

Speeds of Sound and Viscosities in Aqueous Poly(ethylene glycol) Solutions at 303.15 and 308.15 K

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Speeds of sound (u) and viscosities (η) of water + glycol (mono-, di-, tri-, and tetraethylene glycol) mixtures have been measured at 303.15 K and 308.15 K. The results have been combined with those of our previous results for excess molar volumes converted to densities to obtain isentropic compressibilities. From the experimental data deviations in isentropic compressibility and viscosity have been calculated over the entire composition range.

Introduction

In a previous study on aqueous poly(ethylene glycol) solutions, the excess molar volumes (Pal et al., 1994) were determined. In this paper measurements of speeds of sound and viscosities of binary liquid mixtures of poly-(ethylene glycol) + water [$x\text{HOCH}_2(\text{CH}_2\text{—O—CH}_2)_n\text{CH}_2\text{OH} + (1 - x)\text{H}_2\text{O}$] ($n = 0, 1, 2, \text{ and } 3$) at 303.15 K and 308.15 K have been made.

Experimental Section

Materials. Materials were of the same origin and purity as used in earlier studies (Pal et al., 1994). All samples were kept over 4 Å molecular sieves to reduce the water content and protected against atmospheric moisture and CO_2 . The water used was deionized by ion-exchange resins and then doubly distilled. Its conductivity was always lower than $1.0 \times 10^{-6} \Omega^{-1} \text{ cm}^{-1}$. Prior to the measurements, all liquids were partially degassed under vacuum.

Apparatus and Procedure. Ultrasonic speed measurements in both the pure liquids and the binary mixtures were carried out at 303.15 K and 308.15 K using a NUSONIC (Mapco, Model 6080 Concentration Analyser) velocimeter based on the sing-around technique (Garnsey et al., 1969) with 4 MHz acoustic waves. The original glass sample jar supplied with the instrument was replaced with a specially designed low-volume cell (fabricated by the Glass Technology Unit, National Physical Laboratory, New Delhi) in which the first component of about 80 cm^3 is required to cover the transducer portion of the transducer assembly.

The second component is introduced into the cell from a glass syringe by mass, taking into account the effect of buoyancy. The composition of each mixture was obtained with an accuracy of 1×10^{-4} from the measured apparent masses of the components. The ultrasonic speeds u at a given temperature were obtained from the average round trip period of the ultrasonic wave in the fixed path length between the piezoelectric transducer and reflector. The velocimeter was calibrated at both temperatures by measurements of the ultrasonic speeds in water, and the relative error of speeds measured against water [1509.127 $\text{m}\cdot\text{s}^{-1}$ at 303.15 K and 1519.808 $\text{m}\cdot\text{s}^{-1}$ at 308.15 K (Del Grosso and Mader, 1972)] is estimated to be less than 0.2 $\text{m}\cdot\text{s}^{-1}$.

Viscosities of the pure liquids and the mixtures were measured at 303.15 K and 308.15 K with a calibrated suspended level Ubbelohde viscometer. The viscometers were calibrated, and two constants, A and B , of the viscometer in the equation $\eta = \rho(At - B/t)$ were obtained by measuring the flow time t with high-purity benzene, toluene, and carbon tetrachloride at the working temperatures (Riddick et al., 1986; Aminabhavi et al., 1990; Rout et al., 1994). The average of four or five sets of flow times for each liquid was taken for the purpose of the calculation of viscosity. The flow-time measurements were made with an electronic stopwatch having a precision of ± 0.01 s. Viscosities are reproducible to ± 0.003 $\text{mPa}\cdot\text{s}$. Densities of pure liquids were measured using a double-armed pycnometer with an accuracy of $\pm 1 \times 10^{-4} \text{ g}\cdot\text{cm}^{-3}$. A thermostatically controlled, well stirred water bath whose temperature was controlled to ± 0.01 K was used for all the measurements. Densities of liquid mixtures at any

