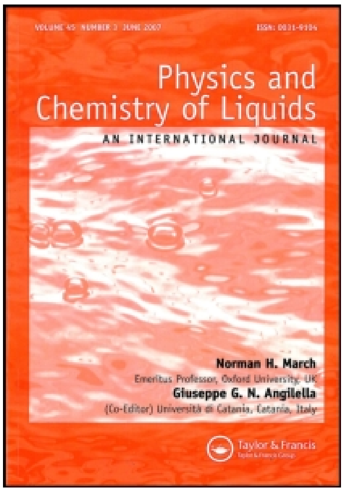

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Effect of temperature on partial molar volumes and viscosities of dilute aqueous solutions of 1-butanol, 1,2-butanediol, 1,4-butanediol, 1,2,4-butanetriol, and butanetetrol

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Experimental densities and viscosities of dilute aqueous binary mixtures of 1-butanol, 1,2-butanediol, 1,4-butanediol, 1,2,4-butanetriol, and 1,2,3,4-butanetetrol (erythritol) were measured in the temperature range from 283.15 to 323.15 K. Apparent molar volumes were determined as a function of composition from density measurements. The limiting partial molar volumes $\overline{V_2^\infty}$ of alcohols in aqueous solution were evaluated through extrapolation. The relative viscosity values were adjusted by least-squares to a second order equation to obtain the viscosity *B* coefficient which has been related to the size and shape of the solute molecule. The temperature dependence of the partial molar volume at infinite dilution and the viscosity *B* coefficient are discussed in terms of the relationship among polar and non polar groups on water structure and the effect of the position of hydroxyl groups in the molecule.

Keywords: Partial molar volumes; Viscosity; Molecular interactions; Alcohols; Polyols

1. Introduction

Thermodynamic properties of highly dilute aqueous solutions of organic compounds are of considerable importance in the analysis of hydrophobic and hydrophilic interactions between these solutes and water and their effect on solvent structure.

Although there is much literature about volumes and viscosities of aqueous alcohol mixtures [1–19], less work has been reported on alkanediols and polyols-water systems [20–32].

Some studies suggest that the thermodynamic behavior of these solutes depends on the number and position of hydroxyl groups and the effect that these factors exert on changes in the hydrophobic–hydrophilic balance and the possibility of inter and intramolecular hydrogen bonding. In a previous work the influence of the relative

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positions of OH groups on the volumetric properties of four carbon atoms alcohols and polyols in dilute aqueous solution at 298.15 K was reported. The effect of the increase in the number of OH groups as well as their position on infinite partial molar volume is examined in a systematical way [19].

This article presents the effect of temperature on partial molar volumes and viscosities of dilute aqueous solutions of 1-butanol (1BuOH), 1,2-butanediol (12BD), 1,4-butanediol (14BD), 1,2,4-butanetriol (124BT), and butanetetrol (1234BT). The data reported here were obtained performing density and viscosity measurements in the temperature range from 283.15 to 323.15 K. The temperature dependence of the partial molar volume at infinite dilution and the viscosity *B* coefficient is discussed in terms of the balance between hydrophobic and hydrophilic interactions between alcohols and polyols in water and their effect on solvent structure.

2. Experimental

The materials used were the following: 1-butanol (J.T. Baker), 1,2-butanediol 98% (Aldrich), 1,4-butanediol 99% (Aldrich), (±)1,2,4-butanetriol 96% (Aldrich), and 1,2,3,4-butanetretol $\geq$99% (Sigma). The alcohols and liquid polyols were degassed before use. Water content was determined by the Karl Fischer method. Water was doubly distilled and deionized according to literature and degassed before use [33]. All the solutions were prepared by weight using a Mettler balance AT-261 dual range with sensitivity of 10^{-5} g in the lower range. Final concentrations were corrected according to water content analysis.

Solution densities were measured using pycnometers of the Wood-Brusie type with a bulb volume of $50\,\mathrm{cm}^3$. Pycnometers were calibrated at all temperatures with water. All measurements were realized in a constant temperature bath with temperature controlled to ±0.05 K. with an uncertainty of $\pm 5\cdot 10^{-5}\,\mathrm{g\,cm}^{-3}$. Density values are the average of three independent measurements.

