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Densities and Viscosities of Binary Solutions of Benzene-1,3-diol + Water, Ethanol, Propan-1-ol, and Butan-1-ol at T = (293.15 to 333.15) K

Jingru Liu, Chunying Zhu,* and Youguang Ma

School of Chemical Engineering and Technology, State Key Laboratory of Chemical Engineering, Tianjin University, Tianjin 300072, P. R. China

ABSTRACT: The densities and the viscosities of benzene-1,3-diol in different solvents of water, ethanol, propan-1-ol and butan-1-ol have been determined under atmospheric pressure and at T = (293.15 to 333.15) K over the mole fraction ranging from 0.0400 to 0.2000 for benzene-1,3-diol + water and 0.0700 to 0.3500 for benzene-1,3-diol + ethanol, propan-1-ol, and butan-1-ol. The measured densities of benzene-1,3-diol solutions showed a good linear relation to the temperature or to the molar concentration, whereas the viscosities of benzene-1,3-diol solutions displayed a nonlinear relation to the temperature or to the molar concentration. The experimental data of densities and viscosities of benzene-1,3-diol solutions were fitted respectively using Gonzáles equation and the extended Jones-Dole equation.

INTRODUCTION

The densities and the viscosities are basic data for describing material property in chemical engineering design and process optimization involving chemical separations, fluid flow, heat transfer, and mass transfer.[1−3] They are also essential to further study the molecular thermodynamic of solutions.[4,5] benzene-1,3-diol is an important fine chemical product and could easily induce many chemical reactions such as hydrogenation, halogenation, alkylation, and nitrification due to its particular molecular structure. Therefore, benzene-1,3-diol has been widely used in the fields of rubber industry, timber processing, agricultural chemicals and dyestuff and so on.[6] Water and alcohols are excellent solvents for benzene-1,3-diol, and it is useful and necessary to know the physicochemical properties of benzene-1,3-diol in water and alcohols. However, few measurements have been made on the physicochemical properties of benzene-1,3-diol in the solvents of water and alcohols in the literature. Harkins and Gibson measured densities of aqueous benzene-1,3-diol solution at (293.15 and 298.15) K, respectively.[7,8] Roy et al. presented the densities and the viscosities of aqueous benzene-1,3-diol solutions at T = (298.15, 308.15, and 318.15) K.[9] Baluja conducted ultrasonic study of benzene-1,3-diol in protic and aprotic solvents at 313.15 K.[10] Bayram et al. studied the effect of structural isomerism on the behavior of dihydroxybenzenes in aqueous solutions by the determination of densities.[11] However, most of the researchers focused mainly on the densities of dilute aqueous benzene-1,3-diol solutions.[12,13] The densities and the viscosities of benzene-1,3-diol especially in the solvents of alcohols have not been yet reported systematically. In the present work, the densities and the viscosities of benzene-1,3-diol in water, ethanol, propan-1-ol and butan-1-ol at T = (293.15 to 333.15) K were determined. Meanwhile, the experimental data of the densities of benzene-1,3-diol solutions were fitted using Gonzáles equation,[14] and the extended Jones-Dole equation[15] was used to fit the experimental data of the viscosities of benzene-1,3-diol solutions, the average deviations and the standard deviations were also calculated.

EXPERIMENTAL SECTION

The analytical-grade ethanol, propan-1-ol and butan-1-ol were purchased from Tianjin Kermel Chemical Reagent Co., Ltd.; the mass purity is >0.997 for ethanol, >0.995 for propan-1-ol and butan-1-ol. benzene-1,3-diol was provided by Changzhou Changyu Chemical Co., Ltd., and its mass purity is >0.996. The doubly distilled deionized water was used for the preparation of solutions.

The solutions for the whole molality range at room temperature were prepared by mass using an analytical balance with an uncertainty of 0.0001 g. The uncertainty in the molality for each binary solution is 0.0001 $mol \cdot kg^{-1}$.

