Density, Speed of Sound, and Refractive Index of Aqueous Binary Mixtures of Some Glycol Ethers at $T = 298.15$ K

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The experimental data of densities (ρ), speeds of sound (u), and refractive indices (n_D) of aqueous solutions of different glycol ethers, namely, ethylene glycol *iso*-propyl ether (EGIPE), ethylene glycol monobutyl ether (EGMBE), diethylene glycol dimethyl ether (DEGDME), diethylene glycol monobutyl ether (DEGMBE), propylene glycol monomethyl ether (PGMME), and dipropylene glycol monomethyl ether (DPGMME), over the entire composition range, at temperature $T = 298.15$ K, and at one atmospheric pressure have been obtained. The derived parameters such as the apparent molar volume (ϕ_V) of solute, isentropic compressibility of solution (β_S), apparent molar isentropic compressibility (ϕ_{KS}) of solute, excess molar volume (V^E) of solution, molar refraction ($[R]_{1,2}$), deviation in refractive index (Δn_D) of solutions, deviation in molar refraction ($\Delta[R]_{1,2}$), and the deviation in isentropic compressibility ($\Delta\beta_S$) have been computed. The limiting apparent molar volumes of solutes (ϕ_V^0), limiting apparent molar isentropic compressibilities of solutes (ϕ_{KS}^0), and the limiting excess partial molar volumes of solutes ($\bar{V}_2^{0E}$) have also been obtained. The results are interpreted in terms of hydrogen bonding, solute−solute, and solute−solvent interactions.

Introduction

Volumetric and acoustic investigations of aqueous solutions of nonelectrolytes play an important role in understanding the nature and extent of molecular interactions occurring between solute and solvent molecules.[1−3] It has always interested chemists to study thermodynamic properties of aqueous solutions of nonelectrolytes like glycol ethers as they are important from both a theoretical and an industrial point of view.[4] Different short-chained glycol ethers are miscible with different solvents including water.[5,6] However, investigation of various thermodynamic and spectroscopic studies reveals that some glycol ethers in the water-rich region exhibit anomalous behavior.[7−11] The short chained glycol ethers exhibit the tendency to demix with the increasing temperature and separate at lower critical solution temperature.[12] Glycol ethers self-associate by intermolecular hydrogen bonding due to alcoholic −OH and partially etheric −O− and also by the hydrophobic interaction between alkoxyethyl groups in aqueous solution.[13−17]

The thermodynamic properties of aqueous solutions of various glycol ethers such as ethylene glycol monomethyl ether (EGMME), ethylene glycol monoethyl ether (EGMEE), diethylene glycol monomethyl ether (DEGMME), and diethylene glycol monoethyl ether (DEGMEE) over the entire composition range at $T = 298.15$ K have been reported earlier.[18]

In this work, we report the densities (ρ), speed of sound (u), and refractive indices (n_D) of aqueous solutions of EGIPE, EGMBE, DEGDME, DEGMBE, PGMME, and DPGMME over the entire composition range at temperature 298.15 K and at one atmospheric pressure. Although some of the properties for the above systems have been reported earlier,[8,10,19−21] it was thought worthwhile to undertake systematic measurements of density, speed of sound, and refractive index of aqueous solutions of the above glycol ethers at 298.15 K. The derived parameters such as the apparent molar volume of solute in water, the apparent molar isentropic compressibility of solute in water, excess molar volume of solution, molar refraction, and isentropic compressibility of solutions have been computed by using the experimental data. The deviation in the isentropic compressibility, deviation in refractive index, and deviation of molar refraction of the aqueous binary mixtures of the glycol ethers have been computed. Limiting apparent molar volume, limiting excess partial molar volume, and limiting apparent molar isentropic compressibility have been obtained for aqueous binary mixtures of solutes. The values of excess molar volume, deviation in isentropic compressibility, deviation in refractive index, and deviation in molar refraction have been processed to obtain the coefficients by fitting into the Redlich−Kister-type polynomial equation.[22] The results are interpreted in terms of extensive hydrogen bonding as well as various types of interactions taking place among solute and solvent molecules.

