



Density, viscosity and excess properties for binary system of 1,2-ethanediamine + polyethylene glycol 400 at $T = (293.15, 298.15, 303.15, 308.15, 313.15, \text{ and } 318.15)$ K under atmospheric pressure

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ABSTRACT

Density (ρ) and viscosity (η) data for the binary system of polyethylene glycol 400 (PEG) + 1,2-ethanediamine (EDA) over the whole concentration range as a function of composition were measured at $T = (293.15, 298.15, 303.15, 308.15, 313.15, \text{ and } 318.15)$ K under atmospheric pressure. Based on experimental density and viscosity data, the excess properties of the PEG (1) + EDA (2) mixtures, including excess molar volume (V_m^E) and viscosity deviation ($\Delta\eta$), excess free energies of activation (ΔG^{*E}), apparent molar volume ($V_{\phi,1}$ and $V_{\phi,2}$), and partial molar volumes (\bar{V}_1 and \bar{V}_2), were calculated respectively. Meanwhile, the results of V_m^E , $\Delta\eta$ and ΔG^{*E} were fitted by a Redlich–Kister equation to obtain coefficients and to evaluate the standard deviations (σ) between the experimental and calculated quantities. Additionally, based on FTIR and UV–Vis spectral results of PEG (1) + EDA (2) mixtures with various concentrations, intermolecular interaction of PEG with EDA was discussed.

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1. Introduction

One of the most disturbing global environmental problems is global warming and climate change [1]. The increasing carbon dioxide (CO_2) concentration due to burning fossil fuels (coal, petroleum and natural gas) was the main cause of the problem. The CO_2 's contribution to global warming has exceed 60% [2,3], so it is necessary to develop the CO_2 capture technology.

Recently, EDA showed a strong capability to absorb CO_2 with about 0.78 g CO_2 per 1.00 g EDA [4]. However, EDA is not wildly used because EDA with relatively high volatility can produce several negative effects in a gas treating unit, including higher operating cost and higher capital expenditures caused by the loss of solvent. Sengwa [5] reported the hydrogen bonding interactions and hydrogen bonded structures in amine–alcohol mixed solvents. Meanwhile, the previous studies [6–9] showed that there was hydrogen bonding interactions in the mixtures of ethylene glycol (EG) and its similar compounds with EDA, which might be used to decrease the loss of amines. In the previous studies [10–13], EG, diethylene glycol (DEG), triethylene glycol (TEG), and polyethylene glycol 300 (PEG 300) were added into EDA and received a good effect for restricting the volatilization of amines.

In this work, polyethylene glycol 400 (PEG) was added into EDA to reduce the solvent's loss and improve the absorption performance of CO_2 . Physicochemical properties of absorbents including density (ρ), viscosity (η) and excess properties over a wide temperature range are

necessary for detailed characterization of the solvent for industrial application and in the process modeling, simulation and design of gas–liquid contactor columns for CO_2 absorption and regeneration processes. However, these data were lacking in the basic data, so we had to determine these data for the system PEG (1) + EDA (2). In addition, the possible intermolecular interaction of PEG with EDA was investigated using FTIR and UV spectroscopic techniques. As part of the systematic studies on application of EDA in CO_2 capture processes, the results of this work may be used to provide important basic data for the fixation of EDA with potential industrial application of the binary system PEG (1) + EDA (2).

2. Experimental section

2.1. Materials

The analytical grade EDA and PEG were purchased from Tianjin Reagent Company. EDA was dehydrated by Na_2SO_4 and distilled method. The purity of final PEG, as found by a gas chromatograph (GC), was better than 99.4%. Meanwhile, EDA was purified from EDA (98.0%) dried over molecular sieves (type 4A) and degassed by ultrasound just before the experiment. The purity of the final EDA, as found by the GC, is better than 99.3%. The purity of PEG and EDA was checked by measuring and comparing the density and viscosity of the samples with corresponding literature values in the temperature range of (293.15 to 318.15) K. Complete specification of chemical samples is listed in Table 1. The density and viscosity of EDA at 293.15 K were found to be $0.9002 \text{ g}\cdot\text{cm}^{-3}$ and $1.60 \text{ mPa}\cdot\text{s}$, and the density and viscosity of PEG at 293.15 K were found to be $1.1266 \text{ g}\cdot\text{cm}^{-3}$ and $113 \text{ mPa}\cdot\text{s}$ respectively. Solvent mixtures

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Table 1
Specification of chemical samples.

Chemical name	Source ^a	Initial mass fraction purity	Purification method
1,2-Diaminoethane	Tianjin Beilian Reagent Co., Ltd., China	≥99.0%	Desiccation ^b and Degasification ^c
Polyethylene glycol 400	Tianjin Beilian Reagent Co., Ltd., China	≥99.0%	Desiccation ^b and Degasification ^c
Ethanol ^d	Beijing Tongguang Industry of Fine Chemicals Co., Ltd., China	≥99.7%	

^a Declared by the supplier.^b Molecular sieve type 4A.^c Ultrasound.^d Chromatographic grade.

were prepared by mass using an analytical balance, which showed a precision of 0.01%, and the uncertainty in the mole fraction for each binary mixture is lower than 0.01%. FTIR spectra were recorded on a Nicolet (Nexus 670) FTIR spectrometer with a resolution of 1 cm^{-1} in the range from 4000 to 400 cm^{-1} . The spectrometer possesses auto-align energy optimization and a dynamically aligned interferometer and is fitted with two constraining BaF₂ pellets for the measurement of aqueous solution. A baseline correction was made for the spectra recorded in air; and then 10 μL solutions were used to perform on the FTIR spectrometer in each measurement and the thickness of sample layers was less than a typical thickness of 2 μm . All spectral experiments of binary mixture PEG (1) + EDA (2) were performed at room temperature and atmospheric pressure.

