0 %) was obtained from the Feixiang Chemical Industry Co., Ltd., Suzhou, China. The aqueous solutions of amines were prepared with doubly distilled and deionized water with a conductivity of 17 $\mu S \cdot cm^{-1}$. Glycine was neutralized with an equimolar quantity of potassium hydroxide to form the glycinate. Metered glycine was dissolved into deionized water to form a glycine solution, and then an equimolar quantity of KOH was added into the solution gradually until the end of reaction to form the glycinate solution. The glycinate solution was transferred into a 50 mL standard flask, and then deionized water was added into the flask to the mark. PZ as a component was added into the glycinate solution to form the blended solution. The blended solution was prepared in a 50 mL standard flask to a given concentration. The process of the preparation used the method with an analytical balance (FA2004, Shanghai Shang Tian Precision Instrument Co., Ltd., China.) with a precision and accuracy of ± 0.0001 g.

Density. The densities of the solutions were measured with a Gay-Lussac pycnometer (5 mL). The pycnometer containing the solution was immersed in a well-stirred constant temperature water bath (X-II, Nanjing Instrument Factory, China). The temperature of the bath was controlled with a digital thermometer system and with a

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Table 1. Comparison of the Densities, Viscosities, and Surface Tensions of Aqueous MDEA Measured in This Work with Literature Values at (288.15 and 313.15) K

		ρ/g·cm^{-3}		η/mPa·s		γ/mN·m^{-1}	
T/K	mass fraction/%	this work	ref 13	this work	ref 13	this work	ref 14
288.15	10	1.0071	1.0079	1.711	1.703		
	20	1.0175	1.0167	2.631	2.624		
313.15	10	1.0011	1.0004	0.905	0.899	58.71	58.45
	20	1.0081	1.0086	1.312	1.305	54.68	54.38

platinum resistance probe within ± 0.001 K precision. The readings from this thermometer were calibrated with a standard thermometer. Once the solution temperature reached the desired value, it was weighed by the analytical balance. Each reported density data point is the average of three measurements. The experimental uncertainty in the measured density was estimated to be ± 0.05 %.

Viscosity. Solution viscosities were measured with a suspended level Ubbelohde viscometer (R200, Yuyao Precise Instrument Co., Ltd., Zhejiang, China), which was placed in a well-stirred constant temperature water bath (X-II, Nanjing Instrument Factory, China). The viscometer was used at temperatures from (288.15 to 323.15) K. The temperature of the water bath was controlled within a ± 0.01 K uncertainty. The temperature was measured by a mercury thermometer. The flow time was measured with an accurate ± 0.01 s stop watch. Pure water was used for calibration. Measurements were repeated at least 10 times for each solution and temperature. The uncertainty of the viscosity measurements was estimated to be ± 0.5 %.

Surface Tension. The surface tensions of the solutions were determined at 288.15 K using a surface tension meter (CAM 200, KSV Instruments Co., Ltd., Finland), which was employed by means of the pendant drop method. Measurements were based on the drop shape of a fluid hanging from a syringe tip which revealed information on the fluid itself about the surrounding media.[12] The emergent drop image was captured by the apparatus. The data were calculated by the Young−Laplace equation by the PDA program. Thus, this method only needs the density data and a picture of the drop of the solution to calculate the surface tension. The surface tension was measured with a 1.7 % uncertainty. Each reported surface tension value was an average of five measurements.

RESULTS AND DISCUSSION

The average absolute deviation (AAD) between experimental and calculated values were estimated from the following equation

$$\mathrm{AAD}\% = \frac{100}{N}\sum_{i=1}^{N}\frac{|X_{\exp,i} - X_{\mathrm{cal},i}|}{X_{\exp,i}} \tag{1}$$

where N is the total number of data in the sample. Subscript exp and cal denote experimental and calculated values, respectively. X represents density (ρ, g·cm^{-3}), viscosity (η, mPa·s), or surface tension (γ, mN·m^{-1}).

