Densities and Viscosities of Glycine, DL-α-Alanine, DL-α-Amino-*n*-butyric Acid, and L-Leucine in Aqueous 1,2-Propanediol Solutions at 298.15 K

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Densities, ρ, and viscosities, η, of glycine, DL-α-alanine, DL-α-amino-*n*-butyric acid, and L-leucine in 1.0, 2.0, 4.0, and 6.0 mol·kg^{-1} aqueous 1,2-propanadiol solutions have been determined at 298.15 K. The partial molar volumes, V_2^∞, obtained from densities have been used to calculate the corresponding volumes of transfer, $\Delta_t V^\infty$, at infinite dilution from water to different concentrations of aqueous 1,2-propanediol solutions for rationalizing various interations. The Jones−Dole equation has been fitted to the viscosity data to calculate *B*-coefficients. Results show that in the case of glycine and DL-α-alanine the ion−dipolar interactions are dominating while hydrophobic−hydrophobic and hydrophilic−hydrophobic interactions are also showing their predominant role in other cases.

Introduction

The volumetric and transport properties of amino acids in an electrolyte and organic aqueous solutions are required to understand the amino acid−ion−solvent and amino acid−solvent interactions.[1−9] In continuation of our earlier studies of some amino acids in different solvents,[3,8] we now systematically investigate the effect of increasing alkyl side chain of amino acids (glycine, DL-α-alanine, DL-α-amino-*n*-butyric acid, and L-leucine) in aqueous 1,2-propanediol solutions. The apparent molar volumes, $V_{2,\phi}$, and viscosities, η, of amino acids have been determined in water and in aqueous 1,2-propanediol solutions (1.0, 2.0, 4.0, and 6.0) mol·kg^{-1} at 298.15 K by measuring the densities using a vibrating tube digital densimeter and an Ubbelohde viscometer, respectively.

Experimental Section

Glycine, DL-α-alanine, DL-α-amino-*n*-butyric acid, and L-leucine of highest purity (99%) were obtained from Sigma Chemical Co. These, along with 1,2-propanediol (AR, Thomas Baker) were used without further purification. The solutions were prepared in deionized, doubly distilled water which was degassed immediately prior to use. All solutions were prepared by mass using a Mettler Balance having an accuracy of ±0.01 mg.

The densities of the solutions were measured using a vibrating-tube digital densimeter (Model DMA 60/602 Anton Paar, Austria). The working of the densimeter was checked by measuring the densities of aqueous sodium chloride solutions, which agreed within ±0.5% with the literature values.[10] The densities were accurate to 3×10^{-6} g·cm^{-3} while the precision was estimated to be 1×10^{-6} g·cm^{-3}, as obtained from triplicate measurements. Viscosities of the solutions were measured using an Ubbelohde type viscometer which was calibrated using the efflux time of water from 298.15 to 318.15 K. Efflux time was measured with an electronic stop watch with a resolution of ±0.01 s, as an average of at least four readings of the flow time. The viscosities of solutions were calculated by the following expression

$$\eta/\rho = at - b/t$$

where ρ is the density of the solution and t is efflux time. The a and b are the viscometer constants obtained from the efflux time of water at different temperatures. The measured viscosity values are accurate up to ±0.001 mPa·s. The temperature of water flowing around the densimeter cell and viscometer was controlled within ±0.01 K.

Results and Discussion

Densities of amino acids in aqueous 1,2-propanediol solutions at 298.15 K as a function of molality are summarized in Table 1. Apparent molar volumes of amino acids, $V_{2,\phi}$, were calculated as

$$V_{2,\phi} = M/\rho - [(\rho - \rho_0)1000/m\rho\rho_0] \quad (1)$$

where M is the molar mass of the amino acid, m is molality, and ρ and ρ_0 are the densities of solution and solvent, respectively. An analogous equation is also applicable for calculating the apparent molar volumes of 1,2-propanediol, $V_{2,\phi,\mathrm{pd}}$, in aqueous amino acids with ρ_0 as the density of amino acid in water at different molalities (m_s/mol·kg^{-1}) of 1,2-propanediol. The $V_{2,\phi,\mathrm{pd}}$ values of the cosolute vary nonlinearly for glycine and DL-α-alanine at $m_s = 1.0$ (only representative plots of $V_{2,\phi,\mathrm{pd}}$ vs m_s are shown in Figures 1 and 2). However, the variation is linear up to ≃0.4 mol·kg^{-1} for both amino acids and then becomes nonlinear. At higher concentration of cosolute, the linear dependence has been observed in all the cases but the variation in the magnitude decreases. In the case of L-leucine at higher concentration of 1,2-propanediol, the $V_{2,\phi,\mathrm{pd}}$ is almost independent of