The densities of solutions and the corresponding pure solvents were measured using a vibrating tube density meter DMA 4500 M (Anton Paar, Austria). Two integrated Pt100 platinum thermometers together with built-in peltier elements provide an extremely precise thermosetting of the sample and the temperature was kept constant within ± 0.01 K. The uncertainties of the apparatus is $\pm 5.0 \times 10^{-5}$ $g \cdot cm^{-3}$. Before each measurement, the apparatus was calibrated with doubly distilled water and dry air at atmospheric pressure.[16] Triplicate measurements were conducted to obtain the average value of density.

The viscosities of the benzene-1,3-diol + water, ethanol, propan-1-ol, and butan-1-ol binary solutions were measured using an Ubbelohde capillary viscometer (Shanghai Qihang Glass Instruments Factory, China) of about 0.50 mm diameter. A thoroughly cleaned and dried viscometer filled with experimental solution was placed exactly vertical in an insulated jacket, where constant temperature (± 0.01 K) was maintained by circulating water from a thermoelectric controller TC-502D (Brookfield Engineering Laboratories, U.S.A.) at the required temperature. An electronic digital stopwatch with uncertainty of 0.01 s was used for flow time measurements. At least three repetitions of each data point obtained were reproducible to ± 0.2 s, and the

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Table 1. Comparison of Experimental Densities and Viscosities of Alcohols with Literature Values

		$10^{-3}\rho/(\mathrm{kg\cdot m^{-3}})$		$\eta/(\mathrm{mPa\cdot s})$	
	T/K	exp	lit	exp	lit
ethanol	293.15	0.79008	0.78900[18] 0.7912[19] 0.789921[20] 0.7890[21] 0.7902[22] 0.78833[23]	1.21	1.2050[27] 1.203[19] 1.1617[21] 1.2103[22]
	303.15	0.78100	0.78089[24] 0.7816[19] 0.7818[25] 0.781313[20] 0.7803[21] 0.78072[23]	1.00	0.995[28] 1.002[19] 0.9940[29] 0.9645[21]
	313.15	0.77231	0.77220[26] 0.7740[19] 0.7734[25] 0.772569[20] 0.7720[21] 0.77261[23]	0.83	0.8290[24] 0.8100[21]
	323.15	0.76335	0.76324[24] 0.7652[19] 0.763643[20] 0.7630[21] 0.76414[23]	0.69	0.6868[21] 0.715[19]
	333.15	0.75420	0.75440[21] 0.7554[19]	0.58	0.5872[21] 0.602[19]
propan-1-ol	293.15	0.80376	0.80362[30] 0.8034[31] 0.80364[32]	2.19	2.238[30] 2.198[38] 2.188[32]
	303.15	0.79574	0.7957[33] 0.7955[34] 0.7959[35] 0.79565[36]	1.73	1.745[30] 1.719[39] 1.725[36] 1.722[40]
	313.15	0.78746	0.78737[37] 0.7873[31] 0.7875[34]	1.37	1.381[30] 1.361[38] 1.363[39] 1.378[32]
	323.15	0.77902	0.77391[30]	1.10	1.115[30]
	333.15	0.77045	0.76731[30]	0.90	0.907[30]
butan-1-ol	293.15	0.80979	0.80982[30] 0.8095[31] 0.80956[36]	2.93	2.963[30] 2.941[38] 2.864[42]
	303.15	0.80209	0.80203[30] 0.80195[36] 0.80201[37] 0.8020[35]	2.26	2.271[37] 2.255[38] 2.268[43] 2.273[42]
	313.15	0.79437	0.79437[39] 0.79438[41] 0.79432[42] 0.79435[36]	1.77	1.783[39] 1.754[38] 1.7734[33] 1.7567[41]
	323.15	0.78643	0.78655[30] 0.78670[42]	1.41	1.421[30]
	333.15	0.77815	0.77781[30]	1.14	1.137[30]

results were averaged. Because all flow times were greater than 100 s and the capillary diameter (0.5 mm) was far less than its length (120 mm), the kinetic energy and the end corrections were found to be negligible. The viscosity η of the solutions was calculated from the following equation:[17]

$$\frac{\eta}{\eta_w} = \frac{\rho t}{\rho_w t_w} \tag{1}$$

where η, ρ, and t and η_w, ρ_w, and t_w are viscosities, densities, and flow times of the solutions and water, respectively. The viscosity and the density of pure water were obtained from Lange's Handbook of Chemistry.[16] The uncertainty of the viscosity measurement is $\pm$ 0.07 mPa·s.