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Table 1. Properties of Pure Liquids at 303.15 and 308.15 K

liquid	T/K	$\rho/\text{g}\cdot\text{cm}^{-3}$		$u/\text{m}\cdot\text{s}^{-1}$		$\eta/\text{mPa}\cdot\text{s}$	
		exptl	lit.	exptl	lit.	exptl	lit.
water	303.15		0.995647 ^b		1509.127 ^c		0.79726 ^b
	308.15		0.994032 ^b		1519.808 ^c		0.71903 ^b
ethylene glycol	303.15	1.1066	1.106488 ^a 1.10635 ^b	1644.2		13.543	13.55 ^b
	308.15	1.1031	1.102964 ^a	1632.1		11.069	10.968 ^e
diethylene glycol	303.15	1.1095	1.1096 ^d	1570.2		21.754	21.922 ^e
	308.15	1.1062	1.1061 ^d	1557.9		17.364	17.030 ^e
triethylene glycol	303.15	1.1164	1.1167 ^e	1597.3		29.256	29.224 ^e
	308.15	1.1127	1.1129 ^e	1582.1		22.853	22.961 ^e
tetraethylene glycol	303.15	1.1170	1.1168 ^e	1582.7		34.589	35.694 ^e
	308.15	1.1131	1.1128 ^e	1566.6		26.928	27.938 ^e

^a Douhéret et al. (1991). ^b Riddick et al. (1986). ^c Del Grosso and Mader (1972). ^d Rinkenbach (1927). ^e Interpolated data from Kumagai et al. (1993).

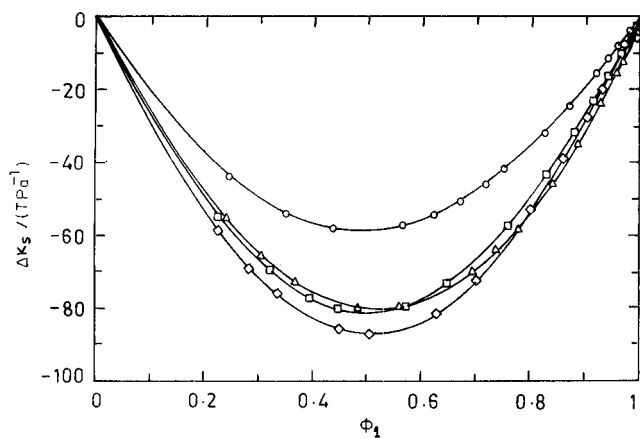


Figure 1. Deviations in isentropic compressibility ($\Delta\kappa_s$) for $[\text{HOCH}_2(\text{CH}_2\text{-O-CH}_2)_n\text{CH}_2\text{OH (1) + H}_2\text{O (2)}]$ at 303.15 K: (○) $n = 0$; (△) $n = 1$; (□) $n = 2$; (◇) $n = 3$. The solid curves were calculated from eq 5 using coefficients a_{j-1} of Table 4.

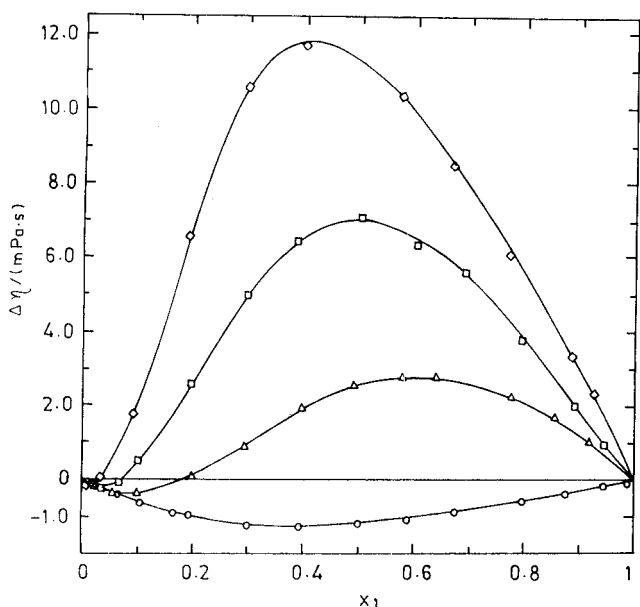


Figure 2. Deviations in viscosity ($\Delta\eta$) for $[\text{HOCH}_2(\text{CH}_2\text{-O-CH}_2)_n\text{CH}_2\text{OH (1) + H}_2\text{O (2)}]$ at 303.15 K: (○) $n = 0$; (△) $n = 1$; (□) $n = 2$; (◇) $n = 3$. The solid curves were calculated from eq 5 using coefficients a_{j-1} of Table 4.

composition were computed from the excess molar volume (V_m^E), reported earlier (Pal et al., 1994), using the relation

$$\rho = \frac{x_1 M_1 + x_2 M_2}{V_m^E + x_1 V_1 + x_2 V_2} \quad (1)$$

where x_1 and x_2 are the mole fractions, M_1 and M_2 are molar masses, and V_1 and V_2 are molar volumes of glycols (1) and water (2), respectively. The accuracy in ρ due to the estimated accuracy in excess molar volume ($\pm 0.003 \text{ cm}^3\text{-mol}^{-1}$) is $1 \times 10^{-4} \text{ g-cm}^{-3}$. The densities, speeds of sound, and viscosities of the component liquids are given in Table 1, together with some values taken from the literature.