Table 2. Densities for (PG + PZ + H_2O) from (288.15 to 323.15) K

	ρ/g·cm^{-3}				
x_{PG}/x_{PZ}	1.0/0	0.9/0.1	0.8/0.2	0.7/0.3	0.6/0.4
T = 288.15 K	1.0648	1.0599	1.0547	1.0481	1.0432
T = 303.15 K	1.0619	1.0579	1.0520	1.0452	1.0412
T = 313.15 K	1.0602	1.0561	1.0496	1.0435	1.0397
T = 323.15 K	1.0578	1.0540	1.0479	1.0421	1.0378

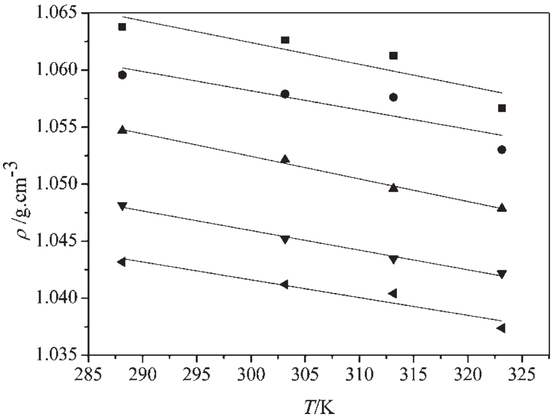

Figure 1. Density ρ of the (PG + PZ + H_2O) system as a function of temperature; ■, x_{PG}/x_{PZ} = 1.0/0; ●, x_{PG}/x_{PZ} = 0.9/0.1; ▲, x_{PG}/x_{PZ} = 0.8/0.2; ▼, x_{PG}/x_{PZ} = 0.7/0.3; left-pointing triangle, x_{PG}/x_{PZ} = 0.6/0.4, solid lines correlated from eq 2.

To validate this experimental procedure and data of the measurements, the densities, viscosities, and surface tensions of mass fractions of 10 % and 20 % MDEA aqueous solutions were measured at (288.15 and 313.15) K and compared with the values reported by Paul and Mandal[13] and Aguila-Hernández et al.[14] These are given in Table 1. The average absolute deviations of the measurements are 0.07 % for densities, 0.48 % for viscosities, and 0.51 % for surface tensions. Thus, the density data obtained in this work are in good agreement with the data of literature.[13,14]

Density. The densities for the aqueous solutions of (PG + PZ) were determined at (288.15, 303.15, 313.15, and 323.15) K with compositions of x_{PG}/x_{PZ} (mol/mol) = 1.0/0, 0.9/0.1, 0.8/0.2, 0.7/0.3, and 0.6/0.4. x_{PG} and x_{PZ} are the mole fractions of PG and PZ in the solution, respectively. The total concentration of the solutions of (PG + PZ) was 1.0 mol·L^{-1}. The data of the measured densities are presented in Table 2. As shown in Table 2, the densities of (PG + PZ) aqueous solutions decrease with an increase of temperature and with a decrease of mole fraction of PG in the mixture. As x_{PG}/x_{PZ} = 1.0/0, namely, aqueous solutions of PG, a deviation of 0.05 % is found when compared with the experimental data of Kumar et al.[15] The density results are consistent with other authors and compliant with general law.

The experimental data, expressed in a form density, ρ versus temperature T, are presented in Figure 1. The results show that the density is linear with respect to temperature. Also, it illustrates that, under a constant total concentration of solution, the density of the solution decreases as the amine is added into the amino acid salt. A correlation was developed to allow the prediction of the density for the aqueous solutions of (PG + PZ) as a function of temperature by means of linear fitting using the experimental results obtained in this work. The correlation uses the following equation,

$$\rho = a + b\cdot T \tag{2}$$

where ρ is the density in g·cm^{-3}, T is the temperature in K, and a and b are coefficients. The values of a and b and average absolute

Table 3. Values of *a* and *b* Coefficients for eq 2 and Average Absolute Deviation

x_{PG}/x_{PZ}	a	$10^4\ b$	R^2	AAD%
1.0/0	1.1219	−1.9682	0.9972	0.03
0.9/0.1	1.1084	−1.6766	0.9880	0.02
0.8/0.2	1.1117	−1.9776	0.9963	0.01
0.7/0.3	1.0977	−1.7252	0.9949	0.01
0.6/0.4	1.0873	−1.5271	0.9929	0.02