The experimental densities and viscosities of alcohols were compared with the available data in literature, and the results were given in Table 1.[18−43] It could be found that the experimental result for the densities of benzene-1,3-diol solutions showed good agreement with the data in literature, but discrepancies appeared between the measured viscosities of benzene-1,3-diol solutions and the results in literature, which may be resulted from reagents purity or accuracy of the apparatuses.

RESULTS AND DISCUSSION

The experimental densities and viscosities of benzene-1,3-diol + water, ethanol, propan-1-ol and butan-1-ol binary solutions as a function of benzene-1,3-diol concentration and temperature are reported in Table 2.

It could be found that densities of benzene-1,3-diol + water, ethanol, propan-1-ol, and butan-1-ol binary solutions show linear relation with benzene-1,3-diol molar concentration or temperature,

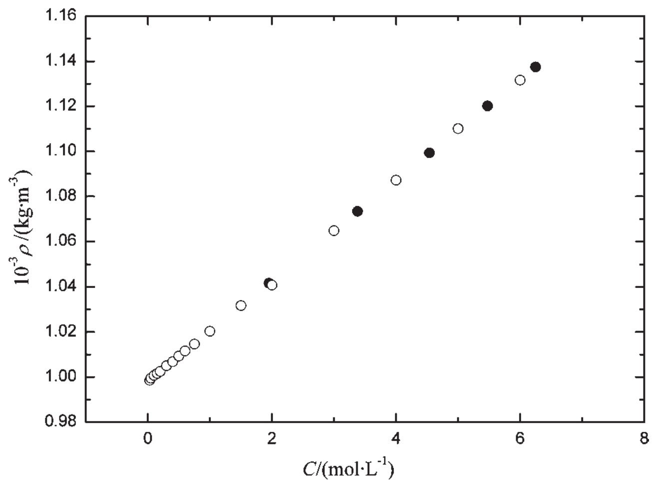

Figure 1. Comparison of densities of benzene-1,3-diol + water with ref 7 at T = 293.15 K: ●, experimental density; ○, literature density.

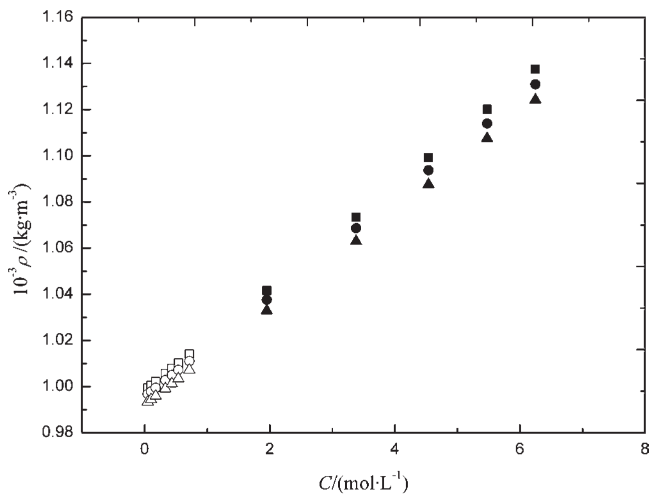

Figure 2. Comparison of densities of benzene-1,3-diol + water with ref 11 at T = (293.15, 303.15, and 313.15) K: ■, experimental density at 293.15 K; ●, experimental density at 303.15 K; ▲, experimental density at 313.15 K; □, literature density at 293.15 K; ○, literature density at 303.15K; Δ, literature density at 313.15 K; C, molar concentration at 293.15 K.