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Table 1. Densities, ρ, for Some Amino Acids in Aqueous 1,2-Propanediol Solutions as a Function of Molalities of Amino Acids and 1,2-Propanediol at 298.15 K

m	ρ	m	ρ	m	ρ
mol·kg^{-1}	g·cm^{-3}	mol·kg^{-1}	g·cm^{-3}	mol·kg^{-1}	g·cm^{-3}
		Glycine			
		m_s[a]/mol·kg^{-1} = 1.0			
0.075 24	1.004 737	0.281 03	1.011 078	0.614 55	1.020 903
0.118 79	1.006 097	0.425 06	1.015 377	0.696 48	1.023 255
0.200 71	1.008 634	0.544 87	1.018 892		
		m_s/mol·kg^{-1} = 2.0			
0.114 98	1.010 672	0.271 28	1.015 381	0.356 75	1.017 923
0.163 50	1.012 146	0.312 59	1.016 619	0.400 10	1.019 191
0.211 78	1.013 599				
		m_s/mol·kg^{-1} = 4.0			
0.071 12	1.017 849	0.197 86	1.021 587	0.557 23	1.031 866
0.101 50	1.018 750	0.309 51	1.024 831	0.746 01	1.037 045
		m_s/mol·kg^{-1} = 6.0			
0.079 71	1.024 127	0.420 67	1.033 803	0.631 96	1.039 545
0.331 41	1.031 308	0.506 75	1.036 162	0.675 93	1.040 718
		DL-α-Alanine			
		m_s/mol·kg^{-1} = 1.0			
0.199 07	1.007 829	0.399 77	1.013 296	0.642 94	1.019 672
0.298 78	1.010 570	0.528 69	1.016 706	0.690 03	1.020 879
		m_s/mol·kg^{-1} = 2.0			
0.091 80	1.011 076	0.296 47	1.016 679	0.602 29	1.024 681
0.205 09	1.014 199	0.412 03	1.019 750	0.679 01	1.026 627
		m_s/mol·kg^{-1} = 4.0			
0.092 39	1.020 808	0.349 86	1.027 592	0.499 15	1.031 385
0.242 49	1.024 812	0.428 19	1.029 602		
		m_s/mol·kg^{-1} = 6.0			
0.151 30	1.026 587	0.264 24	1.029 517	0.359 06	1.031 916
0.210 09	1.028 127	0.305 19	1.030 559	0.459 89	1.034 426
		DL-α-Amino-*n*-butyric Acid			
		m_s/mol·kg^{-1} = 1.0			
0.069 96	1.005 626	0.163 61	1.008 163	0.198 71	1.009 088
0.130 54	1.007 270	0.176 02	1.008 495		
		m_s/mol·kg^{-1} = 2.0			
0.105 96	1.010 889	0.143 58	1.011 907	0.227 10	1.014 122
0.118 59	1.011 229	0.206 94	1.013 590		
		m_s/mol·kg^{-1} = 4.0			
0.097 87	1.020 061	0.135 94	1.021 050	0.205 80	1.022 834
0.120 79	1.020 657	0.177 59	1.022 117		
		m_s/mol·kg^{-1} = 6.0			
0.091 42	1.024 723	0.125 50	1.025 582	0.165 20	1.026 559
0.104 61	1.025 055	0.132 16	1.025 737		
		L-Leucine			
		m_s/mol·kg^{-1} = 1.0			
0.023 22	1.006 420	0.058 47	1.007 250	0.078 42	1.007 711
0.051 78	1.007 094	0.070 54	1.007 530		
		m_s/mol·kg^{-1} = 2.0			
0.022 99	1.007 484	0.057 19	1.008 284	0.080 28	1.008 830
0.035 03	1.007 769	0.066 77	1.008 513	0.095 96	1.009 203
0.044 53	1.007 988	0.071 54	1.008 616		
		m_s/mol·kg^{-1} = 4.0			
0.025 72	1.017 457	0.041 41	1.017 797	0.054 46	1.018 076
0.035 81	1.017 676	0.048 41	1.017 949		
		m_s/mol·kg^{-1} = 6.0			
0.023 80	1.023 039	0.037 48	1.023 320	0.048 16	1.023 527
0.028 54	1.023 132	0.039 93	1.023 369	0.055 62	1.023 681
0.030 25	1.023 169	0.039 98	1.023 364		

