

# Density, Viscosity, and Excess Properties for 1,2-Diaminoethane + 1,2-Ethanediol at (298.15, 303.15, and 308.15) K

Chunping Li,<sup>†</sup> Jianbin Zhang,<sup>\*,†</sup> Tong Zhang,<sup>†</sup> Xionghui Wei,<sup>\*,‡</sup> Enqi Zhang,<sup>†</sup> Na Yang,<sup>†</sup> Nana Zhao,<sup>†</sup> Min Su,<sup>†</sup> and Hui Zhou<sup>†</sup>

College of Chemical Engineering, Inner Mongolia University of Technology, Huhhot 010051, China, and Department of Applied Chemistry, College of Chemistry and Molecular Engineering, Peking University, Beijing 100190, China

Density ( $\rho$ ) and viscosity ( $\eta$ ) were determined over the whole concentration range for the binary mixtures of 1,2-diaminoethane + 1,2-ethanediol at (298.15, 303.15, and 308.15) K. The experimental results are compared with data published in the previous literature. The density values were used to calculate the excess molar volume ( $V_m^E$ ). The calculated results are fitted to a Redlich–Kister equation to obtain the coefficients and estimate the standard deviations between the experimental and the calculated quantities. The values of  $V_m^E$  are negative over the entire composition range.

## Introduction

This paper is a continuation of the systematic program on the properties of solution containing 1,2-ethanediol. 1,2-Ethanediol is an important industrial solvent with the potential gas scrubbing because of its favorable properties such as low vapor pressure, low toxicity, low viscosity, and low melting temperature. 1,2-Ethanediol and its similar solvents, including diethylene glycol (DEG) and poly(ethylene glycol) (PEG), present native hydrogen bonding sites<sup>1,2</sup> so that the potential desorption characters are presented in the regenerative processes of solutions dissolving SO<sub>2</sub>; therefore, in previous studies, great attention has been paid to aqueous 1,2-ethanediol and its similar solvents for SO<sub>2</sub> removal.<sup>3–7</sup>

In recent months, 1,2-diaminoethane had been put into 1,2-ethanediol to boost the absorption capacity of SO<sub>2</sub> in solutions containing 1,2-ethanediol in our research group. The main advantages of 1,2-diaminoethane and 1,2-ethanediol are also a high solubility capability for SO<sub>2</sub>, which leads to lower energy requirements for absorption processes of SO<sub>2</sub>. In actual use, the physical properties of 1,2-diaminoethane + 1,2-ethanediol mixtures are extremely important. Especially, the density and viscosity data of aqueous solutions are significant from the practical and theoretical viewpoint. Previously published data<sup>8–16</sup> showed the density and viscosity data of pure 1,2-diaminoethane and pure 1,2-ethanediol, but the density and viscosity data over the whole concentration range for the binary mixtures of 1,2-diaminoethane + 1,2-ethanediol are generally limited to ambient temperatures.

In the present work, the density and viscosity of 1,2-diaminoethane and 1,2-ethanediol were determined over the whole concentration range for the binary mixtures of 1,2-diaminoethane + 1,2-ethanediol at (298.15, 303.15, and 308.15) K. Meanwhile, the excess molar volume was derived.

## Experimental Section

**Materials.** The analytical grade 1,2-diaminoethane and 1,2-ethanediol were purchased from Beijing Reagent Company (Beijing, China). It was used after drying over molecular sieves (type 4A) and decompression filtration before measurements. The purity of the samples was checked by density determination at 298.15 K. The density of 1,2-ethanediol at 298.15 K was found to be 1.1096 g·cm<sup>-3</sup>, in good agreement with literature,<sup>13–15</sup> and the density of 1,2-diaminoethane at 298.15 K was found to be 0.8945 g·cm<sup>-3</sup>, in good agreement with literature.<sup>14</sup> Bidistilled water and high-performance liquid chromatography (HPLC) grade ethanol were used.

