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Physical properties of aqueous solutions of potassium carbonate+glycine as a solvent for carbon dioxide removal

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Abstract: The physical properties, such as densities, viscosities, and refractive indices, of aqueous solutions of potassium carbonate (PC) blended with glycine (Gly) as solvent blends for CO₂ capture were measured. The properties were measured at ten different temperatures from (298.15 to 343.15) K. The mass fractions (w_1+w_2) of the (PC+Gly) blends were 0.05+0.01, 0.10+0.02, 0.15+0.03, 0.20+0.04, 0.25+0.05, 0.30+0.06, 0.35+0.07 and 0.40+0.08 %, respectively. An analysis of the experimental results showed that the densities, viscosities, and refractive indices of the aqueous (PC+Gly) blends increase with increasing concentration of potassium carbonate and glycine, and decrease with increasing the temperature. The experimental data of density, viscosity and refractive index were correlated by a least-squares method as a function of temperature. The predicted data were estimated from coefficients of correlation (*SD*). The experimental data were consistent with the predicted data.

Keywords: density; viscosity; refractive index; potassium carbonate; glycine; CO₂ capture.

INTRODUCTION

The concern of researchers to overcome the climate change issue is increasing with increasing level of carbon dioxide (CO₂) in the atmosphere. At present, the concentration of CO₂ in the atmosphere is 396.80 ppm, which is more than the acceptable limit (350 ppm). The CO₂ concentration is expected to grow further above 400 ppm by 2015.^{1,2} The main causes of the increase in the CO₂ level are the rapid growth of industry and transport, mechanization of agricultural activities, the energy sector and domestic sources.^{3,4} The major reasons of the endeavors undertaken to overcome the CO₂ issues are the appalling effects of this

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greenhouse gas on the global environment and its inhabitants.⁵ In continuation of these endeavors; various technologies have been developed to mitigate CO₂ emission. These technologies include absorption, adsorption, membrane and cryogenic processes. These technological options are in operation to capture CO₂ but efforts are being undertaken to improve these techniques.^{6,7} The most developed and functioning technique is the absorption process by chemical solvents.⁸

The solvents that are very common and commercially proven are amine--based solvents, such as monoethanolamine, diethanolamine, triethanolamine, methyldiethanolamine and 2-amino-2-methyl-1-propanol, etc.9-11 These amine--based solvents have been widely used for years on the commercial scale, but some issues were identified and reported in the literature, including, the short life of the solvent due to thermal and oxidative degradation, corrosiveness, volatility, and high-energy consumption during regeneration.^{12–15} An aqueous solution of potassium carbonate has also been used as a CO₂ capture agent since it has a lower rate of degradation and lower cost as compared to amines.^{16,17} However, the use of this solvent is limited due to the low rate of CO_2 absorption. One of the promising solutions is the addition of suitable promoters in order to remove the acid gases effectively from various gas streams. A varieties of promoters are used nowadays with potassium carbonate, such as monoethanolamine, diethanolamine and piperazine, etc.^{17,18} Potassium carbonate promoted with these additives offers better CO₂ removal; however, the identified drawbacks of amines restrict the use of these promoters with potassium carbonate. The challenge is to search for the most appropriate promoter to overcome the drawback of amines. One of the potential promoters is an amino acid, such as sarcosine, which has been used for this purpose.¹⁸ This is due to their benefits of having very low degradation, negligible volatility, able to be regenerate, environmental friendly and commercially available.^{19–22} These prospective merits of amino acids have encouraged us to investigate another type of potential amino acid, *i.e.*, glycine as a promoter.

Physical properties, such as density, viscosity and refractive index, are crucial for the practical development, design and implementation of a solvent.^{23,24,35–37} Knowledge of the density and viscosity are a key requirement to establish rate modeling and to determine the constants of the reaction rate.²⁵ Refractive index data are important to calculate the molar refraction, which is useful for a detailed understanding of the molecular interaction of a solvent.^{26–28} In this study, the density, viscosity, and refractive index of aqueous solutions of potassium carbonate (PC) blended with glycine (Gly) were determined and are reported. The concentration variations of the blends were selected in order to investigate their impact on the CO₂ removal performance.