Table 4. Viscosities for ($PG + PZ + H_2O$) from (288.15 to 323.15) K

	η/mPa·s				
x_{PG}/x_{PZ}	1.0/0	0.9/0.1	0.8/0.2	0.7/0.3	0.6/0.4
T = 288.15 K	1.254	1.285	1.294	1.308	1.326
T = 303.15 K	0.964	0.971	0.974	0.991	0.989
T = 313.15 K	0.830	0.842	0.842	0.847	0.853
T = 323.15 K	0.747	0.750	0.750	0.753	0.753

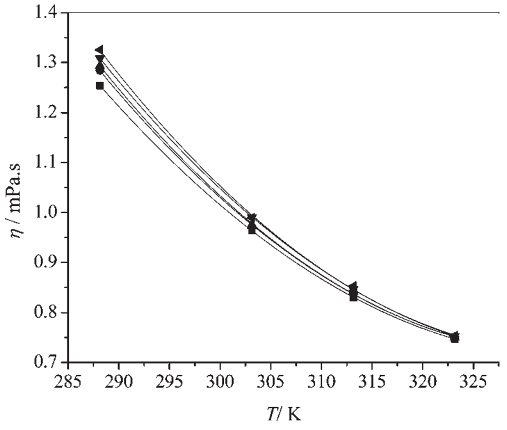

Figure 2. Viscosity η of the ($PG + PZ + H_2O$) system as a function of temperature; ■, x_{PG}/x_{PZ} = 1.0/0; ●, x_{PG}/x_{PZ} = 0.9/0.1; ▲, x_{PG}/x_{PZ} = 0.8/0.2; ▼, x_{PG}/x_{PZ} = 0.7/0.3; left-pointing triangle, x_{PG}/x_{PZ} = 0.6/0.4, solid lines correlated from eq 3.

deviations are given in Table 3. The maximal deviation is 0.08 % between the prediction and the experimental values.

Viscosity. The viscosities of the aqueous solutions of (PG + PZ) were measured at (288.15 to 323.15) K with compositions of x_{PG}/x_{PZ} =1.0/0, 0.9/0.1, 0.8/0.2, 0.7/0.3, and 0.6/0.4. The results of viscosity for the aqueous solutions of (PG + PZ) are presented in Table 4. It can be observed that, as expected, the viscosities for the aqueous solutions decrease with an increase of temperature and increase with an increase of PZ mole fraction in the solution.

The experimental data, expressed in the form viscosity η versus temperature T, are presented in Figure 2. The results show that the change of viscosity for the aqueous solutions of (PG + PZ) is larger at low temperatures than at high temperatures. Differences of the viscosity get smaller and smaller with the increase of PZ mole fraction in the solution. Furthermore, the higher the temperature, the phenomenon is more obvious. A correlation was developed to allow the prediction of the viscosity for the aqueous solutions of (PG + PZ) as a function of temperature by means of polynomial fitting using the experimental data obtained in this work. The following expression is used for the regression equation of the experimental data,

Table 5. Values of the *k*, *m*, and *n* Parameters in eq 3 and Average Absolute Deviation

x_{PG}/x_{PZ}	k	m	$10^4\ n$	R^2	AAD %
1.0/0	28.1670	−0.1638	2.4417	1	0.85
0.9/0.1	30.6755	−0.1794	2.6860	0.9994	0.34
0.8/0.2	31.5293	−0.1847	2.7685	0.9995	0.38
0.7/0.3	30.2565	−0.1759	2.6186	1	0.62
0.6/0.4	32.7456	−0.1918	2.8710	0.9991	0.96

Table 6. Surface Tension for ($PG + PZ + H_2O$) at 288.15 K

	γ/mN·m^{-1}				
x_{PG}/x_{PZ}	1.0/0	0.9/0.1	0.8/0.2	0.7/0.3	0.6/0.4
	76.16	75.21	74.62	74.06	73.50