**Measurements.** Solvent mixtures were prepared by mass using an analytical balance with a precision of  $\pm 0.0001$  g (Sartorius BS 224S). The uncertainty in the mole fraction for each binary mixture is less than 0.0001.

Densities of pure liquids and their mixtures were determined using a bicapillary pycnometer having a bulb volume of 10 cm<sup>3</sup>. The volume of the pycnometer was calibrated as a function of temperature using distilled, deionized, and degassed water at various temperatures.<sup>17–19</sup> The pycnometer filled with liquid was kept in a thermostatically controlled and well-stirred water bath (maintained constant to  $\pm 0.01$  K) for (25 to 30) min to attain thermal equilibrium. The density measurements were carried out at (298.15, 303.15, and 308.15) K. Each experimental density value was an average of at least three measurements. The uncertainty of the density measurement was estimated to be  $\pm 0.02$  %.

The kinematic viscosity in both the pure components and their mixtures was determined with a commercial capillary viscometer of the Ubbelohde type with a 0.6 mm diameter, which was calibrated with high pure water and ethanol (HPLC grade) at the experimental temperature range whose viscosity and density were well-known, as has been described in literature.<sup>17–22</sup> A thoroughly cleaned and perfectly dried viscometer, filled with experimental solution, was placed exactly vertical in an insulated jacket, wherein constant temperature ( $\pm 0.02$  K) was maintained by circulating water from a thermoelectric controller at the required temperature. After thermal stability was attained, the flow times of the

\* To whom correspondence should be addressed. Tel.: +86-010-62751529. Fax: +86-010-62751529. E-mail: xhwei@pku.edu.cn (X. Wei) and tadzhang@pku.edu.cn (J. Zhang).

<sup>†</sup> Inner Mongolia University of Technology.

<sup>‡</sup> Peking University.

**Table 1. Comparison of Experimental Densities ( $\rho$ ) and Viscosities ( $\eta$ ) of 1,2-Ethanediol and 1,2-Diaminoethane with Literature Values at Various Temperatures**

$T/K$	$\rho/(\text{g}\cdot\text{cm}^{-3})$		$\eta/(\text{mPa}\cdot\text{s})$	
	expt.	lit.	expt.	lit.
1,2-Ethanediol				
298.15	1.1096	1.10987 <sup>11</sup>	16.795	15.312 <sup>16</sup>
		1.1085 <sup>13</sup>		18.6826 <sup>24</sup>
		1.1100 <sup>14,16</sup>		
		1.10988 <sup>15</sup>		
303.15	1.1058	1.10636 <sup>11</sup>	13.396	13.646 <sup>13</sup>
		1.10640 <sup>15</sup>		13.8678 <sup>24</sup>
		1.10665 <sup>23</sup>		
		1.11009 <sup>23</sup>		
308.15	1.1030	1.10282 <sup>11</sup>	11.333	11.6956 <sup>24</sup>
		1.1032 <sup>13</sup>		
		1.10289 <sup>15</sup>		
		1.10296 <sup>23</sup>		
1,2-Diaminoethane				
298.15	0.8945	0.891 <sup>12</sup>	1.276	
		0.8948 <sup>14</sup>		
		0.8952 <sup>26</sup>		
303.15	0.8908	0.890325 <sup>25</sup>	1.211	1.290 <sup>25</sup>
		0.8906 <sup>26</sup>		
308.15	0.8856	0.885616 <sup>25</sup>	1.107	1.167 <sup>25</sup>
		0.8859 <sup>26</sup>		