The density data obtained for each solution were used to calculate the apparent molar volume ϕ_v using the equation:

$$\phi_\mathrm{v} = \frac{M}{d} + \frac{1000(d_\mathrm{o} - d)}{mdd_\mathrm{o}} \tag{1}$$

where M is the molecular weight of the solute, m its molal concentration, d, and d_o are the densities of the solution and the solvent, respectively.

Apparent molar volumes were fitted by least-squares to obtain by extrapolation the infinite dilution apparent molar volume $\Phi^\mathrm{o}_\mathrm{v}$ which at infinite dilution equals $\bar{V}^\mathrm{o}$ the infinite dilution partial molar volume. The estimated uncertainty for infinite dilution partial molar volumes is $0.05\,\mathrm{cm}^3\,\mathrm{mol}^{-1}$.

Viscosity was determined using two modified suspended-level Ubbelohde capillary viscometers with efflux times ~300 s for water and were calibrated with water at the selected temperatures. The viscometers were held in a thermostated bath with temperature controlled to ±0.05 K. Reproducibility of efflux times was in all cases better than 0.05%. Viscosity values are the average of at least five independent measurements.

The viscosity data were obtained from the relation:

$$\eta = \alpha d t - \beta \frac{d}{t} \tag{2}$$

where α and β are the viscometer constants, d is the density, and t the efflux time. The relative viscosities $\eta_r = \eta/\eta_o$ were calculated from the solution and solvent viscosities, respectively.

3. Results and discussion

Tables 1–5 present the experimental results for solution density, solute apparent molar volume, and viscosity as a function of alcohol mole fraction at each temperature. For all systems, apparent molar volumes of alcohols and polyols increase with temperature.

Table 1. Densities (d), viscosities (η), and apparent molar volumes (ϕ), for dilute aqueous solutions of 1-Butanol as a function of temperature.

	d(g cm^{-3})			ϕ_V (cm^3 mol^{-1})			η (mPa s)		
	X_{OH}			X_{OH}			X_{OH}		
T(K)	0.0050	0.0100	0.0150	0.0050	0.0100	0.0150	0.0050	0.0100	0.0150
278.15	0.99730	0.99537	0.99373	83.77	82.57	81.88	1.641	1.840	2.020
283.15	0.99693	0.99476	0.99360	84.18	83.24	81.73	1.439	1.554	1.673
288.15	0.99629	0.99427	0.99275	84.38	83.09	82.11	1.235	1.333	1.432
293.15	0.99526	0.99287	0.99098	84.97	84.12	83.31	1.077	1.155	1.232
298.15	0.99395	0.99140	0.98923	85.64	84.82	84.17	0.947	1.003	1.058
303.15	0.99245	0.98955	0.98732	86.15	85.82	84.96	0.842	0.883	0.930
308.15	0.99064	0.98755	0.98502	87.06	86.69	86.01	0.758	0.794	0.841
313.15	0.98860	0.98545	0.98276	88.05	87.42	86.76	0.682	0.712	0.746
318.15	0.98648	0.98327	0.98046	88.70	87.97	87.35	0.620	0.638	0.662
323.15	0.98422	0.98103	0.97813	89.25	88.32	87.78	0.566	0.580	0.602

Table 2. Densities (d), viscosities (η), and apparent molar volumes (ϕ), for dilute aqueous solutions of 1,2-Butanediol as a function of temperature.

	d (g cm^{-3})				ϕ_V (cm^3 mol^{-1})				η (mPa s)			
	X_{OH}				X_{OH}				X_{OH}			
T(K)	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200
278.15	1.00130	1.00268	1.00403	1.00535	84.99	84.94	84.90	84.86	1.735	1.923	2.110	2.310
283.15	1.00106	1.00242	1.00376	1.00516	85.15	85.06	85.00	84.86	1.461	1.601	1.791	1.913
288.15	1.00041	1.00177	1.00312	1.00453	85.39	85.21	85.10	84.94	1.251	1.381	1.502	1.626
293.15	0.99945	1.00069	1.00196	1.00323	85.70	85.62	85.50	85.40	1.087	1.188	1.285	1.372
298.15	0.99821	0.99940	1.00056	1.00173	86.09	85.96	85.91	85.83	0.964	1.034	1.129	1.204
303.15	0.99673	0.99782	0.99890	1.00000	86.51	86.42	86.35	86.27	0.865	0.919	0.978	1.052
308.15	0.99507	0.99615	0.99720	0.99831	86.82	86.65	86.60	86.47	0.783	0.824	0.874	0.929
313.15	0.99320	0.99429	0.99536	0.99651	87.16	86.89	86.78	86.60	0.689	0.732	0.780	0.844
318.15	0.99114	0.99218	0.99331	0.99446	87.54	87.26	87.01	86.82	0.632	0.669	0.714	0.761
323.15	0.98880	0.98967	0.99080	0.99186	88.34	88.08	87.62	87.42	0.572	0.600	0.644	0.686