which is the same as Harkins's[7] and Bayram's[11] studies. But the molality range of benzene-1,3-diol in aqueous solution was confined merely from (0.0479 to 0.7623) mol·kg^{-1} in Bayram's study.[11] The densities of benzene-1,3-diol + water show good agreement with Harkins's[7] data at T = 293.15 K in Figure 1 and the densities show the same trend as Bayram's[11] data at T = (293.15, 303.15, and 313.15) K in Figure 2. For a given molality of benzene-1,3-diol, the density decreases with the rise of temperature. In the whole range of molality studied, the viscosities of benzene-1,3-diol increase nonlinearly with the increase of molality, but decrease with the rise of temperature at a given molality of benzene-1,3-diol.

The Gonzáles equation was used to correlate the experimental data[14]

$$\rho = A_1 + A_2 T + A_3 C \tag{2}$$

where A_1, A_2, and A_3 are the empirical constants determined by fitting experimental data, C is the molar concentration of

Table 2. Densities, ρ, Viscosities, η of Benzene-1,3-diol + Water, Ethanol, Propan-1-ol and Butan-1-ol Binary Solutions

		T/K = 293.15		T/K = 303.15		T/K = 313.15		T/K = 323.15		T/K = 333.15	
C	m	$10^{-3}\rho$	η	$10^{-3}\rho$	η	$10^{-3}\rho$	η	$10^{-3}\rho$	η	$10^{-3}\rho$	η
$mol \cdot L^{-1}$	$mol \cdot kg^{-1}$	$kg \cdot m^{-3}$	$mPa \cdot s$	$kg \cdot m^{-3}$	$mPa \cdot s$	$kg \cdot m^{-3}$	$mPa \cdot s$	$kg \cdot m^{-3}$	$mPa \cdot s$	$kg \cdot m^{-3}$	$mPa \cdot s$
					Benzene-1,3-diol + Water						
0.0000	0.0000	0.99823		0.99568		0.99223		0.98806		0.98218	
		0.99820[16]	1.002[16]	0.99565[16]	0.7977[16]	0.99222[16]	0.6532[16]	0.98804[16]	0.5470[16]	0.98320[16]	0.4665[16]
1.9515	2.3605	1.04162	1.61	1.03763	1.25	1.03294	1.01	1.02774	0.83	1.02198	0.70
3.3795	4.8190	1.07342	2.48	1.06864	1.87	1.06296	1.47	1.05726	1.18	1.05076	0.97
4.5378	7.5673	1.09932	3.68	1.09372	2.72	1.08757	2.09	1.08127	1.63	1.07455	1.31
5.4754	10.5860	1.12013	5.35	1.11397	3.84	1.10755	2.87	1.10086	2.19	1.09426	1.73
6.2476	13.8995	1.13742	7.61	1.13096	5.36	1.12423	3.90	1.11762	2.91	1.11053	2.25
					Benzene-1,3-diol + Ethanol						
0.0000	0.0000	0.79008	1.21	0.78100	1.00	0.77231	0.83	0.76335	0.69		
1.1729	1.6338	0.84702	2.14	0.83853	1.71	0.83012	1.42	0.82165	1.26		
2.2836	3.5365	0.89716	3.74	0.88903	3.08	0.88071	2.29	0.87263	1.95		
3.3224	5.7700	0.94164	6.37	0.93372	4.87	0.92587	3.65	0.91818	2.86		
4.2924	8.4413	0.98115	11.33	0.97342	8.49	0.96564	5.94	0.95795	4.30		
5.1987	11.6930	1.01703	21.33	1.00747	15.56	1.00206	9.50	0.99449	6.58		
					Benzene-1,3-diol + Propan-1-ol						
0.0000	0.0000	0.80376	2.19	0.79574	1.73	0.78746	1.37	0.77902	1.10	0.77045	0.90
0.9316	1.2524	0.84641	3.60	0.83823	2.74	0.83012	2.13	0.82185	1.67	0.81335	1.37
1.8504	2.7087	0.88687	6.04	0.87870	4.38	0.87062	3.29	0.86249	2.47	0.85418	1.95
2.7502	4.4230	0.92461	10.37	0.91662	7.22	0.90869	5.14	0.90065	3.74	0.89248	2.86
3.6293	6.4707	0.96050	18.52	0.95255	12.05	0.94474	8.18	0.93683	5.70	0.92884	4.19
4.4834	8.9594	0.99410	33.22	0.98639	20.29	0.97872	12.82	0.97097	8.20	0.96310	6.41
					Benzene-1,3-diol + Butan-1-ol						
0.0000	0.0000	0.80979	2.93	0.80209	2.26	0.79437	1.77	0.78643	1.41	0.77815	1.14
0.7712	1.0155	0.84432	4.52	0.83669	3.36	0.82892	2.54	0.82101	1.97	0.81292	1.67
1.5533	2.1963	0.87829	7.01	0.87007	5.06	0.86236	3.71	0.85451	2.81	0.84652	2.33
2.3410	3.5864	0.91051	11.56	0.90298	8.02	0.89533	5.55	0.88757	4.09	0.87968	3.27
3.1346	5.2467	0.94260	19.75	0.93510	12.64	0.92753	8.60	0.91986	6.10	0.91209	4.84
3.9317	7.2647	0.97413	34.34	0.96670	20.69	0.95920	13.18	0.95160	8.80	0.94392	6.64

