

Excess properties and viscous flow thermodynamics of the binary system 1,2-ethanediamine+triethylene glycol at T=(298.15, 303.15, 308.15, and 313.15) K for CO₂ capture

Yan Chang, Jianbin Zhang[†], Qiang Li, Lihua Li, Bo Guo, and Tianxiang Zhao

College of Chemical Engineering, Inner Mongolia University of Technology, Huhhot 010051, China
(Received 2 February 2014 • accepted 1 July 2014)

Abstract—Liquid densities and viscosities are reported for the binary system of 1,2-ethanediamine (EDA)+triethylene glycol (TEG) at T=(298.15, 303.15, 308.15, and 313.15) K. Densities were measured using a capillary pycnometer and viscosities were determined using an Ubbelohde capillary viscometer. The experimental results are compared with data published in the literatures. Based on the density data and kinematic viscosity data, excess molar volumes (V_m^E) and deviation in kinematic viscosity ($\Delta\nu$) were calculated and the calculated results were fitted to a Redlich-Kister equation to obtain the coefficients and estimate the standard deviations between the experimental and calculated quantities. The values of V_m^E are negative in the whole composition range, whereas the values of $\Delta\nu$ are positive over the major composition range. From kinematic viscosity data, Gibbs energies of activation of viscous flow (ΔG^*), enthalpy of activation for viscous flow (ΔH^*), and entropy of activation for the viscous flow (ΔS^*) were also calculated.

Keywords: Density, Viscosity, Excess Molar Volume, Viscous Flow Thermodynamics

INTRODUCTION

The concentration of atmospheric carbon dioxide (CO₂), which is mainly due to CO₂ emission from the combustion of fossil fuels, gas flaring, and cement production, has substantially increased [1]. Meanwhile, the Intergovernmental Panel on Climate Change (IPCC) has proposed CO₂ capture and storage (CCS) [2,3] as a sustainable technology to mitigate greenhouse gas emissions. Therefore, it is an exigent necessity to develop the CO₂ abatement techniques [4].

According to the principle of CO₂ separation, chemical absorption is considered to be the most popular method among these technologies, which includes membranes, adsorption, cryogenics, and absorption. Some of the most important typical chemical absorbents include monoethanolamine (MEA) [5-7], diethanolamine (DEA) [8], triethanolamine (TEA) [9], diisopropanolamine (DIPA) [10], N-methyldiethanolamine (MEDA) [5,10], diglycolamine [10,11], and 1,2-diaminoethane [12]. However, the costs of the amine-based absorption processes are relatively high due to volatilization of amines, so that they cost large amounts of energy consumption [13] in the solvent regeneration processes.

In our recent work, 1,2-ethanediamine (EDA) shows stronger solubility capability for CO₂; however, EDA is also a high volatile absorbent in the absorption and desorption processes of CO₂; therefore, triethylene glycol (TEG) is used as a fixing reagent to reduce the volatilization of EDA aqueous. Meanwhile, knowledge of the density and viscosity data is an indispensable requirement for the absorption and desorption processes of CO₂. But the density and viscosity data are very lacking in the basic data, so we have to determine the density and viscosity data for the system EDA+TEG

system for the future industrial application of the solutions containing EDA.

The present work is mainly focused on investigating the density data and viscosity data of EDA+TEG for the whole composition range at T=(298.15, 303.15, 308.15, and 313.15) K. Based on experimental results, the excess molar volumes (V_m^E), deviations in kinematic viscosity ($\Delta\nu$), Gibbs energies of activation for viscous flow (ΔG^*), enthalpy of activation for viscous flow (ΔH^*), and entropy of activation for the viscous flow (ΔS^*) were calculated.

EXPERIMENTAL

1. Materials

Analytical grade EDA and TEG were purchased from Beijing Reagent Company (Beijing, China). The chemicals were dried over molecular sieves (type 4A) and degassed by ultrasonic method before use. The purity of final TEG, as found by gas chromatograph (GC), was better than 99.3%. The purity of the sample was checked by density and kinematic viscosity determination at T=298.15 K. The density of EDA at T=298.15 K was found to be 0.8945 g·cm⁻³, and the density of TEG at T=298.15 K was found to be 1.1200 g·cm⁻³, which were in good agreement with literatures [14-19]. Bi-distilled water and high-performance liquid chromatography (HPLC) grade ethanol (Tianjin Kernel Chemicals Co., Ltd., China) were used. All specification of chemical samples is listed in Table 1.