[a] m_s = molality of 1,2-propanediol in water.

concentration of amino acid. At infinite dilution the apparent molar volumes, $V_{2,\phi}^{\infty}$, and partial molar volumes, V_2^{∞}, are identical ($V_{2,\phi}^{\infty} = V_2^{\infty}$). In the case of negligible concentration dependence of $V_{2,\phi}$ (within the uncertainty limits of measurements), V_2^{∞} was determined by taking the average of all the data points. However, where finite concentration dependence was observed, V_2^{∞} was determined by least-squares fitting of the data by the following equation

$$V_{2,\phi} = V_2^{\infty} + S_v m \tag{2}$$

where S_v is the limiting slope. The S_v and V_2^{∞} values with their standard deviations have been summarized in Table 2. The experimental values of V_2^{∞} for the above studied amino acids in water agreed well with those reported in the literature values.[1,6] The standard partial molar volumes of transfer, $\Delta_t V^{\infty}$, of a particular amino acid from water to aqueous cosolute solution at infinite dilution have been estimated as follows

$$\Delta_t V^{\infty} = V_2^{\infty}(\text{in aqueous cosolute}) - V_2^{\infty}(\text{in water}) \tag{3}$$

The values for $\Delta_t V^{\infty}$ are summarized in Table 2, and plots of $\Delta_t V^{\infty}$ versus m_s have been illustrated in Figure 3. Both positive and negative values of $\Delta_t V^{\infty}$ have been observed for the amino acids studied. Positive values of $\Delta_t V^{\infty}$ have been observed for glycine and DL-α-alanine over the entire concentration range of cosolute and increase with the increase in the concentration of cosolute. The magnitude of $\Delta_t V^{\infty}$ is slightly more in the case of glycine than in the case of DL-α-alanine. The $\Delta_t V^{\infty}$ value for L-leucine is negative at lower concentration and passes through the minimum at ≃1.5 mol·kg^{-1}, having $\Delta_t V^{\infty} = -1.0$ cm^3·mol^{-1}, and then increases and becomes positive after ≃4.0 mol·kg^{-1} of cosolute. For DL-α-amino-*n*-butyric acid, values of $\Delta_t V^{\infty}$ also pass through the minimum at ≃1.5 mol·kg^{-1} and remain negative throughout the concentration range of the cosolute. We have also reported $\Delta_t V^{\infty}$ values of some amino acids from water to aqueous glycerol solutions at 298.15 K, and the behavior is almost similar.[8] Mishra and Ahluwalia[11] have reported the enthalpies, heat capacities, and apparent molar volumes of transfer of some amino acids from water to aqueous *tert*-butyl alcohol solutions. The behavior of apparent molar volumes of transfer at 0.1 mol·kg^{-1} concentration of amino acids in aqueous solutions of *tert*-butyl alcohol shows a minimum for the studied amino acids except for glycine which lies around 0.03 mole fraction of *tert*-butyl alcohol. The negative values of $\Delta_t V^{\infty}$ around this mole fraction increase with the increase in side chain length of the amino acids, which is similar to the case observed in aqueous glycerol[8] and 1,2-propanediol solutions.

The V_2^{∞} values of the studied α-amino acids in water and in cosolute solution vary linearly with the number of carbon atoms (n_c) in their alkyl side chains at 298.15 K. A similar behavior of V_2^{∞} has been reported for some homologous series of α- or ω-amino acids in aqueous potassium thiocyanate[12] or guanidine hydrochloride solutions.[13] The alkyl side chains of the homologous series of α-amino acids investigated in this work are as follows: CH_2- (glycine), CH_3CH- (DL-α-alanine), CH_3CH_2CH- (DL-α-amino-*n*-butyric acid), and $(CH_3)_2CHCH_2CH-$ (L-leucine). A linear regression analysis of the V_2^{∞} values versus number of carbon atoms in water and in cosolute solutions was carried out by using the following equation

$$V_2^{\infty} = V_2^{\infty}(NH_3^+, COO^-) + n_c V_2^{\infty}(CH_2) \tag{4}$$

where $V_2^{\infty}(NH_3^+, COO^-)$ and $V_2^{\infty}(CH_2)$ represent the zwitterionic end groups and the methylene group contributions, respectively. The results are given in Table 3. The contributions of the methylene group and zwitterionic groups to