Table 3. Coefficients of eq 2 for Densities of Benzene-1,3-diol + Water, + Ethanol, + Propan-1-ol, and + Butan-1-ol Binary Solution

	A_1	$10^4 A_2$	$10^5 A_3$	AD	SD
	$g \cdot cm^{-3}$	$g \cdot cm^{-3} \cdot K^{-1}$	$g \cdot mol^{-1}$		$g \cdot cm^{-3}$
benzene-1,3-diol + water	1.1584	−5.3720	2.1435	0.001	0.002
benzene-1,3-diol + ethanol	1.0312	−7.9414	4.2542	0.002	0.002
benzene-1,3-diol + propan-1-ol	1.0434	−8.0156	4.1830	0.001	0.001
benzene-1,3-diol + butan-1-ol	1.0399	−7.7286	4.1221	0.0008	0.0008

benzene-1,3-diol solutions at 293.15 K. The fitting parameters A_1, A_2, and A_3 are shown in Table 3.

The relative viscosities for benzene-1,3-diol + water, ethanol, propan-1-ol, and butan-1-ol binary solutions were calculated by the extended Jones-Dole equation[15]

$$\eta_r = \frac{\eta}{\eta_0} = 1 + B_1 m^{1/2} + B_2 m + B_3 m^2 \qquad (3)$$

where η_r is the relative viscosity; η and η_0 are the viscosities of the benzene-1,3-diol solution and the solvent (water, ethanol, propan-1-ol and butan-1-ol), respectively; m is the molality of benzene-1,3-diol in the solutions; B_1, B_2, and B_3 are the constants at a given temperature and characteristic of the solute and the solvent. B_1, generally positive values, accounts for solute−solute interactions. B_2 is an empirical constant which depends on solute−solvent interactions and can have positive or negative

Table 4. Coefficients of eq 3 for Viscosities of Benzene-1,3-diol + Water + Ethanol, + Propan-1-ol, and + Butan-1-ol Binary Solutions