Table 3. Densities (d), viscosities (η), and apparent molar volumes (ϕ), for dilute aqueous solutions of 1,4-Butanediol as a function of temperature.

	d (g cm^{-3})				ϕ_V (cm^3 mol^{-1})				η (mPa s)			
	X_{OH}				X_{OH}				X_{OH}			
T(K)	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200
278.15	1.00062	1.00144	1.00233	1.00340	87.60	87.25	87.00	86.67	1.740	1.951	2.201	2.411
283.15	1.00032	1.00104	1.00179	1.00281	87.73	87.51	87.37	87.01	1.515	1.715	1.924	2.210
288.15	0.99969	1.00038	1.00120	1.00203	87.90	87.67	87.41	87.24	1.298	1.447	1.660	1.868
293.15	0.99879	0.99947	1.00023	1.00105	88.00	87.78	87.58	87.39	1.126	1.272	1.460	1.616
298.15	0.99764	0.99832	0.99909	0.99988	88.09	87.87	87.66	87.50	0.970	1.075	1.193	1.314
303.15	0.99624	0.99693	0.99773	0.99852	88.19	87.97	87.73	87.59	0.860	0.960	1.067	1.191
308.15	0.99463	0.99532	0.99606	0.99695	88.34	88.11	87.93	87.68	0.779	0.863	0.942	1.068
313.15	0.99280	0.99349	0.99425	0.99518	88.52	88.28	88.08	87.79	0.720	0.770	0.853	0.958
318.15	0.99078	0.99147	0.99227	0.99321	88.76	88.50	88.23	87.93	0.643	0.700	0.758	0.845
323.15	0.98858	0.98925	0.99012	0.99103	89.05	88.77	88.38	88.12	0.586	0.626	0.674	0.737

Table 4. Densities (d), viscosities (η), and apparent molar volumes (ϕ), for dilute aqueous solutions of 1,2,4-Butanetriol as a function of temperature.

	d (g cm^{-3})				Φ_V (cm^3 mol^{-1})				η (mPa s)			
	X_{OH}				X_{OH}				X_{OH}			
T(K)	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200
278.15	1.00513	1.01016	1.01516	1.02011	87.57	87.06	86.85	86.61	1.763	1.969	2.176	2.400
283.15	1.00475	1.00976	1.01465	1.01950	87.60	87.33	87.17	86.95	1.514	1.714	1.924	2.170
288.15	1.00409	1.00907	1.01388	1.01864	87.87	87.60	87.42	87.24	1.320	1.470	1.660	1.880
293.15	1.00317	1.00810	1.01285	1.01754	88.03	87.74	87.65	87.50	1.089	1.271	1.460	1.617
298.15	1.00199	1.00688	1.01158	1.01622	88.16	87.93	87.87	87.74	0.984	1.096	1.193	1.328
303.15	1.00059	1.00542	1.01008	1.01468	88.30	88.15	88.09	87.97	0.889	0.968	1.067	1.195
308.15	0.99896	1.00374	1.00837	1.01293	88.47	88.39	88.33	88.21	0.798	0.863	0.942	1.058
313.15	0.99712	1.00185	1.00646	1.01099	88.67	88.65	88.57	88.46	0.700	0.769	0.853	0.937
318.15	0.99509	0.99978	1.00437	1.00887	88.99	88.93	88.84	88.71	0.656	0.703	0.770	0.857
323.15	0.99287	0.99753	1.00210	1.00659	89.22	89.22	89.11	88.97	0.595	0.626	0.678	0.732

Table 5. Densities (d), viscosities (η), and apparent molar volumes (η), for dilute aqueous solutions of 1,2,3,4-Butanetetrol as a function of temperature.