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EXPERIMENTAL

Materials

Potassium carbonate (\geq 99 % pure), glycine (\geq 99 % pure), and methyldiethanolamine (\geq 99 % pure) were purchased from Merck Sdn. Bhd, Malaysia. The additional information about the chemicals used in this study is specified in Table I. All the chemicals were used without further purification. Double distilled water was used to prepare all the solutions. The aqueous blends of potassium carbonate (PC) and glycine (Gly) were prepared in mass. An electronic analytical balance (Sartorius, model BSA-224S-CW) with the measurement accuracy of $\pm 1 \times 10^{-4}$ g was used. The different mass fractions (w_1+w_2) of the (PC+Gly) blends were 0.05+0.01, 0.10+0.02, 0.15+0.03, 0.20+0.04, 0.25+0.05, 0.30+0.06, 0.35+0.07 and 0.40+0.08 %. Furthermore, the uncertainty in the mass fraction was determined to be $\pm 1 \times 10^{-3}$. All the properties were measured within the temperature range of 298.15 to 343.15 K. The maximum mass fraction in the blend was kept at 0.40 for potassium carbonate, and 0.08 for glycine, which are as per the commercial suitability of the solvent.

TABLE I. Specifications of the chemicals

Name of chemical	Chemical formula	Purity	Method of purification	Source
Glycine	H ₂ NCH ₂ COOH	\geq 99 % pure	None	Merck
Potassium carbonate	K_2CO_3	\geq 99 % pure	None	Merck
Methyldiethanolamine	$CH_3N(C_2H_4OH)_2$	\geq 99 % pure	None	Merck
Double distilled water	H ₂ O	99 % pure	Distillation	-

Apparatus and procedure

The density of different aqueous (PC+Gly) blends was measured using a digital densimeter (Anton Par, model, DMA-4500M) with an accuracy of ±5×10⁻⁵ g·cm⁻³. The apparatus was calibrated each time before and after the measurement in order to obtain accurate results. Standard water of Millipore quality was used in the calibration process. Each experiment was performed in triplicate, and the data reported are the average values. The density and temperature uncertainty was $\pm 6 \times 10^{-5}$ g cm⁻³ and ± 0.01 K, respectively. A digital rolling ball microviscometer (Anton Par, model Lovis-2000M / ME) with an accuracy of up to 0.5 % was used to measure the viscosity of the aqueous (PC+Gly) blends. Before filling the sample in a suitable capillary, the capillary was properly washed with acetone, and air-dried to avoid any error in the reading. Before and after each experiment, the viscometer was carefully calibrated with Millipore water. For the measurement, the capillary was filled with the sample by the help of the syringe, kept inside the viscometer until the set temperature was achieved, and finally, the measurement was started. Each experiment was repeated three times, and the average values of the viscosity are reported. The uncertainties in the viscosity and temperature were estimated to be $\pm 7 \times 10^{-3}$ mPa·s and ± 0.02 K, respectively. For measurement of the refractive index of the aqueous blends of (PC+Gly), a digital Abbemat automatic refractometer (Anton Par, model WR), with an accuracy of $\pm 4 \times 10^{-5} n_{\rm D}$, was used. In order to obtain accurate readings, the refractometer was calibrated with water of Millipore quality each time the sample was changed. Before pouring the sample into the sample mould, the prism face was carefully cleaned with acetone and dried to prevent any disturbance in the results because of possible minute sediments on the prism face. Measurement was started after pouring the sample into the sample mould and setting the required temperature. Each experiment was

conducted three times and the average value is reported. The uncertainties in the refractive index and temperature were $\pm 5 \times 10^{-5} n_{\rm D}$ and ± 0.03 K, respectively.

RESULTS AND DISCUSSION

The experimental results for pure methyldiethanolamine (MDEA) were compared with the corresponding literature data in order to validate the results. The comparison results for density, viscosity and refractive index of pure MDEA are given in Table II. The % average absolute deviations (% *AAD*) reported in Table II were calculated using Eq. (1):³²

%
$$AAD = \frac{1}{n} \sum \left| \frac{X_{\exp} - X_{\text{lit}}}{X_{\text{lit}}} \right| \times 100$$
 (1)

where X_{exp} is experimental values of density, ρ , viscosity, η , and refractive index, n_D , whereas X_{lit} denotes the literature data of density, viscosity and refractive index, and n is the number of data points. The deviation values show that there was good consistency of experimental and literature data.