Results and Discussion

The experimental results of the speeds of sound (u) and viscosities (η) for four binary mixtures of water + ethylene glycol, + diethylene glycol, + triethylene glycol, + tetraethylene glycol for a number of mole fractions at 303.15 K and 308.15 K are presented in Tables 2 and 3.

Table 2. Speeds of Sound (u), Isentropic Compressibilities (κ_s), and Deviations in Isentropic Compressibilities ($\Delta\kappa_s$) at 303.15 and 308.15 K

x_1	ϕ_1	$T = 303.15 \text{ K}$			$T = 308.15 \text{ K}$			
		$u/\text{m}\cdot\text{s}^{-1}$	κ_s/TPa^{-1}	$\Delta\kappa_s/\text{TPa}^{-1}$	$u/\text{m}\cdot\text{s}^{-1}$	κ_s/TPa^{-1}	$\Delta\kappa_s/\text{TPa}^{-1}$	
Ethylene Glycol (1) + Water (2)								
0.0953	0.2462	1617.2	371.4	-43.4	0.2464	1618.8	371.6	-40.4
0.1483	0.3505	1654.5	350.2	-53.4	0.3509	1652.4	352.0	-50.1
0.2004	0.4372	1678.1	336.8	-57.5	0.4376	1673.3	339.7	-54.1
0.2986	0.5689	1699.1	323.7	-56.6	0.5693	1691.2	327.7	-53.6
0.3480	0.6233	1702.1	320.7	-53.8	0.6236	1693.2	325.2	-51.0
0.3975	0.6716	1702.3	319.1	-50.2	0.6719	1692.6	323.9	-47.7
0.4499	0.7171	1699.9	318.8	-45.7	0.7174	1689.6	323.8	-43.5
0.4937	0.7514	1697.1	318.9	-41.9	0.7517	1686.5	324.0	-39.9
0.6047	0.8258	1686.1	321.3	-31.5	0.8261	1675.0	326.7	-30.2
0.6885	0.8726	1677.4	323.6	-24.2	0.8728	1665.1	329.5	-22.9
0.7897	0.9209	1665.2	327.4	-15.3	0.9210	1653.3	333.2	-14.6
0.8445	0.9439	1659.2	329.3	-10.9	0.9440	1647.4	335.1	-10.6
0.8922	0.9625	1654.2	330.9	-7.4	0.9625	1642.3	336.8	-7.1
0.9479	0.9826	1648.9	332.7	-3.5	0.9826	1636.9	338.6	-3.4
Diethylene Glycol (1) + Water (2)								
0.0553	0.2363	1622.7	368.2	-55.0	0.2366	1623.6	368.5	-52.1
0.0767	0.3051	1650.6	352.3	-65.7	0.3054	1648.7	353.9	-62.4
0.0981	0.3651	1671.6	340.6	-72.9	0.3654	1667.1	343.2	-69.3
0.1490	0.4807	1698.6	324.7	-80.0	0.4810	1690.6	328.6	-76.6
0.1957	0.5626	1706.0	318.7	-79.9	0.5629	1696.0	323.3	-76.7
0.2986	0.6923	1694.7	318.7	-70.0	0.6926	1682.6	324.3	-67.6
0.3463	0.7369	1684.7	321.4	-64.0	0.7371	1672.3	327.1	-61.9
0.3936	0.7743	1674.2	324.6	-58.0	0.7746	1661.3	330.6	-56.1
0.4941	0.8377	1651.8	332.6	-45.6	0.8379	1638.4	338.7	-44.0