2. Measurements

Solvent mixtures were prepared by mass using an analytical balance with a precision of 0.0001 g (Sartorius BS 224S). The uncertainty in the mole fraction for each binary mixture was estimated to be ± 0.0001 .

Densities of pure liquids and their mixtures were determined using a bicapillary pycnometer having a bulb volume of 10 cm³. The volume of the pycnometer was calibrated as a function of temperature

[†]To whom correspondence should be addressed.

E-mail: tadzhang@pku.edu.cn

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Table 1. Specification of chemical samples

Chemical name	Source ^b	Initial mass fraction purity	Purification method
1,2-Ethanediamine	Tianjin Reagent Co., Ltd., China	≥0.99	Desiccation ^c and Degasification ^d
Tri-ethylene glycol	Beijing Reagent Co., Ltd., China	≥0.99	Desiccation ^c and Degasification ^d
Ethanol ^a	Beijing Tongguang Industry of Fine Chemicals Co., Ltd., China	≥0.997	Desiccation ^c and Degasification ^d

^aChromatographic grade^bDeclared by the supplier^cMolecular sieve type 4A^dUltrasound

using distilled, deionized, and degassed water at four temperatures of T=(298.15, 303.15, 308.15 and 313.15) K. A thermostatically controlled and well-stirred water bath, which was controlled to ±0.01 K, was used for all the density and kinematic viscosity measurements. The pycnometer filled with liquid was kept in the water bath for 25 min to attain thermal equilibrium. Each experimental density value was an average of at least three measurements. The uncertainty of density measurement was estimated to be ±0.0002 g·cm⁻³.

The kinematic viscosities in both the pure components and their mixtures were measured with a commercial capillary viscometer

of the Ubbelohde type, which was calibrated with bi-distilled water and HPLC grade ethanol at T=(298.15, 303.15, 308.15 and 313.15) K. The flow time was determined with a hand-held digital stopwatch capable of measuring time within ±0.01 s. The average of sixteen flow times for each fluid was taken for the purpose of the calculation of viscosities.

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

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

where ν denotes the kinematic viscosity; t denotes flow time of liquids; and A and B denote viscometer constants, respectively. A and B are calculated from measurements with the calibration fluids. The absolute viscosity (η) was obtained by multiplying the determined ν by the measured density ($\eta = \nu\rho$). The uncertainty of kinematic viscosity measurement was estimated to be lower than ±0.3%.

The experimental densities and viscosities of pure EDA and TEG compared with the available literature values are given in Table 2.

Table 2. Comparison of experimental densities (ρ) and viscosities (η) of TEG and EDA with literature values at various temperatures^a

T/K	$\rho/(\text{g} \cdot \text{cm}^{-3})$		$\eta/(\text{mPa} \cdot \text{s})$	
	Expt.	Lit.	Expt.	Lit.
Triethylene glycol				
298.15	1.1200	1.1192 [14]	37.1	35.8 [14]
		1.11984 [15]		37.26 [22]
		1.11991 [16]		
303.15	1.1162	1.1151 [14]	29.2	29.256 [23]
		1.11588 [20]		29.224 [21]
		1.11585 [16]		
308.15	1.1118	1.1118 [14]	21.6	21.308 [24]
		1.11196 [16]		21.306 [25]
313.15	1.1087	1.1081 [14]	17.3	18.3 [14]
		1.10802 [16]		19.8 [26]
		1.1080 [21]		
1,2-Ethanediamine				
298.15	0.8945	0.8945 [17]	1.30	1.276 [17]
		0.8948 [18]		
		0.8952 [19]		
303.15	0.8881	0.8908 [17]	1.21	1.211 [17]
		0.890325 [27]		1.290 [27]
		0.8906 [19]		
308.15	0.8833	0.8856 [17]	1.07	1.107 [17]
		0.885616 [27]		1.167 [27]
		0.8859 [19]		
313.15	0.8797	0.8809 [27]	0.97	1.060 [27]

^aStandard uncertainties u are $u(T)=0.01$ K, $u(\rho)=0.0002$ g·cm⁻³ and $u(\eta)=\pm 0.3\%$