TABLE II. Comparison of experimental data of density, ρ , viscosity, η , and refractive index, n_D , of pure methyl diethanolamine with literature data

<i>T</i> / K	Present work	Literature data ³⁴	<i>AAD</i> / %
		o∕g·cm ⁻³	
303.15	1.03356	1.03325	0.02085
313.15	1.02553	1.02565	
		η∕mPa s	
303.15	59.756	59.76	0.021
313.15	37.886	37.9	
		n _D	
303.15	1.46728	1.46796	0.03716
313.15	1.46397	1.46438	

The measured values of the density of the aqueous blends of (PC+Gly) at various temperatures from 298.15 to 343.15 K are presented in Table III. It was found that with increasing mass fraction of potassium carbonate and glycine in the blend, the density increased; however, the density decreased with increasing temperature. This could be due to the wider spaces between the blend molecules at higher temperatures.²³ This density trend is similar to that previously reported work.^{29,30}

The investigated data for the viscosity of different concentrations of aqueous (PC+Gly) blends in the temperature range of 298.15 to 343.15 K are listed in Table IV. After analysis of results, it was noticed that the viscosity decreased with increasing temperature. This could be due to a decrease in the internal resistance of the molecules with increasing temperature, which allows the solution molecules to flow easily, thereby reducing the viscosity.^{30,31} However, with

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increasing concentration of potassium carbonate and glycine in the aqueous solutions, the viscosity tended to increase. The higher concentrated solutions had a higher viscosity than the lower ones, which may be due to the increased molecular resistance in the more concentrated solutions.^{30,31} The trends of the variation in viscosity values with changing concentration and temperature were the same as those found in published studies.^{30,31}

TABLE III. Density, ρ / g cm⁻³, of aqueous blends of potassium carbonate (1)+ glycine(2)

TK				(w ₁ +и	/ %			
1 / K	0.05 + 0.01	0.10 + 0.02	0.15 + 0.03	0.20 + 0.04	0.25 + 0.05	0.30 + 0.06	0.35 + 0.07	0.40 + 0.08
298.15	1.04219	1.08767	1.13833	1.18964	1.24263	1.29839	1.35564	1.41943
303.15	1.04044	1.08565	1.13608	1.18721	1.24006	1.29570	1.35286	1.41656
308.15	1.03854	1.08351	1.13373	1.18470	1.23743	1.29298	1.35005	1.41367
313.15	1.03648	1.08125	1.13129	1.18213	1.23475	1.29021	1.34721	1.41078
318.15	1.03427	1.07887	1.12876	1.17948	1.23200	1.28739	1.34433	1.40784
323.15	1.03193	1.07637	1.12614	1.17675	1.22919	1.28451	1.34140	1.40487
328.15	1.02944	1.07377	1.12342	1.17395	1.22632	1.28158	1.33843	1.40185
333.15	1.02684	1.07108	1.12062	1.17109	1.22340	1.27862	1.33541	1.39880
338.15	1.02410	1.06827	1.11774	1.16816	1.22042	1.27561	1.33237	1.39573
343.15	1.02126	1.06537	1.11478	1.16517	1.21739	1.27256	1.32930	1.39265

TABLE IV. Viscosity, η / mPa s, of aqueous blends of potassium carbonate (1)+glycine (2)

T / K				$(w_1 + w_1)$	v ₂) / %			
1 / K	0.05 + 0.01	0.10 + 0.02	0.15 + 0.03	0.20 + 0.04	0.25 + 0.05	0.30 + 0.06	0.35 + 0.07	0.40 + 0.08
298.15	0.993	1.164	1.372	1.590	2.014	2.567	3.541	5.010
303.15	0.891	1.048	1.236	1.431	1.788	2.292	3.128	4.387
308.15	0.806	0.954	1.120	1.297	1.613	2.063	2.813	3.879
313.15	0.733	0.869	1.021	1.183	1.458	1.870	2.540	3.464
318.15	0.672	0.795	0.937	1.083	1.331	1.705	2.306	3.111
323.15	0.619	0.733	0.863	1.005	1.222	1.562	2.103	2.816
328.15	0.572	0.682	0.799	0.923	1.112	1.439	1.931	2.583
333.15	0.532	0.634	0.743	0.857	1.045	1.331	1.780	2.367
338.15	0.496	0.594	0.693	0.800	0.972	1.237	1.648	2.179
343.15	0.464	0.555	0.648	0.749	0.911	1.152	1.531	2.017