**Table 2. Experimental Densities ( $\rho$ ) of 1,2-Diaminoethane (1) + 1,2-Ethanediol (2)**

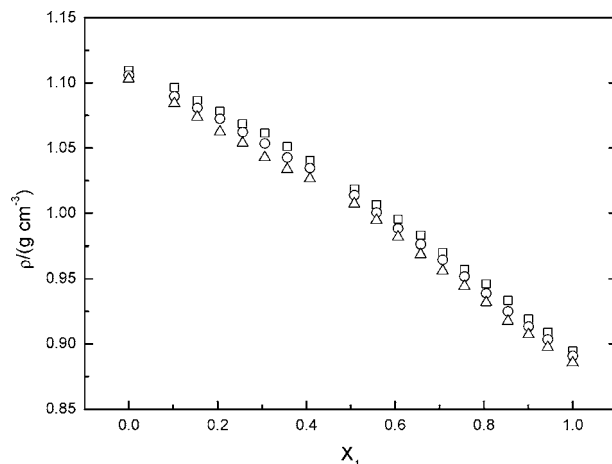
$x_1$	$\rho/(\text{g}\cdot\text{cm}^{-3})$		
	$T/K = 298.15$	$T/K = 303.15$	$T/K = 308.15$
0.0000	1.1096	1.1058	1.1030
0.1027	1.0966	1.0898	1.0844
0.1541	1.0866	1.0809	1.0738
0.2052	1.0784	1.0724	1.0624
0.2561	1.0685	1.0622	1.0538
0.3066	1.0614	1.0535	1.0429
0.3568	1.051	1.0428	1.0338
0.4078	1.0404	1.0348	1.0266
0.5080	1.0185	1.0139	1.0072
0.5579	1.0066	1.0007	0.9947
0.6065	0.9954	0.9883	0.9818
0.6573	0.9831	0.9762	0.9687
0.7071	0.9701	0.9645	0.9561
0.7560	0.9573	0.9517	0.9444
0.8051	0.9462	0.9388	0.9317
0.8542	0.9334	0.9249	0.9175
0.9003	0.9191	0.9134	0.9072
0.9438	0.9088	0.9034	0.8975
1.0000	0.8945	0.8908	0.8856

solutions were recorded with an electronic digital stopwatch correct to  $\pm 0.01$  s. At least 16 repetitions of each datum point obtained were reproducible to 0.06 s, and the results were averaged. Because all flow times were greater than 15 s and the capillary diameter (0.6 mm) was far less than its length (120 mm), the kinetic energy and end corrections, respectively, were found to be negligible. The viscosity was then calculated from the fundamental relationship.

The kinematic viscosity ( $\nu$ ) was calculated from the following equation

$$\nu = At - \frac{B}{t} \quad (1)$$

where  $\nu$  is the kinematic viscosity,  $A$  and  $B$  are viscometer contents, and  $t$  is its flow time in the viscometer, respectively.  $A$  and  $B$  are determined from measurements with the calibration fluids of water and ethanol. The absolute viscosity ( $\eta$ ) was obtained by multiplying the determined kinematic viscosity ( $\nu$ ) by the measured density ( $\eta = \nu\rho$ ). Each experimental point was

**Figure 1.** Experimental densities with mole fraction for 1,2-diaminoethane (1) + 1,2-ethanediol (2):  $\square$ , 298.15 K;  $\circ$ , 303.15 K;  $\triangle$ , 308.15 K.**Table 3. Excess Molar Volumes ( $V_m^E$ ) for 1,2-Diaminoethane (1) + 1,2-Ethanediol (2)**

$x_1$	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$		
	$T/K = 298.15$	$T/K = 303.15$	$T/K = 308.15$
0.0000	0.0000	0.0000	0.0000
0.1027	-0.6756	-0.5249	-0.4109
0.1541	-0.8272	-0.7332	-0.5371
0.2052	-1.0621	-0.9523	-0.6060
0.2561	-1.1979	-1.0708	-0.8165
0.3066	-1.4746	-1.2595	-0.8872
0.3568	-1.5604	-1.3248	-1.0458
0.4078	-1.6345	-1.5436	-1.3169
0.5080	-1.6897	-1.6540	-1.5248
0.5579	-1.6394	-1.5231	-1.4397
0.6065	-1.6016	-1.4074	-1.2961
0.6573	-1.5093	-1.3219	-1.1493
0.7071	-1.3428	-1.2334	-1.0029
0.7560	-1.1574	-1.0430	-0.8855
0.8051	-1.0703	-0.8290	-0.6849
0.8542	-0.8495	-0.5218	-0.3547
0.9003	-0.4620	-0.3232	-0.2431
0.9438	-0.3018	-0.1801	-0.1240
1.0000	0.0000	0.0000	0.0000