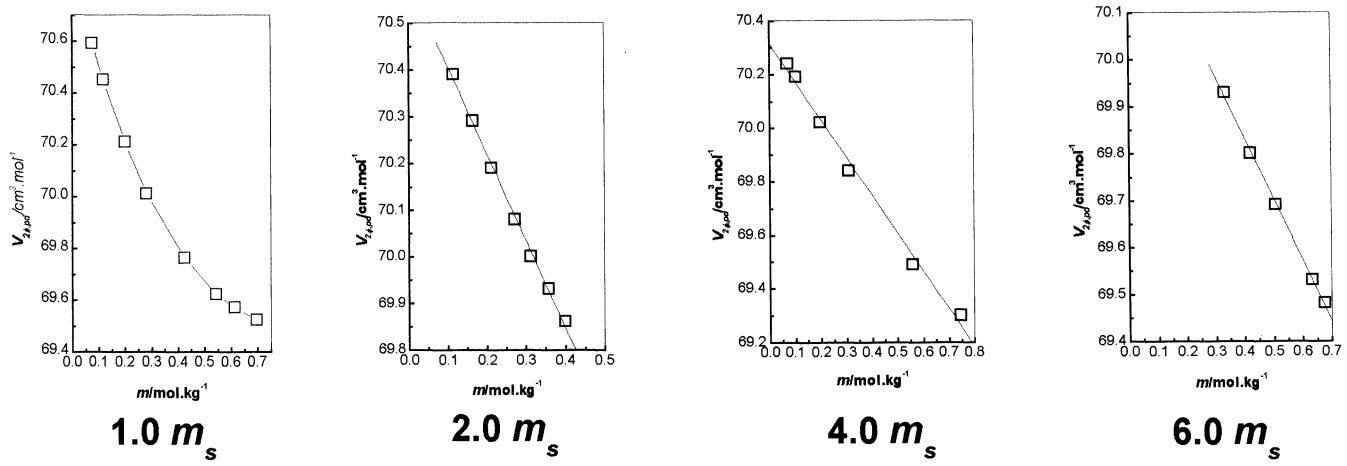

Figure 1. Apparent molar volumes, $V_{2,\phi,\mathrm{pd}}$, of 1,2-propanediol (m_s = 1.0, 2.0, 4.0, or 6.0) vs molality, m, of glycine at 298.15 K.