Table 3. Experimental densities (ρ) of EDA (1)+TEG (2) at T=(298.15 to 313.15) K^a

x_1	$\rho/(g \cdot cm^{-3})$			
	T/K=298.15	T/K=303.15	T/K=308.15	T/K=313.15
0.0000	1.1200	1.1162	1.1118	1.1087
0.1163	1.1108	1.1071	1.1031	1.1004
0.2174	1.1011	1.0974	1.0930	1.0901
0.3061	1.0909	1.0872	1.0835	1.0805
0.3846	1.0795	1.0765	1.0725	1.0695
0.4545	1.0687	1.0647	1.0607	1.0575
0.5172	1.0569	1.0528	1.0491	1.0460
0.5738	1.0443	1.0407	1.0370	1.0340
0.625	1.0323	1.0279	1.0237	1.0209
0.6716	1.0198	1.0156	1.0112	1.0080
0.7142	1.0073	1.0026	0.9985	0.9954
0.7534	0.9956	0.9911	0.9866	0.9834
0.7895	0.9834	0.9787	0.9744	0.9714
0.8228	0.9705	0.9658	0.9613	0.9578
0.9574	0.9161	0.9101	0.9053	0.9019
1.0000	0.8945	0.8881	0.8833	0.8797

^aStandard uncertainties u are $u(x_1)=0.0001$, $u(T)=0.01$ K and $u(\rho)=0.0002$ g·cm⁻³

RESULTS AND DISCUSSION

Experimental densities of EDA+TEG at T=(298.15, 303.15, 308.15 and 313.15) K throughout the whole concentration range are listed in Table 3.

Table 3 show that the density values decrease with the increasing EDA concentration in binary solutions over the whole concentration range. Meanwhile, the density values decrease with the augment of temperatures at the same composition.

The excess molar volume, V_m^E , was calculated from density measurements according to the following equation:

$$V_m^E = \frac{x_1 M_1 + x_2 M_2}{\rho_m} - \left(x_1 \frac{M_1}{\rho_1} + x_2 \frac{M_2}{\rho_2} \right) \quad (2)$$

where ρ_m denotes the density of the mixture and x_1 , ρ_1 , M_1 , x_2 , ρ_2 , and M_2 denote the mole fractions, densities, and molecular weights

Table 4. Excess molar volumes (V_m^E) for EDA (1)+TEG (2) at T=(298.15 to 313.15) K

x_1	$V_m^E \times 10^{-6} / (\text{m}^3 \cdot \text{mol}^{-1})$			
	T/K=298.15	T/K=303.15	T/K=308.15	T/K=313.15
0.0000	0.0000	0.0000	0.0000	0.0000
0.1163	-0.5402	-0.5796	-0.6354	-0.6908
0.2174	-0.9396	-1.0022	-1.0198	-1.0580
0.3061	-1.2208	-1.3033	-1.4003	-1.4334
0.3846	-1.3328	-1.5017	-1.5705	-1.6078
0.4545	-1.4667	-1.5517	-1.6224	-1.6434
0.5172	-1.4747	-1.5639	-1.6641	-1.6978
0.5738	-1.3878	-1.5340	-1.6342	-1.6794
0.6250	-1.3339	-1.4185	-1.4733	-1.5374
0.6716	-1.2179	-1.3293	-1.3669	-1.3964
0.7142	-1.0862	-1.1630	-1.2262	-1.2653
0.7534	-1.0089	-1.1109	-1.1404	-1.1719
0.7895	-0.8749	-0.9671	-1.0131	-1.0618
0.8228	-0.6692	-0.7671	-0.7954	-0.8023
0.9574	-0.2454	-0.2683	-0.2705	-0.2870
1.0000	0.0000	0.0000	0.0000	0.0000