0.5894	0.8836	1631.2	339.9	-34.5	0.8837	1618.5	346.3	-33.5
0.6983	0.9244	1611.6	347.6	-23.6	0.9245	1598.5	354.5	-22.7
0.7939	0.9540	1596.2	354.1	-14.9	0.9541	1583.5	360.9	-14.4
0.8441	0.9662	1589.7	356.9	-11.2	0.9663	1576.9	363.8	-10.8
0.8927	0.9778	1583.3	359.7	-7.5	0.9778	1570.5	366.7	-7.2
0.9398	0.9880	1577.5	362.3	-4.2	0.9880	1564.7	369.3	-3.9
Triethylene Glycol (1) + Water (2)								
0.0377	0.2256	1626.3	366.1	-54.6	0.2259	1626.9	366.6	-51.6
0.0592	0.3187	1667.7	343.1	-69.2	0.3191	1664.0	345.5	-65.6
0.0796	0.3913	1693.5	329.2	-76.6	0.3917	1686.9	332.7	-72.9
0.0985	0.4482	1708.7	320.7	-80.8	0.4486	1700.0	325.0	-76.3
0.1504	0.5682	1723.2	310.5	-79.4	0.5686	1710.9	315.9	-76.1
0.1982	0.6476	1718.3	309.5	-73.2	0.6480	1704.4	315.6	-70.4
0.2977	0.7591	1693.0	315.8	-56.9	0.7594	1678.4	322.4	-55.0
0.3942	0.8287	1669.5	323.4	-43.1	0.8289	1654.2	330.6	-41.6
0.4946	0.8792	1649.5	330.5	-31.4	0.8793	1634.2	337.9	-30.4
0.5901	0.9145	1634.8	336.0	-22.7	0.9147	1619.1	343.7	-21.8
0.6868	0.9422	1623.0	340.6	-15.7	0.9423	1607.5	348.4	-15.1
0.7841	0.9643	1614.0	344.2	-10.1	0.9643	1598.3	352.1	-9.6
0.8510	0.9770	1608.3	346.5	-6.7	0.9770	1592.9	354.4	-6.4
0.8848	0.9828	1605.5	347.6	-5.6	0.9828	1590.2	355.5	-4.8
0.9493	0.9929	1601.4	349.3	-2.4	0.9929	1586.2	357.2	-2.3
Tetraethylene Glycol (1) + Water (2)								
0.0295	0.2261	1630.1	363.9	-58.2	0.2264	1630.4	364.6	-55.2
0.0397	0.2843	1658.2	348.4	-68.8	0.2847	1655.4	350.5	-65.3
0.0494	0.3331	1678.7	337.4	-75.8	0.3335	1674.1	340.1	-72.2
0.0787	0.4508	1716.2	317.2	-86.1	0.4513	1706.6	321.7	-82.4
0.0966	0.5068	1725.7	311.3	-87.3	0.5073	1714.5	316.4	-83.9
0.1489	0.6270	1724.9	307.1	-81.5	0.6273	1710.8	313.2	-78.7
0.1971	0.7023	1710.1	310.1	-72.2	0.7027	1695.0	316.8	-69.9
0.2963	0.8018	1674.5	321.2	-52.7	0.8021	1659.7	328.2	-51.6
0.3893	0.8597	1649.2	330.3	-38.8	0.8599	1633.6	337.9	-37.9
0.4941	0.9037	1628.7	338.2	-27.3	0.9039	1612.6	346.2	-26.6
0.5823	0.9305	1615.3	343.5	-19.7	0.9307	1599.3	351.7	-19.2
0.6794	0.9532	1605.3	347.6	-13.8	0.9533	1588.4	356.4	-13.0
0.8013	0.9748	1595.4	351.9	-7.6	0.9749	1578.7	360.6	-7.2
0.9030	0.9889	1589.1	354.6	-3.7	0.9890	1572.4	363.4	-3.4