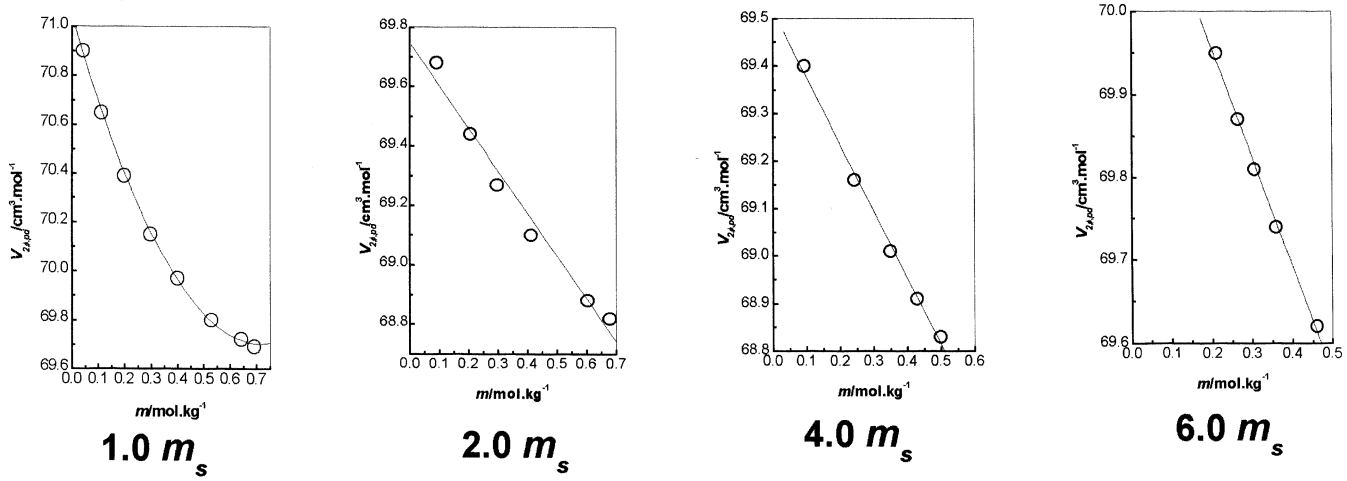

Figure 2. Apparent molar volumes,$V_{2,\phi,\mathrm{pd}}$, of 1,2-propanediol (m_s = 1.0, 2.0, 4.0, or 6.0) vs molality, m, of alanine at 298.15 K.

Table 2. Standard Partial Molar Volumes, V_2^∞, and Volumes of Transfer, $\Delta_t V_2^\infty$, at Infinite Dilution for Some Amino Acids in Aqueous 1,2-Propanediol Solutions at 298.15 K

	V_2^∞/cm^3·mol^{-1} at various m_s[a]/mol·kg^{-1}				$\Delta_t V_2^\infty$/cm^3·mol^{-1} at various m_s[a]/mol·kg^{-1}			
amino acid	1.0	2.0	4.0	6.0	1.0	2.0	4.0	6.0
glycine	43.43 ± 0.01[b] (1.05)	43.96 ± 0.01 (0.81)	44.76 ± 0.01 (0.05)	45.33 ± 0.01 (0.72)	0.25	0.78	1.58	2.15
DL-α-alanine	60.49 ± 0.01 (0.72)	60.51 ± 0.01 (0.65)	61.08 ± 0.01 (1.03)	61.31 ± 0.01 (1.47)	0.03	0.05	0.62	0.85
DL-α-amino-*n*-butyric acid	75.08 ± 0.01 (1.47)	75.84 ± 0.01 (1.50)	74.41 ± 0.003 (1.77)	75.51 ± 0.01 (4.00)	−0.59	−0.83	−0.26	−0.16
L-leucine	106.39 ± 0.01 (6.69)	106.62	107.13 ± 0.01 (10.25)	108.29	−1.14	−0.91	−0.40	0.76

[a] m_s = molality of 1,2-propanediol in water. [b] Standard deviations; parentheses contain S_V/cm^3·mol^{-2}·kg.

Table 3. Contribution of (NH_3^+, COO^-) and R Groups to the Standard Partial Molar Volumes, V_2^∞, and Volumes of Transfer, $\Delta_t V_2^\infty$, for Some Amino Acids from Water to Aqueous 1,2-Propanediol Solutions at 298.15 K

	V_2^∞/cm^3·mol^{-1} at various m_s[a]/mol·kg^{-1}					$\Delta_t V_2^\infty$/cm^3·mol^{-1} at various m_s[a]/mol·kg^{-1}			
group	0	1.0	2.0	4.0	6.0	1.0	2.0	4.0	6.0
(NH_3^+, COO^-)	27.72	28.36	28.82	29.43	29.48	0.64	1.10	1.71	1.76
$-CH_2$	15.99	15.63	15.60	15.51	15.68	−0.36	−0.39	−0.48	−0.31
$-CHCH_3$	31.98	31.26	31.20	31.02	31.36	−0.72	−0.78	−0.96	−0.62
$-CHCH_2CH_3$	47.97	46.89	46.80	46.53	47.40	−1.08	−1.17	−1.44	−0.93
$-CHCH_2CH(CH_3)_2$	79.95	78.15	78.10	77.55	78.48	−1.80	−1.95	−2.40	−1.55

[a] m_s = molality of 1,2-propanediol in water.