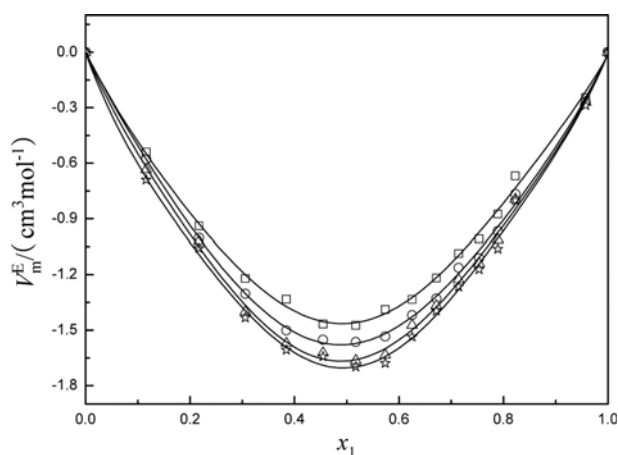


Fig. 1. Excess molar volumes with mole fraction for EDA (1)+TEG (2): □, 298.15 K; ○, 303.15 K; △, 308.15 K; ☆, 313.15 K.

of pure EDA and pure TEG, respectively. The results of V_m^E are listed in Table 4, and the dependence of V_m^E on various temperatures is displayed in Fig. 1.

From Table 4 and Fig. 1, V_m^E presents negative values over the whole composition range at each temperature, and it hardly changes at various temperatures. It was found that the minimum is at about $x_1=0.52$, which indicates one EDA molecule can combine one TEG molecule to form tighter complexes in the binary system of EDA+TEG. Moreover, the V_m^E values become more negative with the increasing temperatures.

A Redlich-Kister relation was used to correlate the excess molar volume data,

$$V_m^E \times 10^{-6} / \text{m}^3 \cdot \text{mol}^{-1} = x_1 x_2 \sum_{i=0}^n A_i (2x_1 - 1)^i \quad (3)$$

where x_1 denotes the mole fraction of EDA, x_2 denotes the mole fraction of TEG, and A_i denotes the polynomial coefficients.

The standard deviation values, σ , between the calculated and experimental data points are obtained by the following equation:

$$\sigma_{V_m^E} = [\sum (V_{m, \text{calc}}^E - V_m^E)^2 / (N - m)]^{1/2} \quad (4)$$

where N denotes the total number of experimental points and m

Table 5. Coefficients and standard deviations of excess mole volumes (V_m^E) for EDA+TEG

T/K	A_4	A_3	A_2	A_1	A_0	$\sigma / (\text{cm}^3 \cdot \text{mol}^{-1})$
298.15	-1.915	0.049	2.277	0.055	-6.010	0.0312
303.15	-1.835	-0.621	2.247	0.415	-6.302	0.0269
308.15	-2.916	0.489	7.824	11.199	0.193	0.0321
313.15	-3.470	0.241	2.930	0.264	-6.806	0.0393

Table 6. Experimental viscosities (ν) of EDA (1)+TEG (2) at T=(298.15 to 313.15) K^a

x_1	$\nu / (\text{mm}^2 \cdot \text{s}^{-1})$			
	T/K=298.15	T/K=303.15	T/K=308.15	T/K=313.15
0.0000	33.1	26.3	19.5	15.6
0.1163	38.1	30.9	22.8	18.1
0.2174	43.1	34.5	25.0	19.6
0.3061	45.5	36.0	26.0	20.3
0.3846	44.7	34.9	24.8	19.3
0.4545	39.7	31.1	22.5	17.5
0.5172	33.3	26.0	19.3	15.1
0.5738	26.5	20.7	15.7	12.5
0.6250	20.3	16.1	12.4	9.92
0.6716	15.5	12.6	9.70	7.94
0.7142	11.8	9.60	7.61	6.35
0.7534	9.12	7.67	6.11	5.11
0.7895	7.19	6.05	4.94	4.18
0.8228	5.99	5.09	4.19	3.57
0.9574	2.23	1.93	1.70	1.55
1.0000	1.46	1.36	1.21	1.11

^aStandard uncertainties u are $u(x_1)=0.0001$, $u(T)=0.01$ K and $u(\nu)=\pm 0.3\%$