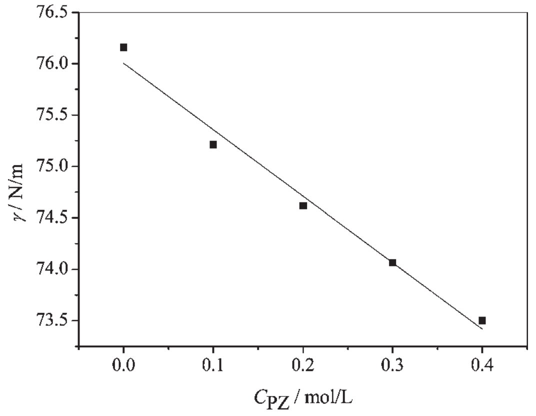

Figure 3. Surface tension γ of the ($PG + PZ + H_2O$) system as a function of PZ concentration; ■, PZ concentration; solid lines correlated from eq 4.

$$\eta = k + m \cdot T + n \cdot T^2 \tag{3}$$

where η is the viscosity in mPa·s, T is the temperature in K, and k, m, and n are parameters. The values of the parameters and average absolute deviations are given in Table 5. The average absolute deviation for viscosities is less than 0.60 %. For the small AAD values such as x_{PG}/x_{PZ} = 0.7/0.3 (AAD % = 0.06), the predictions from eq 3 are in excellent agreement with the experimental values.

Surface Tension. The surface tensions of the aqueous solutions of (PG + PZ) were determined at 288.15 K with compositions of x_{PG}/x_{PZ} = 1.0/0, 0.9/0.1, 0.8/0.2, 0.7/0.3, and 0.6/0.4. The results of surface tensions are presented in Table 6. It can be found that the surface tensions for the aqueous solutions decrease with an increase of PZ mole fraction in the solution.

The experimental data, expressed in the form surface tension γ versus PZ concentration C_{PZ}, are given in Figure 3. As shown in Figure 3, at the given temperature, the surface tensions of the aqueous solutions of (PG + PZ) almost decrease linearly as the

Table 7. Values of p and q Parameters for eq 4 and Average Absolute Deviation

p	q	R^2	AAD%
76.01	−6.468	0.9853	0.13

PZ concentration increases. Similarly, a correlation is developed to allow the prediction of the surface tension for the aqueous solutions of (PG + PZ) as a function of PZ concentration by means of fitting the experimental data obtained in this work to a linear equation. The regression equation is

$$\gamma = p + qC_{\mathrm{PZ}} \tag{4}$$

where γ is the surface tension in $\mathrm{mN \cdot m^{-1}}$, C_{PZ} is the PZ concentration in $\mathrm{mol \cdot L^{-1}}$, and p and q are parameters. The values of the parameters and average absolute deviations are listed in Table 7. Equation 4 correlates the surface tension values of the aqueous solutions with an AAD of 0.13 %, and as can be seen the agreement is acceptable. Figure 3 also shows the prediction values are in agreement with the experimental values over the whole concentration range.

CONCLUSIONS

The densities and viscosities of aqueous solutions of (PG + PZ) have been measured at (288.15 to 323.15) K, respectively. The surface tensions have been determined at 288.15 K. The experiments covered the compositions of $x_{\mathrm{PG}}/x_{\mathrm{PZ}}$ (mol/mol) = 1.0/0, 0.9/0.1, 0.8/0.2, 0.7/0.3, and 0.6/0.4. Correlations by means of mathematical fitting were developed for the prediction of the density, viscosity, and surface tension of the aqueous solutions of (PG + PZ) using the experimental data of this work. For a given concentration of the aqueous solutions, the densities and viscosities decrease as the temperature increases. The densities of the studied solutions decrease with an increase of PZ mole fraction in the solutions, whereas the viscosities of the solutions increase. The surface tensions of the solutions almost decrease linearly as the PZ mole fraction in the solution increases at 288.15 K. The prediction values from correlation equations for density, viscosity, and surface tension are in good agreement with the experimental values. The average deviations are small and acceptable.

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