The experimental data for the refractive index of the aqueous solutions of (PC+Gly) in the temperature range 298.15 to 343.15 K are reported in Table V. From experimental values, it was observed that the refractive index increased with increasing concentration of the blends but decreased slightly with increasing temperature. The decrease in refractive index with increasing temperature could be due to an increase in the speed of the particles in the aqueous (PC+Gly) blends, causing the light to strike fewer molecules, thereby reducing the refractive index. In case of concentration, since additional molecules were present in the solution, the chances of light striking the molecules was greater, thereby increasing the refractive index.³⁸ The manner in which the refractive index

decreased with temperature and increased with increasing concentration was same as reported in the literature.^{32,33}

TABLE V. Refractive index, n_D , of aqueous blends of potassium carbonate (1)+glycine (2)

T/V				(w_1+w_1)	/2) / %			
1 / K	0.05 + 0.01	0.10 + 0.02	0.15 + 0.03	0.20 + 0.04	0.25 + 0.05	0.30 + 0.06	0.35 + 0.07	0.40 + 0.08
298.15	1.34158	1.35042	1.36001	1.36994	1.37893	1.38850	1.39801	1.40815
303.15	1.34091	1.34969	1.35921	1.36861	1.37817	1.38763	1.39714	1.40727
308.15	1.34020	1.34892	1.35843	1.36788	1.37733	1.38674	1.39626	1.40639
313.15	1.33951	1.34819	1.35776	1.36710	1.37654	1.38596	1.39542	1.40553
318.15	1.33876	1.34754	1.35717	1.36660	1.37592	1.38528	1.39464	1.40470
323.15	1.33800	1.34667	1.35680	1.36636	1.37540	1.38468	1.39402	1.40399
328.15	1.33727	1.34597	1.35657	1.36596	1.37495	1.38430	1.39361	1.40334
333.15	1.33662	1.34511	1.35601	1.36557	1.37453	1.38398	1.39323	1.40288
338.15	1.33602	1.34421	1.35583	1.36550	1.37445	1.38355	1.39311	1.40277
343.15	1.33546	1.34347	1.35536	1.36541	1.37421	1.38302	1.39282	1.40270

Experimentally measured data for density were converted into graphical form with respect to temperature, and based on various series of concentrations, the best fit was found by the least-squares method. The following equation was used to report the correlation coefficients for density:

$$\rho = P_0 + P_1(T / K) + P_2(T / K)^2$$
(2)

where ρ represents the density, P_0 , P_1 and P_2 are the optimized correlation parameters and *T* is the temperature. These fitting parameters are listed in Table VI along with the *SD* calculated using Eq. (3), as also mentioned in literature:²³

$$SD = \left[\frac{\sum_{i=1}^{n} (X_{\exp,i} - X_{calc}, i)^2}{n} \right]^{0.5}$$
3)

where *SD* is the standard deviation, X_{exp} is the experimental value of the density, ρ , viscosity, η , or refractive index, n_D , X_{cal} is the calculated value of the same property, and *n* is the number of data points.

Measured viscosity data was transformed into graphical form with respect to the temperature. The fittings were conducted by the least- squares method using an exponential function. The following equation was used to correlate the viscosity data:

$$\eta = Q_0 \exp\left(-Q_1 T / K\right) \tag{4}$$

where η is the viscosity, Q_0 and Q_1 are the optimized coefficients, and *T* is the temperature. The optimized parameters of the correlation equation are reported in Table VII with the SD calculated using Eq. (3).

TABLE VI. Correlation Eq. (2) parameters and SD for the density, $\rho / g \cdot cm^{-3}$, of aqueous blends of potassium carbonate (1)+glycine (2)

$(w_1+w_2) / \%$	P_0	$10^4 P_1$	$10^{6}P_{2}$	R^2	$10^3 SD$
0.05+0.01	0.89973	12.982	-2.751	0.999	0.075
0.10 + 0.02	1.01042	9.155	-2.201	0.999	0.013
0.15 + 0.03	1.11189	6.208	-1.784	0.999	0.073
0.20 + 0.04	1.20693	3.643	-1.416	0.999	0.006
0.25 + 0.05	1.29024	1.890	-1.169	0.999	0.009
0.30 + 0.06	1.37268	0.331	-0.946	0.999	0.008
0.35 + 0.07	1.44956	-0.801	-0.787	0.999	0.009
0.40 + 0.08	1.53266	-1.928	-0.627	0.999	0.012