**Table 4. Coefficients and Standard Deviations of Excess Molar Volumes,  $V_m^E$ , for 1,2-Diaminoethane + 1,2-Ethanediol**

$T/K$	$A_0$	$A_1$	$A_2$	$A_3$	$A_4$	$\sigma/\text{cm}^3\cdot\text{mol}^{-1}$
298.15	-6.788	-0.058	0.824	0.528	-0.522	0.0475
313.15	-6.294	-0.298	2.741	2.194	-0.584	0.0438
308.15	-5.761	-1.519	7.698	3.964	-7.112	0.0529

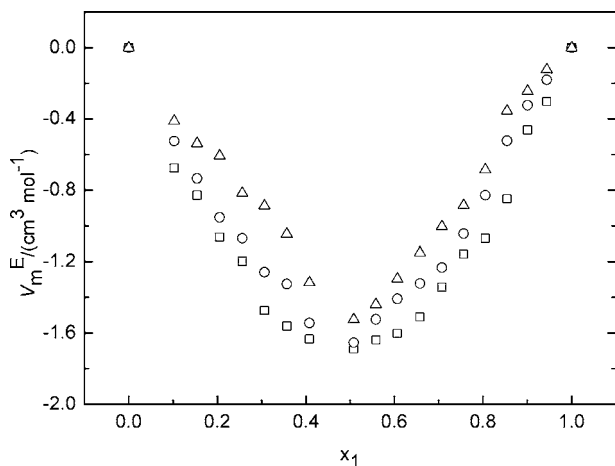
the average of 16 sets with a maximum deviation of  $\pm 0.02$  % in the flow time.

The experimental densities and viscosities of pure 1,2-diaminoethane and 1,2-ethanediol are compared with the available literature values are given in Table 1.

## Results and Discussion

Experimental densities of the binary solutions of 1,2-diaminoethane (1) + 1,2-ethanediol (2) at (298.15, 303.15, and 308.15) K throughout the whole concentration range are summarized in Table 2. The values of the measured density at different temperatures are shown in Figure 1.

Table 2 shows that the density values decrease with the increasing 1,2-diaminoethane concentration in binary solutions over the whole concentration range. The density values decrease with the augment of temperature at the same concentration.



**Figure 2.** Excess molar volumes with mole fraction for 1,2-diaminoethane (1) + 1,2-ethanediol (2): □, 298.15 K; ○, 303.15 K; △, 308.15 K.

**Table 5.** Experimental Viscosities ( $\eta$ ) of 1,2-Diaminoethane (1) + 1,2-Ethanediol (2)

$x_1$	$\eta/(\text{mPa}\cdot\text{s})$		
	$T/\text{K} = 298.15$	$T/\text{K} = 303.15$	$T/\text{K} = 308.15$
0.0000	16.795	13.396	11.333
0.1027	22.284	17.598	14.049
0.1541	25.541	20.556	16.928
0.2052	28.368	22.441	18.213
0.2561	30.733	23.153	19.284
0.3066	31.457	23.656	19.196
0.3568	30.700	22.853	18.495
0.4078	28.318	21.307	17.004
0.5080	20.173	15.184	11.891
0.5579	16.057	12.272	8.772
0.6065	12.509	9.748	6.254
0.6573	9.028	7.157	4.674
0.7071	6.882	5.681	4.008
0.7560	5.154	4.177	3.547
0.8051	3.787	3.154	2.683
0.8542	3.199	2.552	2.128
0.9003	2.273	1.920	1.713
0.9438	1.800	1.561	1.422
1.0000	1.276	1.211	1.107