Isentropic compressibilities (κ_s) and deviations in isentropic compressibility ($\Delta\kappa_s$), which refers to the deviations from linearity on an ideal volume fraction average, are calculated using the relations

$$\kappa_s = 1/u^2 \rho \quad (2)$$

$$\Delta\kappa_s = \kappa_s - (\phi_1 \kappa_{s,1} + \phi_2 \kappa_{s,2}) \quad (3)$$

where ϕ_1 and ϕ_2 are the ideal volume fractions of compo-

Table 3. Viscosities (η) and Deviations in Viscosities ($\Delta\eta$) at 303.15 and 308.15 K

x_1	303.15 K		308.15 K		x_1	303.15 K		308.15 K	
	$\eta/\text{mPa}\cdot\text{s}$	$\Delta\eta/\text{mPa}\cdot\text{s}$	$\eta/\text{mPa}\cdot\text{s}$	$\Delta\eta/\text{mPa}\cdot\text{s}$		$\eta/\text{mPa}\cdot\text{s}$	$\Delta\eta/\text{mPa}\cdot\text{s}$	$\eta/\text{mPa}\cdot\text{s}$	$\Delta\eta/\text{mPa}\cdot\text{s}$
Ethylene Glycol (1) + Water (2)									
0.0003	0.778	-0.023	0.706	-0.016	0.2969	3.435	-1.146	2.919	-0.873
0.0009	0.781	-0.028	0.709	-0.019	0.3955	4.638	-1.200	3.905	-0.907
0.0059	0.808	-0.064	0.733	-0.047	0.4982	6.000	-1.147	5.004	-0.871
0.0100	0.834	-0.091	0.752	-0.071	0.5873	7.264	-1.019	6.040	-0.758
0.0136	0.856	-0.115	0.774	-0.086	0.6734	8.539	-0.841	7.077	-0.612
0.0616	1.180	-0.402	1.044	-0.313	0.7944	10.392	-0.530	8.555	-0.386
0.1054	1.529	-0.612	1.326	-0.484	0.8719	11.576	-0.334	9.501	-0.242
0.1634	2.030	-0.850	1.756	-0.654	0.9410	12.693	-0.098	10.349	-0.109
0.1935	2.348	-0.916	2.016	-0.706	0.9866	13.349	-0.023	10.905	-0.025
Diethylene Glycol (1) + Water (2)									
0.0004	0.778	-0.028	0.704	-0.022	0.2956	7.943	0.951	6.484	0.845
0.0007	0.785	-0.027	0.710	-0.021	0.3956	10.968	1.880	8.911	1.607
0.0015	0.788	-0.041	0.717	-0.027	0.4905	13.618	2.541	10.950	2.067
0.0027	0.804	-0.050	0.730	-0.034	0.5794	15.715	2.775	12.640	2.277
0.0047	0.836	-0.060	0.749	-0.048	0.6406	17.062	2.840	13.654	2.272
0.0193	1.020	-0.182	0.905	-0.135	0.7750	19.296	2.257	15.466	1.847
0.0548	1.579	-0.367	1.375	-0.256	0.8543	20.369	1.668	16.255	1.316
0.0991	2.467	-0.470	2.107	-0.262	0.9170	21.030	1.015	16.820	0.838
0.1966	4.980	0.063	4.169	0.178					
Triethylene Glycol (1) + Water (2)									
0.0004	0.781	-0.028	0.707	-0.021	0.1954	8.931	2.573	7.273	2.229
0.0008	0.788	-0.032	0.714	-0.023	0.2932	14.104	4.963	11.273	4.064
0.0014	0.798	-0.039	0.725	-0.025	0.3871	18.230	6.416	14.474	5.187
0.0024	0.820	-0.046	0.736	-0.036	0.5032	22.215	7.097	17.446	5.589
0.0056	0.875	-0.082	0.787	-0.056	0.5991	24.202	6.355	19.290	5.311
0.0110	0.986	-0.124	0.877	-0.086	0.6914	26.056	5.582	20.449	4.427
0.0317	1.480	-0.219	1.298	-0.123	0.7941	27.166	3.770	21.781	3.485
0.0563	2.254	-0.145	1.934	-0.031	0.8891	28.091	1.991	22.141	1.743
0.0988	4.058	0.449	3.330	0.424	0.9432	28.530	0.890	22.528	0.932
Tetraethylene Glycol (1) + Water (2)									
0.0003	0.784	-0.023	0.710	-0.017	0.1918	13.821	6.542	11.040	5.294
0.0008	0.794	-0.030	0.717	-0.023	0.2972	21.423	10.583	16.695	8.187
0.0012	0.804	-0.034	0.726	-0.024	0.3958	25.894	11.722	20.272	9.179
0.0027	0.843	-0.045	0.758	-0.032	0.5753	30.519	10.349	23.921	8.124
0.0051	0.909	-0.061	0.812	-0.041	0.6691	31.866	8.459	24.934	6.679
0.0115	1.094	-0.092	0.972	-0.048	0.7737	33.079	6.137	26.263	5.266
0.0339	1.958	0.015	1.688	0.080	0.8847	34.033	3.340	26.693	2.787
0.0941	5.694	1.717	4.713	1.528	0.9219	34.266	2.316	26.785	1.904

Table 4. Values of the Parameters of Eq 5 and Standard Deviations at 303.15 and 308.15 K