	d (g cm^{-3})				ϕ_V (cm^3 mol^{-1})				η (mPa s)			
	X_{OH}				X_{OH}				X_{OH}			
T(K)	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200	0.0050	0.0100	0.0150	0.0200
278.15	1.00996	1.01986	1.02946	1.03891	85.34	84.87	84.64	84.37	1.848	2.037	2.187	2.371
283.15	1.00974	1.01954	1.02914	1.03859	85.20	84.97	84.72	84.43	1.582	1.742	1.877	2.020
288.15	1.00890	1.01905	1.02890	1.03858	86.10	84.80	84.31	83.91	1.368	1.504	1.626	1.772
293.15	1.00795	1.01767	1.02745	1.03741	86.33	85.74	85.03	84.21	1.176	1.312	1.420	1.530
298.15	1.00659	1.01602	1.02549	1.03514	87.15	86.70	86.09	85.30	1.050	1.152	1.250	1.327
303.15	1.00500	1.01445	1.02390	1.03340	87.92	87.09	86.40	85.70	0.915	1.021	1.109	1.191
308.15	1.00290	1.01210	1.02127	1.03050	89.78	88.54	87.74	86.99	0.790	0.912	0.968	1.053
313.15	1.00090	1.00996	1.01920	1.02830	90.58	89.24	88.17	87.45	0.690	0.821	0.891	0.965
318.15	0.99889	1.00771	1.01691	1.02670	90.72	89.82	88.64	87.18	0.649	0.744	0.808	0.860
323.15	0.99640	1.00510	1.01412	1.02360	92.02	90.77	89.54	88.17	0.620	0.679	0.736	0.813

In the dilute region apparent molar volumes for 1-butanol, 1,2-butanediol, 1,4-butanediol, and 1,2,3,4-butanetetrol (erythritol) decrease with increasing concentration and decrease as temperature increases. As expected, viscosity increases as alcohol concentration increase and decrease with temperature. The observed trends agree with the experimental behavior reported for similar systems.

Table 6 shows the results for the partial molar volumes of alcohols in water at infinite dilution obtained as the extrapolated value of the adjusted data by least squares of partial molar volumes. Literature values are reported for comparison. No results have been found in literature for 1,2,3-butanetriol at temperatures different from 298.15 K.

The results obtained in this work for infinite dilution partial molar volumes of alcohols and polyols agree well with literature values when they are available. The small differences can be attributed to differences in the concentration range used to make extrapolation and the resulting differences in least squares fitting of experimental data. For all the solutes considered in this work volume increases with temperature. For 1,2-butanediol the change with temperature is larger than in the case of 1,4-butanediol.

The results show that at constant temperature the addition of OH groups does not cause sensible changes in volumetric properties and its contribution does not present a clear dependence on position of OH groups [5,19,27,28]. At all temperatures, the highest value for the infinite dilution partial molar volumes is for 1,4-butanediol in agreement with results presented by other authors who have studied this system [3,5,29]. The values obtained are very similar to those for 1,2,4-butanetriol. In the case of diols, the difference between the infinite dilution partial molar volumes is related to the relative position of the OH groups.

The relative viscosity values were adjusted by least-squares to a second order equation. In this equation B and D are empirical coefficients however, as proposed by

Tabla 6. Infinite dilution partial molar volume of 1-Butanol. 1.2-Butanediol. 1.4-Butanediol. 1.2.4-Butanetriol and 1.2.3.4-Butanetetrol in aqueous solution in the temperature range from 283.15 to 323.15 K.

	$\overline{V_2^0}$(cm^3 mol^{-1})				
T(K)	1-butanol	1,2-butanediol	1,4-butanediol	1,2,4-butanetriol	1,2,3,4-butanetetrol
278.15	84.63 86.00[3]	85.02	87.88 87.81[3] 87.81[7]	87.79	85.01
283.15	85.11 86.25[3]	85.22	87.99 88.00[3]	87.92	85.46
288.15	85.47	85.53	88.12	88.03	86.54
293.15	85.79 86.41[3]	85.80	88.19 88.19[3]	88.15	87.09
298.15	86.34 86.4[31] 86.48[5] 86.23[19]	86.16 86.32[29] 86.37[19]	88.27 88.23[5] 88.36[7] 88.30[32]	88.26 88.15[19]	87.84 87.68[19] 87.61[27]
303.15	86.82	86.58	88.38	88.38	88.62
308.15	87.63	86.91	88.55	88.55	90.19
313.15	88.64 87.61[3]	87.31	88.77 88.91[3]	88.77	91.48
318.15	89.31	87.66	89.05 89.18[7]	89.10	92.09
323.15	89.92	88.67	89.37	89.4	93.31

Table 7. Viscosity B coefficient of 1-butanol. 1,2-butanediol. 1,4-butanediol. 1,2,4-butanetriol and 1,2,3,4-butanetetrol in aqueous solution in the temperature range from 283.15 to 323.15 K.