TABLE VII. Correlation Eq. (3) parameters and SD for the viscosity, η / mPa s, of aqueous blends of potassium carbonate (1)+glycine (2)

${(w_1+w_2)/\%}$	00	O_1	R^2	SD
0.05+0.01	143.793	-0.016	0.992	0.014
0.10 + 0.02	146.897	-0.016	0.994	0.015
0.15+0.03	185.701	-0.016	0.994	0.017
0.20+0.04	221.242	-0.016	0.992	0.050
0.25+0.05	361.613	-0.017	0.995	0.033
0.30+0.06	483.578	-0.017	0.994	0.036
0.35 + 0.07	837.945	-0.018	0.994	0.053
0.40+0.08	1884.389	-0.020	0.994	0.092

TABLE VIII. Correlation Eq. (2) parameters and *SD* for refractive index, n_D , of aqueous blends of potassium carbonate (1)+glycine (2)+water (3)

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$(w_1+w_2) / \%$	P_0	$10^4 P_1$	$10^{6}P_{2}$	R^2	$10^{2}SD$
0.05+0.01	1.41254	-3.236	0.287	0.999	0.005
0.10 + 0.02	1.36221	0.602	-0.334	0.999	0.004
0.15+0.03	1.52877	-9.723	1.362	0.995	0.010
0.20 + 0.04	1.64353	-16.343	2.401	0.992	0.012
0.25 + 0.05	1.60404	-13.181	1.889	0.998	0.005
0.30+0.06	1.58081	-11.031	1.536	0.997	0.009
0.35 + 0.07	1.65388	-15.021	2.160	0.998	0.006
0.40+0.08	1.67178	-15.405	2.203	0.996	0.011

Likewise, the experimentally measured refractive index data were plotted in the form of its relationship *vs*. temperature followed by the series of different concentrations of (PC+Gly) blends. In order to predict the refractive index data, the same correlation Eq. (2) was used as for the density prediction. The parameters of the refractive index fitting equation are presented in Table VIII together with the *SD* estimated using Eq. (3).

After the analysis of the predicted density, viscosity, and refractive index data obtained from the correlation equations, it was observed that the predicted data were in good agreement with the experimental data.

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CONCLUSIONS

The physical properties of aqueous (PC+Gly) blends such as density, viscosity, and refractive index were measured at a range of temperature from 298.15 to 343.15 K. The measured properties were observed to increase with increasing concentration of potassium carbonate and glycine in the solution. However, all properties tended to decrease with increasing temperature. The same trend was reported in the available literature. All experimental data were correlated by least-squares fitting to mathematical equations in order to calculate the predicted data. Based on the deviations calculated between the experimental and predicted data, good agreement was found. Hence, the developed correlations are acceptable and could be used in design calculations of future CO_2 removal systems.

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ИЗВОД

ФИЗИЧКА СВОЈСТВА ВОДЕНИХ РАСТВОРА КАЛИЈУМ-КАРБОНАТА СА ГЛИЦИНОМ КАО РАСТВАРАЧА ЗА УКЛАЊАЊЕ УГЉЕН-ДИОКСИДА

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Мерена су физичка својства, као што су густине, вискозности и индекси рефракције, водених раствора калијум-карбоната (PC) помешаног са глицином (Gly), као растварача за уклањање CO₂. Ова својства су мерена на 10 различитих температура, од 298,15 то 343,15 К. Масени удели (w_1+w_2) раствора (PC+Gly) су били 0,05+0,01, 0,10+0,02, 0,15+0,03, 0,20+0,04, 0,25+0,05, 0,30+0,06, 0,35+0,07 и 0,40+0,08 %, редом. Анализа експерименталних резулата показује да густине, вискозности и индекси рефракције водених (PC+Gly) смеша расту са порастом концентрација калијумкарбоната и глицина, и опадају са смањењем температуре. Експериментални подаци за густине, вискозности и индексе рефракције су корелисани као функције температуре, коришћењем методе најмањих квадрата. Предикције података су одређене на основу коефицијената корелација за сва мерена својства, и приказане са стандардном девијацијом. Експериментални подаци су конзистентни са предикцијама.

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