	T/K	a_0	a_1	a_2	a_3	a_4	a_5	$\sigma(A)$
Ethylene Glycol (1) + Water (2)								
$\Delta\kappa_s/\text{TPa}^{-1}$	303.15	-232.392	11.246	8.512	-6.141	19.955		0.1
	308.15	-219.408	5.677	10.182	-6.493	18.525		0.1
$\Delta\eta/\text{mPa}\cdot\text{s}$	303.15	-4.577	2.323	-0.069	-1.469	0.072	2.853	0.016
	308.15	-3.461	1.864	0.468	-0.450	-1.136	0.877	0.009
Diethylene Glycol (1) + Water (2)								
$\Delta\kappa_s/\text{TPa}^{-1}$	303.15	-321.577	-22.109	13.017	-12.393			0.1
	308.15	-308.017	-30.103	14.055	-1.778	-5.395		0.1
$\Delta\eta/\text{mPa}\cdot\text{s}$	303.15	10.284	9.439	-10.062	2.796	1.405		0.023
	308.15	8.426	6.988	-7.294	2.464	0.526		0.009
Triethylene Glycol (1) + Water (2)								
$\Delta\kappa_s/\text{TPa}^{-1}$	303.15	-323.928	-4.604	35.247	46.803	27.079	-73.135	0.2
	308.15	-309.634	-11.491	34.299	43.014	25.029	-58.602	0.2
$\Delta\eta/\text{mPa}\cdot\text{s}$	303.15	27.998	-1.850	-18.088	22.673	-10.619	-8.491	0.075
	308.15	22.357	-1.417	-9.378	21.267	-11.498	-9.327	0.069
Tetraethylene Glycol (1) + Water (2)								
$\Delta\kappa_s/\text{TPa}^{-1}$	303.15	-349.079	-14.797	37.038	64.810	42.285	-83.944	0.2
	308.15	-335.331	-20.606	45.417	30.649			0.2
$\Delta\eta/\text{mPa}\cdot\text{s}$	303.15	45.501	-22.531	-5.962	38.711	-34.162	9.027	0.083
	308.15	35.231	-16.499	2.724	33.061	-32.875		0.071

nents 1 and 2, respectively, and κ_s , and $\kappa_{s,1}$, and $\kappa_{s,2}$ are the isentropic compressibilities of the mixture and components 1 and 2, respectively. Data on the isentropic compressibility (κ_s) are included in Table 2. The deviations in the viscosity from a linear dependence on mole fraction average is given by

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (4)$$

where η , η_1 , and η_2 are the viscosities of the mixture and

components 1 and 2, respectively. The variation of $\Delta\kappa_s$ with volume fraction and of $\Delta\eta$ with mole fraction at 303.15 K is represented in Figures 1 and 2, respectively. The values of η at 308.15 K for the system ethylene glycol + water are in agreement, within experimental error, with those reported by Krishnaiah et al. (1994).

The composition dependence of the deviation in isentropic compressibility, and viscosity deviation are correlated by the Redlich-Kister (1948) polynomial

$$A = x_1 x_2 \sum_{j=1}^n a_{j-1} (x_2 - x_1)^{j-1} \quad (5)$$

where if A is the viscosity deviation, the composition is in mole fraction and, if A ($=\Delta\kappa_s$) is the compressibility deviation, the composition is in volume fraction and a_{j-1} are the adjustable parameters. The values of the parameters obtained by the method of least squares are given in Table 4 along with the standard deviation $\sigma(A)$. Values of $\sigma(A)$ were obtained from the equation

$$\sigma(A) = \left[\sum (A_{\text{exptl}} - A_{\text{calcd}})^2 / (n - p) \right]^{1/2} \quad (6)$$

where n is the total number of experimental points, p is the number of parameters, and A is the property.

The results given in Table 2 and in Figures 1 and 2 indicate that the deviations in isentropic compressibility ($\Delta\kappa_s$) are negative whereas the viscosity deviations ($\Delta\eta$), except for the ethylene glycol + water system, are negative to positive at both the temperatures over the entire range of compositions, and the magnitude tends to increase with chain length of the glycol. The ultrasonic speed increases with increasing temperature in the range $0.05 < x_1 < 0.10$ and then decreases beyond this range. It would appear that an increase in temperature affects the molecular organization in these systems so as increase the amount of free water and hence contribute to a closer packing of the molecules, thereby producing an increase in ultrasonic speed, but at $x_1 > 0.10$, the influence of the more compressible glycol component dominates the solution behavior and thus leads to a decrease in the ultrasonic speed. The same qualitative arguments can be used to describe the increase in viscosities of water–glycol mixtures above $x_1 = 0.001$ at both temperatures. The above conclusions also find support with the interpretation of the excess and apparent molar volume properties of these binary solvents.

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