V_2^∞ are 15.99 and 27.72 cm^3·mol^{-1}, respectively, which agree well with the literature values[12] (15.3 cm^3·mol^{-1} for the CH_2 group and 27.8 cm^3·mol^{-1} for the zwitterionic group). The contribution of (NH_3^+, COO^-) to V_2^∞ is larger than that of the CH_2 group and increases with the increase in the concentration of cosolute, which indicates that the interactions between cosolute and charged end groups (NH_3^+, COO^-) of amino acids are much stronger than those

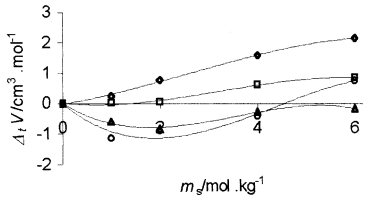

Figure 3. Standard volumes of transfer, $\Delta_t V^\infty$, of some amino acids vs molality, m_s, at 298.15 K: ◇, glycine; □, alanine; △, aminobutyric acid; ○. leucine.

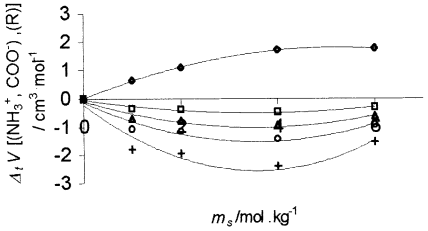

Figure 4. Contribution of NH_3^+, COO^- (◇) and $-CH_2$ (□), $-CHCH_3$ (△), $-CHCH_2CH_3$ (○), or $-CHCH_2CH(CH_3)_2$ (+) groups to standard volumes of transfer, $\Delta_t V^\infty$, vs molality, m_s, at 298.15 K.

between cosolute and CH_2. However, the contribution from the side chain increases with the increase in the size of the side chain of the amino acids.

The partial molar volumes of transfer of the zwitterionic end group [$\Delta_t V^\infty(NH_3^+, COO^-)$] and the alkyl side chain group $\Delta_t V^\infty(R)$ [R = $-CH_2$, $-CHCH_3$, $-CHCH_2CH_3$, and $-CHCH_2CH(CH_3)_2$] of amino acids from water to cosolute solution have been calculated as follows

$$\Delta_t V^\infty(NH_3^+, COO^-) \text{ or } \Delta_t V^\infty(R) = V_2^\infty(NH_3^+, COO^-) \text{ or } V_2^\infty(R) \text{ (in aqueous cosolute)} - V_2^\infty(NH_3^+, COO^-) \text{ or } V_2^\infty(R) \text{ (in water)} \quad (5)$$

The results are given in Table 3 and illustrated in Figure 4. The contribution of (NH_3^+, COO^-) to $\Delta_t V^\infty$ is positive throughout the concentration range of the cosolute, and its value increases with the increase in the concentration of cosolute. The contributions of R to $\Delta_t V^\infty$ values are negative for all the amino acids, and their magnitude increases with the increase in the number of carbon atoms of the alkyl side chain. After passing through minima which occur at around ≃3.5 mol·kg^{-1} (Figure 4), the contribution of R to $\Delta_t V^\infty$ values increases with the increase in concentration of cosolute.

The relative viscosities, η_r, of studied amino acids in water and in cosolute solutions were calculated using the following equation and are summarized in Table 4.

$$\eta_r = \eta/\eta_0 \quad (6)$$

where η and η_0 are the viscosities of solution and solvent. The *B*-coefficients were calculated by fitting the η_r values to the Jones−Dole equation by a least-squares method as follows

$$\eta_r = \eta/\eta_0 = 1 + Bc \quad (7)$$

where *c* is the molarity (calculated from molality data). The values of *B*-coefficients have been summarized in Table 5 along with the literature values. Good agreement between experimental and literature values has been observed in the case of water.[14−15] *B*-coefficient values are positive for

Table 4. Relative Viscosities, η_r, for Some Amino Acids in Aqueous 1,2-Propanediol Solutions as a Function of Concentrations of Amino Acid and 1,2-Propanediol at 298.15 K