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

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

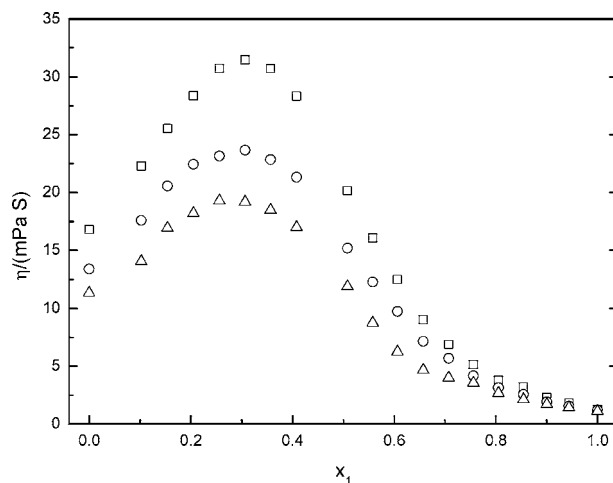
where  $\rho_m$  is the density of the mixture and  $x_1$ ,  $\rho_1$ ,  $M_1$ ,  $x_2$ ,  $\rho_2$ , and  $M_2$  are the mole fractions, densities, and molecular weights of pure 1,2-diaminoethane and pure 1,2-ethanediol, respectively. The results of  $V_m^E$  are listed in Tables 3 and 4, and the dependence of  $V_m^E$  at various temperatures is displayed in Figure 2.

Figure 2 shows that  $V_m^E$  is negative for all of the mixtures over the entire mole fraction range at each temperature. The maximum is at about  $x_1 = 0.50$ . Additionally, these  $V_m^E$  values become less negative with the increasing temperature.

A Redlich–Kister relation was used to correlate the excess volume data.

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

where  $x_1$  is the mole fraction of 1,2-diaminoethane,  $x_2$  is the mole fraction of 1,2-ethanediol,  $A_i$  are the polynomial coef-



**Figure 3.** Experimental viscosities with mole fraction for 1,2-diaminoethane (1) + 1,2-ethanediol (2): □, 298.15 K; ○, 303.15 K; △, 308.15 K.

ficients which were evaluated from the least-squares method, and  $n$  is the polynomial degree.

The standard deviation values,  $\sigma$ , between the calculated and the experimental data points are obtained by the following equation

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

where  $N$  is the total number of experimental points and  $m$  is the number of  $A_i$  coefficients considered. The coefficients  $A_i$  and corresponding standard deviations,  $\sigma$ , are listed in Table 4.

Experimentally measured viscosities of the binary solutions of 1,2-diaminoethane + 1,2-ethanediol at (298.15, 303.15, and 308.15) K are listed in Table 5 and shown in Figure 3. In all cases, the viscosities increase with the increasing 1,2-diaminoethane concentration in the range of 0 to 0.3, decrease with the increasing 1,2-diaminoethane concentration in the range of 0.3 to 1.0, and decrease with the increasing temperature.

## Conclusion

This paper reports experimental data for the densities and viscosities of the binary system of 1,2-diaminoethane + 1,2-ethanediol at (298.15, 303.15, and 308.15) K. The data of pure 1,2-diaminoethane and pure 1,2-ethanediol generally agreed with available literature data. These data have been used to compute excess properties of the system. The calculated  $V_m^E$  values for 1,2-diaminoethane + 1,2-ethanediol were negative at all temperatures and compositions.