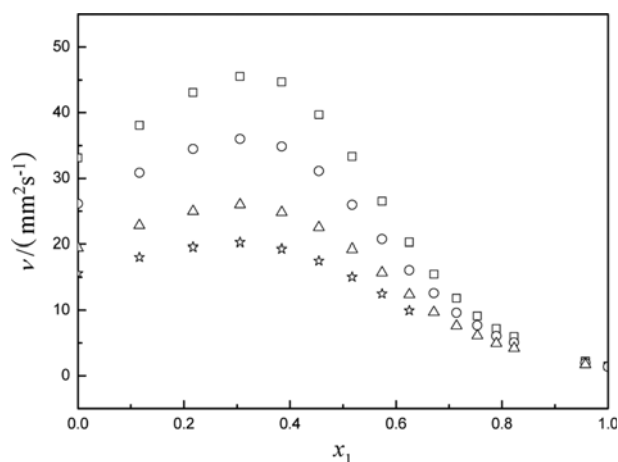


Fig. 2. Experimental viscosities with mole fraction for EDDDA (1)+TEG (2): □, 298.15 K; ○, 303.15 K; △, 308.15 K; ☆, 313.15 K.

denotes the number of A_i coefficients considered. The coefficients A_i and corresponding standard deviations (σ) are listed in Table 5.

Experimental measured viscosities of the system EDA+TEG at $T=(298.15, 303.15, 308.15, 313.15)$ K are listed in Table 6 and shown in Fig. 2. In all cases, the viscosities increase with the increasing EDA concentration in the range of $x_1=(0 \text{ to } 0.31)$, decrease with the increasing EDA concentration in the range of $x_1=(0.31 \text{ to } 1.0)$, and the well-defined maximum is found at about $x_1=0.31$. Meanwhile, the viscosities decrease with the increase of temperatures at the same composition.

The experimental values of ν for the various mixtures were used to calculate the deviation in kinematic viscosity ($\Delta\nu$), which is defined by the following equation:

$$\Delta\nu = \nu - (x_1\nu_1 + x_2\nu_2) \quad (5)$$

Table 7. Deviations in kinematic viscosity ($\Delta\nu$) for EDA (1)+TEG (2) at $T=(298.15 \text{ to } 313.15)$ K

x_1	$\Delta\nu/(\text{mm}^2 \cdot \text{s}^{-1})$			
	T/K=298.15	T/K=303.15	T/K=308.15	T/K=313.15
0.0000	0.00	0.00	0.00	0.00
0.1163	8.64	7.60	5.49	4.13
0.2174	16.8	13.7	9.49	7.12
0.3061	22.1	17.5	12.1	9.09
0.3846	23.7	18.2	12.4	9.28
0.4545	21.0	16.2	11.4	8.47
0.5172	16.6	12.6	9.25	6.96
0.5738	11.6	8.81	6.74	5.19
0.6250	6.96	5.45	4.32	3.37
0.6716	3.62	3.10	2.50	2.07
0.7142	1.30	1.16	1.19	1.10
0.7534	-0.145	0.197	0.406	0.425
0.7895	-0.938	-0.527	-0.109	0.015
0.8228	-1.08	-0.661	-0.257	-0.106
0.9574	-0.582	-0.487	-0.283	-0.179
1.0000	0.00	0.00	0.00	0.00

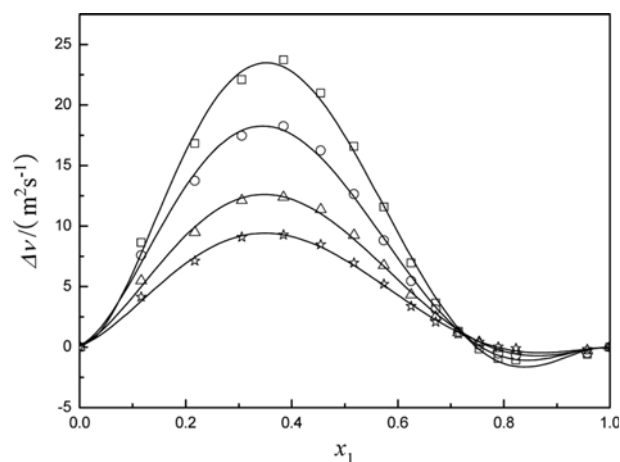


Fig. 3. Deviations in kinematic viscosity with mole fraction for EDA (1)+TEG (2): □, 298.15 K; ○, 303.15 K; △, 308.15 K; ☆, 313.15 K.

where ν is the kinematic viscosity of the mixtures, x_1 , x_2 , ν_1 , and ν_2 represent the mole fractions and the viscosities of pure EDA and pure TEG, respectively. The results of the deviation in kinematic viscosity, $\Delta\nu$, are listed in Table 7 and shown in Fig. 3.