c/mol·dm^{-3}	η_r	c/mol·dm^{-3}	η_r	c/mol·dm^{-3}	η_r
		Glycine			
		m_s[a]/mol·kg^{-1} = 1.0			
0.07517	1.0168	0.27827	1.0406	0.59973	1.0910
0.11846	1.0239	0.46139	1.0674	0.67723	1.1054
0.19944	1.0328	0.53309	1.0816		
		m_s/mol·kg^{-1} = 2.0			
0.11521	1.0140	0.26996	1.0412	0.39588	1.0616
0.16348	1.0228	0.31050	1.0453		
0.21130	1.0331	0.35367	1.0554		
		m_s/mol·kg^{-1} = 4.0			
0.07201	1.0074	0.31000	1.0405	0.73262	1.1114
0.10262	1.0108	0.40824	1.0561		
0.19917	1.0180	0.54196	1.0850		
		m_s/mol·kg^{-1} = 6.0			
0.04967	1.0014	0.14239	1.0163	0.33348	1.0510
0.08114	1.0056	0.20492	1.0222	0.42158	1.0608
		DL-α-Alanine			
		m_s/mol·kg^{-1} = 1.0			
0.19713	1.0457	0.39115	1.0970	0.62007	1.1655
0.29403	1.0789	0.51334	1.1320	0.66364	1.1836
		m_s/mol·kg^{-1} = 2.0			
0.04413	1.0139	0.29366	1.0825	0.58573	1.1704
0.09206	1.0258	0.40529	1.1163		
0.20427	1.0597	0.50982	1.1453		
		m_s/mol·kg^{-1} = 4.0			
0.04603	1.0114	0.19125	1.0522	0.42467	1.1198
0.09342	1.0240	0.24325	1.0735	0.49290	1.1396
0.13835	1.0352	0.34865	1.0977		
		m_s/mol·kg^{-1} = 6.0			
0.08600	1.0193	0.26578	1.0676	0.40302	1.1103
0.15326	1.0369	0.30623	1.0801	0.45702	1.1243
0.21203	1.0530	0.35903	1.0953		
		DL-α-Amino-*n*-butyric Acid			
		m_s/mol·kg^{-1} = 1.0			
0.06985	1.0270	0.11748	1.0397	0.17749	1.0641
0.07506	1.0312	0.13148	1.0466	0.20048	1.0758
0.10062	1.0363	0.16491	1.0605		
		m_s/mol·kg^{-1} = 2.0			
0.06528	1.0261	0.14559	1.0553	0.23025	1.0828
0.09047	1.0340	0.19058	1.0695		
0.10710	1.0418	0.20971	1.0736		
		m_s/mol·kg^{-1} = 4.0			
0.06398	1.0149	0.13689	1.0444	0.20612	1.0760
0.09884	1.0307	0.14221	1.0517	0.23505	1.0872
0.12177	1.0419	0.17825	1.0667		
		m_s/mol·kg^{-1} = 6.0			
0.09283	1.0303	0.13254	1.0470	0.16520	1.0544
0.10609	1.0352	0.13374	1.0483		
0.12706	1.0401	0.15147	1.0514		
		L-Leucine			
		m_s/mol·kg^{-1} = 1.0			
0.02330	1.0164	0.05179	1.0300	0.07402	1.0481
0.03738	1.0210	0.05845	1.0344	0.08449	1.0564
		m_s/mol·kg^{-1} = 2.0			
0.02501	1.0157	0.03424	1.0178	0.04754	1.0283
0.03004	1.0163	0.03994	1.0258	0.05024	1.0317
		m_s/mol·kg^{-1} = 4.0			
0.00869	1.0090	0.02608	1.0160	0.04896	1.0274
0.01437	1.0127	0.03627	1.0212	0.05505	1.0302
0.02244	1.0154	0.04192	1.0238		
		m_s/mol·kg^{-1} = 6.0			
0.02427	1.0086	0.03817	1.0182	0.05651	1.0292
0.02909	1.0137	0.04070	1.0201		
0.03083	1.0139	0.04898	1.0248		

[a] m_s = molality of 1,2-propanediol in water.

Table 5. Viscosity *B*-Coefficients, *B*, for Some Amino Acids in Aqueous 1,2-Propanediol Solutions at 298.15 K

	B/dm^3·mol^{-1} at various m_s[a]/mol·kg^{-1}			
amino acid	1.0	2.0	4.0	6.0
glycine	0.153	0.152	0.146	0.135
DL-α-alanine	0.265	0.287	0.282	0.265
DL-α-amino-*n*-butyric acid	0.366	0.364	0.357	0.337
L-leucine	0.603	0.601	0.581	0.502

[a] m_s = molality of 1,2-propanediol in water.

all amino acids, and these increase with the increase in the size of the side chain of the amino acids.

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