## Literature Cited

- Zhang, J. B.; Han, F.; Wei, X. H.; Shui, L. K.; Gong, H.; Zhang, P. Y. Spectral Studies of Hydrogen Bonding and Interaction in the Absorption Processes of Sulfur Dioxide in Poly(ethylene Glycol) 400 + Water Binary System. *Ind. Eng. Chem. Res.* **2010**, *49*, 2025–2030.
- Zhang, J. B.; Zhang, P. Y.; Han, F.; Chen, G. H.; Zhang, L. W.; Wei, X. H. Hydrogen Bonding and Interaction in the Absorption Processes of Sulfur Dioxide in Ethylene Glycol + Water Binary Desulfurization System. *Ind. Eng. Chem. Res.* **2009**, *48*, 1287–1291.
- Li, X. X.; Liu, Y. X.; Wei, X. H. Hydrolysis of Carbonyl Sulfide in Binary Mixture of Diethylene Glycol Diethyl Ether with Water. *Chin. J. Chem. Eng.* **2005**, *13*, 234–238.
- Wei, X. H. Desulfurization & Decarburization Solution Activities. CN Patent 02130605.2, 2002.
- Wei, X. H.; Zhang, J. B.; Zhang, P. Y.; Zhang, L. W.; Li, X. B.; Wan, M. J. Removal of  $\text{SO}_x$  from Flue Gas by Ethylene Glycol. CN Patent 101053746, 2007.
- Zhang, J. B.; Zhang, P. Y.; Chen, G. H.; Han, F.; Wei, X. H. Gas-Liquid Equilibrium Data for Mixture Gas of Sulfur Dioxide/Nitrogen