UV spectra were recorded on a Shimadzu (UV-2450) UV-Vis spectrometer with a resolution of 0.5 nm in the range of (190 to 700) nm at room temperature. The deionized water could be used to make a baseline correction for the spectra because the $n \rightarrow \sigma^*$ electronic transition of oxygen atom in water is often found at the vacuum ultraviolet region.

2.2. Measurements

Densities of pure liquids and their mixtures were determined at $T = (293.15, 298.15, 303.15, 308.15, 313.15, \text{ and } 318.15)\text{ K}$. The volume of the pycnometer was calibrated as a function of temperature using distilled, deionizer, and degassed water. A thermostatically controlled and well-stirred water bath, which has a controlled accuracy of 0.01 K was used to measure all density and viscosity data. The pycnometer filled with liquid was kept in water bath for 25 min to attain thermal equilibrium. Each experimental density value was an average of at least three measurements; furthermore, the uncertainty of density measurements was estimated to be $u(\rho) = 0.2\%$. The viscosity in both the pure liquids and their mixtures were performed using an Ubbelohde type with a commercial capillary viscometer, which had a capillary diameter of 1.1 mm. In the whole experiment process, only one viscometer was used. The Ubbelohde type was calibrated with bidistilled water and ethanol at $T = (293.15, 298.15, 303.15, 308.15, \text{ and } 313.15, 318.15)$

K whose viscosity and density were well-known in the literatures. The flow time was determined with a hand-held digital stopwatch capable of measuring time within $\pm 0.01\text{ s}$. The average of at least 18 flow times for each fluid was taken for the calculation of viscosities. Care was taken to reduce evaporation during the measurements.

The kinematic viscosity (ν) was calculated from the following equation:

$$\nu = At - B/t \quad (1)$$

where t is flow time of liquids in the viscometer; and A and B are instrument constants calculated from measurements with the calibration fluids of water and ethanol. The absolute viscosity was calculated by multiplying the kinematic viscosity by the corresponding density.

The measured density and viscosity data of EDA and PEG were in good agreement with those in literatures at the experimental temperatures as listed in Table 2 [14–25].

3. Results and discussion

3.1. Density

The experiment measured density for the binary system of EDA + PEG were listed in Table 3 and plotted in Fig. 1. As shown in Fig. 1, the density values of PEG (1) + EDA (2) mixtures decreased with the increasing concentrations of PEG at the same temperature; meanwhile, the density values gradually decreased with the increasing temperatures at the same concentration.

The calculated density values ($\rho_{\text{cal.}}$) could be expressed as Eq. (2) [26–27], which relied on temperatures, were matched by a second-order polynomial and presented in Eq.(2), which confirmed the correlation between density and temperature.

$$\rho_{\text{cal.}} = a_0 + a_1T + a_2T^2 \quad (2)$$

where a_i are the undetermined parameters and residual gas constant, T is absolute temperature of mixtures.

Table 2
Density and viscosity values of pure solutions at different temperatures, and comparison with literature data.

T/K	EDA				PEG			
	$\rho/(\text{g}\cdot\text{cm}^{-3})$		$\eta/(\text{mPa}\cdot\text{s})$		$\rho/(\text{g}\cdot\text{cm}^{-3})$		$\eta/(\text{mPa}\cdot\text{s})$	
	Exp.	Lit.	Exp.	Lit.	Exp.	Lit.	Exp.	Lit.
293.15	0.9002	0.8897 [14] 0.8994 [15]	1.601	1.603 [14]	1.1266	1.1262 [20]	112	115 [20]
298.15	0.8957	0.8952 [19] 0.8948 [16]	1.417	1.405 [18] 1.435 [17]	1.1219	1.1223 [21] 1.1218 [22]	79.82	92.79 [22] 90.701 [23]
303.15	0.8902	0.8906 [16] 0.8908 [17]	1.32	1.29 [19] 1.30 [17]	1.1201	1.1197 [23]	73.2	69.1 [24]
308.15	0.8854	0.8856 [21] 0.8855 [19]	1.16	1.167 [19] 1.191 [17]	1.1153	1.1148 [25]	56.3	51.66 [25]
313.15	0.8807	0.8809 [19] 0.8814 [17]	1.06	1.060 [19]	1.1113	1.1115 [24]	43.03	44.4 [24]
318.15	0.8769	0.8767 [17]	0.97	0.98 [17] 0.96 [19]	1.1063	1.1067 [25]	35.6	33.81 [25]

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