	Viscosity B coefficients (kg mol^{-1})				
T(K)	1-butanol	1,2-butanediol	1,4-butanediol	1,2,4-butanetriol	1,2,3,4-butanetetrol
278.15	0.4602	0.5033	0.5383	0.5584	0.7571
283.15	0.3713	0.4428	0.4899	0.5278	0.7033
288.15	0.3038	0.3792	0.4364	0.4754	0.6426
293.15	0.2699	0.3381	0.3806	0.4293	0.6230
298.15	0.238	0.2897	0.3252	0.3639	0.5799
303.15	0.1905	0.2599	0.2858	0.3221	0.5567
308.15	0.1711	0.2358	0.2527	0.2869	0.5311
313.15	0.1447	0.1898	0.2411	0.2663	0.4968
318.15	0.1227	0.1634	0.2272	0.2527	0.4361
323.15	0.1143	0.1310	0.2075	0.2383	0.4176

Tsangaris-Martin for mixed solutes as amino acids [34] and has the same form of the Einstein equation for the viscosity of non electrolyte solutions, when D coefficient is neglected as expressed by Zhao [35]. Spite the empirical character of B the coefficient, it has been assumed that it depends on the size and shape of the solute molecule and is related to solute–solvent interactions.

$$\eta_{\mathrm{r}} = 1 + Bm + Dm^2. \tag{3}$$

The viscosity B coefficients are presented in table 7. At each temperature the coefficients increase with the number of OH groups and decrease with temperature. In the case of diols the coefficient for 1,4-butanediol is considerably larger than the coefficient of 1,2-butanediol.

Several properties have been used to interpret the effect of solutes on water structure. The negative sign of the limiting slope $(\partial \bar{V}_2/\partial X_2)_T$ has been used by several authors as indicative of hydrophobic behavior [36]. In this case all the limiting slopes are negative. The highest value is for butanol showing that 1-butanol is the solute that presents the most hydrophobic behavior which diminishes as the number of OH groups increases.

The temperature dependence of the partial molar volume at infinite dilution is used to interpret the effect of the hydrocarbon chain on water structure according to the general criteria proposed by Hepler about solute effect on water structure [37]. According to this general criterion, the behavior of the second derivative of the infinite dilution partial molar volume with temperature is related to the effect of the solute on water structure. In this way if $(\partial^2 V^0/\partial T^2) < 0$ the solute has a water structure breaking character, but if the behavior shows that $(\partial^2 V^0/\partial T^2) > 0$ the solute has water structure forming effect.

The effect of temperature on volumetric behavior is of 1-butanol, 1,2-butanediol, 1,4-butanediol, 1,2,4-butanetriol, and butanetetrol in aqueous solution is well described by a polynomial second order equation. The second derivative has a very small positive value for all the solutes considered in this work indicating that they exert a forming effect on water structure. However, the calculated values are smaller than the estimated uncertainty for infinite dilution partial molar volumes.

The first derivative of the *B* coefficient with temperature has also been used as indicative of the effect of solute on water structure [35]. The negative sign has been attributed to a structure making effect while the positive sign to a structure breaking effect. For all the solutes considered, dB/dT shows a negative value at 298.15 K suggesting that they have a water structure making character in accordance with the results from volumetric behavior.

The different behavior observed for 1,2-butanediol and for 1,4-butanediol can be attributed to the strength of H bond and to the ability to form inter or intramolecular H bonds. The most stable conformation for vicinal diols such as 1,2-butanediol in which free rotation is allowed, is the gauche. For the pure diol there is no evidence of high electron density along the internuclear axis connecting the H donor and the O acceptor atoms [38]. Under this conditions it is clear that intramolecular hydrogen bonds can not be formed between pure 1,2-butanediol molecules and is possible that in dilute aqueous solution 1,2-butanediol would form intermolecular hydrogen bonds with water. In the case of 1,4-butanediol electron density concentration around the internuclear axis is observed indicating the formation of intramolecular hydrogen bonds.

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