The deviation in kinematic viscosity was also represented by the Redlich-Kister equation as follow:

$$\Delta\nu/\text{mm}^2 \cdot \text{s}^{-1} = x_1 x_2 \sum_{i=0}^n B_i (2x_1 - 1)^i \quad (6)$$

The coefficients B_i and the standard deviation σ are presented in Table 8.

The thermodynamic parameters of activation of viscous flow of the binary mixtures, such as Gibbs energy of activation of viscous flow (ΔG^*), enthalpy of activation for viscous flow (ΔH^*), and entropy of activation for the viscous flow (ΔS^*), were evaluated on the basis of Eyring's approach to Andrade's theory [27] with the kinematic viscosity expressed in the form,

$$\nu = \frac{h N_A}{M} \exp\left(\frac{\Delta G^*}{RT}\right) \quad (7)$$

where $M (=x_1 M_1 + x_2 M_2)$ is the average molar mass, h is Planck's constant, N_A is the Avogadro number, R is the gas constant, and T is the absolute temperature.

By applying the standard thermodynamic equation,

$$\Delta G^* = \Delta H^* - T\Delta S^* \quad (8)$$

It is possible to write

Table 8. Coefficients (B_0 , B_1 , B_2 , B_3 , B_4 , and B_5) and standard deviations (σ , $\text{mm}^2 \cdot \text{s}^{-1}$) of deviation in kinematic viscosity ($\Delta\nu$) for EDA+TEG

T/K	B_4	B_3	B_2	B_1	B_0	$\sigma/(\text{mm}^2 \cdot \text{s}^{-1})$
298.15	46.5	149	-85.7	-142	71.0	0.523
303.15	29.1	104	-53.3	-109	54.3	0.393
308.15	32.4	60.9	-43.6	-70.5	36.7	0.236
313.15	22.1	42.9	-31.1	-51.3	29.4	0.151

$$R \ln \left(\nu \frac{M}{hN_A} \right) = \frac{\Delta H^*}{T} - \Delta S^* \quad (9)$$

According to Eyring's theory, the enthalpy and entropy of activation for the viscous flow were calculated from Eq. (9).

Plots of the term on the left-hand side of Eq. (9) against $1/T$ for each binary mixture are linear in Fig. 4 and all the linearity (R^2) is above 0.99. The ΔH^* values were calculated from the slopes of the curves. The ΔH^* values so obtained, together with the ΔG^* values calculated from Eq. (7), were used to calculate the corresponding ΔS^* values by using Eq. (8). The ΔH^* and ΔS^* values are listed in

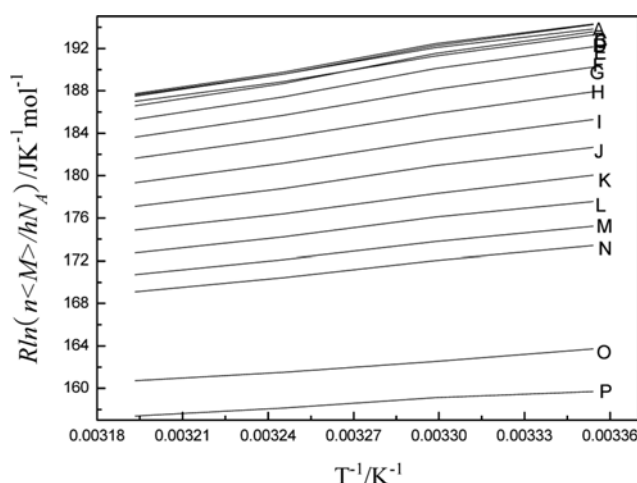


Fig. 4. Plots of $R \ln(\nu \langle M \rangle / hN_A)$ against $1/T$ for the EDA (1)+TEG (2) system at various temperatures, the molar fractions corresponding to lines as follows: A, 0.0000; B, 0.1163; C, 0.2174; D, 0.3061; E, 0.3846; F, 0.4545; G, 0.5172; H, 0.5738; I, 0.6250; J, 0.6716; K, 0.7142; L, 0.7534; M, 0.7895; N, 0.8228; O, 0.9574; P, 1.0000.