T	B_1	B_2	B_3		SD
K	$kg^{1/2} \cdot mol^{-1/2}$	$kg \cdot mol^{-1}$	$kg^2 \cdot mol^{-2}$	AD	mPa·s
		Benzene-1,3-diol + Water			
293.15	0.0039	0.19	0.020	0.003	0.01
303.15	0.0091	0.20	0.015	0.003	0.009
313.15	0.0036	0.20	0.011	0.002	0.007
323.15	0.0036	0.20	0.0079	0.0008	0.002
333.15	0.0027	0.19	0.0056	0.002	0.003
		Benzene-1,3-diol + Ethanol			
293.15	1.2	−0.58	0.14	0.04	0.2
303.15	1.0	−0.45	0.12	0.04	0.2
313.15	0.19	0.20	0.055	0.004	0.02
323.15	0.40	0.16	0.039	0.007	0.02
		Benzene-1,3-diol + Propan-1-ol			
293.15	0.73	−0.33	0.19	0.01	0.2
303.15	0.32	0.049	0.12	0.005	0.05
313.15	0.075	0.28	0.070	0.003	0.04
323.15	−0.18	0.49	0.032	0.02	0.09
333.15	0.25	0.14	0.051	0.005	0.03
		Benzene-1,3-diol + Butan-1-ol			
293.15	0.58	−0.22	0.20	0.009	0.1
303.15	0.25	0.12	0.13	0.007	0.1
313.15	0.050	0.29	0.080	0.004	0.05
323.15	−0.12	0.44	0.045	0.01	0.06
333.15	−0.10	0.49	0.030	0.02	0.07

values. The precise physical meaning of B_3 is still not clear so far, and it seems to account for solute−solute interactions.[15]

The coefficients B_1, B_2, and B_3 listed in Table 4 were calculated by the least-squares deviations using eq 3. The values of the standard deviation (SD) and the average deviation (AD) were calculated by eqs 4 and 5, respectively.

$$\mathrm{SD} = \left[\sum_{i=1}^{p} (y_{\exp,i} - y_{\mathrm{cal},i})^2/(p-q)\right]^{1/2} \tag{4}$$

$$\mathrm{AD} = \frac{1}{p}\sum_{i=1}^{p}\left|\frac{y_{\exp,i} - y_{\mathrm{cal},i}}{y_{\exp,i}}\right| \tag{5}$$

where p is the total number of experimental data point and q is the number of parameters. $y_{\exp,i}$ and $y_{\mathrm{cal},i}$ refer to the experimental values and the calculated values from the equation, respectively.

It could be seen from Table 3 that the maximum values of AD and SD between calculated densities and experimental data are 0.002 and 0.002 $g \cdot cm^{-3}$, respectively. Then it could be observed from Table 4 that for viscosities of benzene-1,3-diol + water, the maximum values of AD and SD are 0.003 and 0.01 mPa·s, respectively; and for viscosities of benzene-1,3-diol + ethanol the AD and SD are 0.04 and 0.2 mPa·s; for benzene-1,3-diol + propan-1-ol 0.02 and 0.2 mPa·s; for benzene-1,3-diol + butan-1-ol 0.02 and 0.1 mPa·s. Calculated results indicated that the Jones-Dole equation is applicable for the correlation of the viscosity data of benzene-1,3-diol + water, ethanol, propan-1-ol, and butan-1-ol binary solutions.

■ CONCLUSIONS

Owing to the unique molecular structure, benzene-1,3-diol has attracted increasing interest of researchers to develop its derivative products in fine chemical field. As a prerequisite, some basic property data for materials are very necessary for related studies and product exploitations. In this work, the densities and viscosities of benzene-1,3-diol + water, ethanol, propan-1-ol and butan-1-ol binary solutions at T = (293.15 to 333.15) K were measured experimentally. The influences of temperature and concentration of benzene-1,3-diol on the densities and viscosities of benzene-1,3-diol solutions were studied. Furthermore, the densities of benzene-1,3-diol solutions were fitted using the Gonzáles equation, and the relative viscosities of benzene-1,3-diol solutions were correlated using the extended Jones-Dole equation, and the parameters in corresponding equations were obtained respectively.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: zhchy971@tju.edu.cn. Fax: +86-22-27404772.

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