- with Ethylene Glycol at Temperatures from 298.15 to 313.15 K under Low Pressures. *J. Chem. Eng. Data* **2008**, *53*, 1479–1485.
- (7) Zhang, J. B.; Zhang, P. Y.; Han, F.; Chen, G. H.; Deng, R. H.; Wei, X. H. Gas-Liquid Equilibrium Data for Mixture Gas of Sulfur Dioxide/Nitrogen with Ethylene Glycol Aqueous Solutions at 298.15 K and 123.15 kPa. *J. Chem. Eng. Data* **2008**, *53*, 2372–2374.
- (8) Burman, A. U.; Strom, K. H. U. Density for (Water + Ethylenediamine) at Temperatures between (283 and 353) K. *J. Chem. Eng. Data* **2008**, *53*, 2307–2310.
- (9) Kapadi, U. R.; Hundiwale, D. G.; Patil, N. B.; Lande, M. K. Effect of Temperature on Excess Molar Volumes and Viscosities of Binary Mixtures of Ethylenediamine and Water. *Fluid Phase Equilib.* **2003**, *205*, 267–274.
- (10) Saleh, M. A.; Akhtar, S.; Ahmed, M. S. Excess Molar Volumes of Aqueous Systems of Some Diamines. *J. Mol. Liq.* **2005**, *116*, 147–156.
- (11) Azizian, S.; Bashavard, N. Equilibrium Surface Tensions of Benzyl Alcohol + Ethylene. *J. Chem. Eng. Data* **2005**, *50*, 709–712.
- (12) Good, W. D.; Moore, R. T. Enthalpies of Formation of Ethylenediamine, 1,2-Propanediamine, 1,2-Butanediamine, 2-Methyl-1,2-propanediamine, and Isobutylamine. *J. Chem. Eng. Data* **1970**, *15*, 150–154.
- (13) Yang, C.; Ma, P.; Jing, F.; Tang, D. Excess Molar Volumes, Viscosities, and Heat Capacities for the Mixtures of Ethylene Glycol + Water from 273.15 to 353.15 K. *J. Chem. Eng. Data* **2003**, *48*, 836–840.
- (14) Gladden, J. K. Excess Thermodynamic Properties of Ethylene Diamine-Ethylene Glycol Solutions at 25 °C. *J. Chem. Eng. Data* **1972**, *17*, 468–471.
- (15) Zorebski, E.; Waligora, A. Densities, Excess Molar Volumes, and Isobaric Thermal Expansibilities for 1,2-Ethanediol + 1-Butanol, or 1-Hexanol, or 1-Octanol in the Temperature Range from (293.15 to 313.15) K. *J. Chem. Eng. Data* **2008**, *53*, 591–595.
- (16) Nagashima, A.; Yoshii, S.; Matsuda, H.; Ochi, K. Measurement and Correlation of Excess Molar Enthalpies for Ethylene Glycol + Alkanol Systems at the Temperatures (298.15, 308.15, and 323.15) K. *J. Chem. Eng. Data* **2004**, *49*, 286–290.
- (17) Henni, A.; Hromek, J. J.; Tontiwachwuthikul, P.; Chakma, A. Volumetric Properties and Viscosities for Aqueous AMP Solutions from 25 °C to 70 °C. *J. Chem. Eng. Data* **2003**, *48*, 551–556.
- (18) Comesana, J. F.; Otero, J. J.; García, E.; Correa, A. Densities and Viscosities of Ternary Systems of Water + Glucose + Sodium Chloride at Several Temperatures. *J. Chem. Eng. Data* **2003**, *48*, 362–366.
- (19) Alvarez, E.; Cancela, A.; Maceiras, R.; Navaza, J. M.; Taboas, R. Density, Viscosity, Excess Molar Volume, and Viscosity Deviation of Three Amyl Alcohols + Ethanol Binary Mixtures from 293.15 to 323.15 K. *J. Chem. Eng. Data* **2006**, *51*, 940–945.
- (20) Gomez Marigliano, A. C.; Solimo, H. N. Density, Viscosity, Excess Molar Volume, Viscosity Deviation, and Their Correlations for Formamide + Three Alkan-1-ols Binary Systems. *J. Chem. Eng. Data* **2002**, *47*, 796–800.
- (21) Djojoputro, H.; Ismadji, S. Density and Viscosity for a Binary Mixture of *cis*-3-Hexenyl Formate, Butyl Acetate, *trans*-2-Hexenyl Acetate, and *cis*-3-Hexenyl Acetate with Ethanol at Several Temperatures. *J. Chem. Eng. Data* **2005**, *50*, 1009–1013.
- (22) Nikam, P. S.; Shirsat, L. N.; Hasan, M. Density and Viscosity Studies of Binary Mixtures of Acetonitrile with Methanol, Ethanol, Propan-1-ol, Propan-2-ol, Butan-1-ol, 2-Methylpropan-1-ol, and 2-Methylpropan-2-ol at (298.15, 303.15, 308.15, and 313.15) K. *J. Chem. Eng. Data* **1998**, *43*, 732–737.
- (23) Jimenez, E.; Cabanas, M.; Segade, L.; Garcia-Garabal, S.; Casas, H. Excess Volume, Changes of Refractive Index and Surface Tension of Binary 1,2-Ethanediol + 1-Butanol Mixtures at Several Temperatures. *Fluid Phase Equilib.* **2001**, *180*, 151–164.
- (24) Tsierekzos, N. G.; Molinou, I. E. Thermodynamic Properties of Water + Ethylene Glycol at 283.15, 293.15, 303.15, and 313.15 K. *J. Chem. Eng. Data* **1998**, *43*, 989–993.
- (25) Kapadi, U. R.; Hundiwale, D. G.; Patil, N. B.; Lande, M. K. Effect of Temperature on Excess Molar Volumes and Viscosities of Binary Mixtures of Ethylenediamine and Water. *Fluid Phase Equilib.* **2003**, *25*, 267–274.
- (26) Saleh, M. A.; Akhtar, S.; Ahmed, S. Excess Molar Volumes of Aqueous Systems of Some Diamines. *J. Mol. Liq.* **2005**, *116*, 147–156.

Received for review March 31, 2010. Accepted June 4, 2010. This work was supported by foundation of Inner Mongolia Autonomous Region's Educational Commission (NJ10079) and Yongfeng Boyuan Industry Co., Ltd. (Jiangxi Province, China).

JE100290A