Table 9. Enthalpy of activation for viscous flow ($\Delta H^*/\text{kJ} \cdot \text{mol}^{-1}$) and entropy of activation for viscous flow ($\Delta S^*/\text{J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$) for the viscous flow about EDA+TEG

x_1	$\Delta S^*/\text{J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$	$\Delta H^*/\text{kJ} \cdot \text{mol}^{-1}$
0.0000	-60.4 ± 4.8	39.65 ± 1.45
0.1163	-61.7 ± 6.5	39.44 ± 2.00
0.2174	-54.5 ± 7.2	41.71 ± 2.19
0.3061	-50.9 ± 6.6	42.79 ± 2.02
0.3846	-45.0 ± 6.6	44.34 ± 2.03
0.4545	-47.4 ± 5.7	43.20 ± 1.76
0.5172	-50.7 ± 3.8	41.61 ± 1.15
0.5738	-55.8 ± 2.7	39.39 ± 0.82
0.625	-59.8 ± 2.7	37.43 ± 0.82
0.6716	-64.9 ± 4.2	35.14 ± 1.27
0.7142	-71.0 ± 2.9	32.51 ± 0.88
0.7534	-75.3 ± 3.8	30.51 ± 1.17
0.7895	-79.9 ± 2.3	28.43 ± 0.72
0.8228	-82.4 ± 2.5	27.15 ± 0.77
0.9574	-100.4 ± 3.2	18.85 ± 0.99
1.0000	-110.2 ± 3.5	14.77 ± 1.07

Table 10. Gibbs energies of activation of viscous flow ($\Delta G^*/\text{kJ} \cdot \text{mol}^{-1}$) for the viscous flow about EDA (1)+TEG (2) at $T=(298.15$ to $313.15)$ K

x_1	$\Delta G^*/\text{kJ} \cdot \text{mol}^{-1}$			
	$T/\text{K}=298.15$	$T/\text{K}=303.15$	$T/\text{K}=308.15$	$T/\text{K}=313.15$
0.0000	57.62	57.99	58.19	58.56
0.1163	57.79	58.23	58.42	58.75
0.2174	57.93	58.34	58.47	58.79
0.3061	57.91	58.29	58.41	58.71
0.3846	57.72	58.06	58.15	58.43
0.4545	57.28	57.63	57.75	58.03
0.5172	56.72	57.04	57.22	57.50
0.5738	56.03	56.34	56.57	56.88
0.6250	55.24	55.59	55.83	56.16
0.6716	54.46	54.85	55.09	55.46
0.7142	53.68	54.06	54.36	54.76
0.7534	52.94	53.39	53.69	54.09
0.7895	52.25	52.69	53.04	53.46
0.8228	51.70	52.15	52.51	52.95
0.9574	48.81	49.27	49.77	50.32
1.0000	47.61	48.23	48.73	49.29

Table 9 and the ΔG^* values are listed in Table 10. The observed data of ΔH^* and ΔG^* are positive for the present binary system of EDA+TEG suggesting a strong specific interaction among constituent molecules. The ΔS^* data are generally negative, which indicates that the viscous flow is an ordered process involving contiguous liquid layers which should retain their structural configuration even moving in a stationary steady state.

CONCLUSION

This paper reports experimental data of the densities and viscosities for the binary system of 1,2-ethanediamine (EDA)+triethylene glycol (TEG) at $T=(298.15, 303.15, 308.15, 313.15)$ K. The data of pure EDA and pure TEG are in good agreement with the literature values. These data were used to compute excess properties of the system. The calculated V_m^E values for EDA+TEG were negative at all temperatures and compositions. The observed data of ΔH^* and ΔG^* are positive and ΔS^* are negative for the present binary system of EDA+TEG suggesting a strong specific interaction between constituent molecules.

ACKNOWLEDGEMENTS

This work was supported by the National Natural Science Foundation of China (21166017), Program for New Century Excellent Talents in University (NCET-12-1017), the Research Fund for the Doctoral Program of Higher Education of China (20111514120002), Program for Young Talents of Science and Technology in Universities of Inner Mongolia Autonomous Region (NJYT-12-B13), the Natural Science Foundation of Inner Mongolia Autonomous Region (2011BS0601), the Inner Mongolia Science and Technology Key Projects, the Inner Mongolia Talented People Development Fund, and Yongfeng Boyuan Industry Co., Ltd. (Jiangxi Province, China).

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