Materials and Methods

Ethylene glycol *iso*-propyl ether used was of synthesis grade (98 %) and was procured from Merck-Schuchardt, Germany.

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Table 1. Comparison of Observed Densities (ρ), Speeds of Sound (u), and Refractive Indices (n_D) for Pure Solutes with the Literature Values at T/K = 298.15

name of compound	CAS no.	ρ/kg·m^{-3} this work	ρ/kg·m^{-3} literature	u/m·s^{-1} this work	u/m·s^{-1} literature	n_D this work	n_D literature
EGIPE	109-59-1	899.69	899.680[g]	1268.6	1267.65[g]	1.4079	
EGMBE	111-76-2	895.79	895.971[a]	1308.7	1305.04[a]	1.4171	1.4177[k]
DEGMBE	112-34-5	950.41	947.98[d]	1360.0	1358.9[d]	1.4295	1.4299[j]
DEGDME	111-96-6	938.78	938.50[b] 938.90[c]	1278.4	1278.6[c]	1.4060	1.40585[b]
PGMME	107-98-2	916.31	917.45[f] 916.39[h]	1261.0	1261.0[h]	1.4010	1.4016[f]
DPGMME	34590-94-8	950.96	951.08[e] 952.70[i]	1302.3	1293.4[i]	1.4199	1.420[j]

[a] Ref 23. [b] Ref 24. [c] Ref 25. [d] Ref 26. [e] Ref 27. [f] Ref 19. [g] Ref 20. [h] Ref 28. [i] Ref 29. [j] Ref 30. [k] Ref 31.

Ethylene glycol monobutyl ether (98 %), diethylene glycol monobutyl ether (97 %), diethylene glycol dimethyl ether (99 %), propylene glycol monomethyl ether (98 %), and dipropylene glycol monomethyl ether (98 %) furnished by S. D. Fine Chemicals, India, were of LR grade. All the solutes were purified by fractional distillation. The purified solutes were stored and protected from atmospheric moisture by keeping over molecular sieves (Type 4A^0, E-Merck, India) in tightly capped dark bottles. They were partially degassed by passing nitrogen gas just prior to use. The purity of all the solvents was checked by comparing the experimental density, speed of sound, and refractive index data for pure liquids with the literature[19,20,23−31] as listed in Table 1.

All the binary mixtures were prepared in freshly prepared doubly distilled water on molality basis by using a Mettler Balance with the precision of ± 0.1 mg.

The densities of the aqueous solutions of solute were measured at temperature T = 298.15 K using modified Lypkin's bicapillary pyknometers (volume ≈22 cm^3). Calibration of pyknometers was done using the density of pure water from the literature.[31] The pyknometers were immersed in the water bath whose temperature was maintained constant by circulating the coolant liquid (water + methanol) from the MK-70 ultracryostat (Germany) which has the accuracy to maintain temperature to ± 0.02 K inside the thermostat. The temperature stability in the experimental water bath was ± 0.002 K. The details have been described elsewhere.[32] The accuracy of the density measurements was confirmed by measuring the densities of aqueous NaCl solution at 298.15 K. Our density data agreed well with the literature[33] to ± 0.05 kg·m^{-3}.

The speed of sound was measured in aqueous solutions of all the glycol ethers using an ultrasonic interferometer (model SI-2 M/s Dr. Steeg and Reuter, Germany) at a fixed frequency of 2 MHz and having temperature control ± 0.1 K. Temperature was maintained constant by circulating coolant liquid from the MLW MK-70 ultracryostat (Germany). The details are given elsewhere.[34] The interferometer was calibrated by measuring speed of sound in freshly prepared double distilled water.[35] The accuracy in speed of sound measurement was better than ± 0.5 m·s^{-1}.

Refractive index measurements were made for sodium light using Abbe's Refractometer (Carl Ziess, Germany) having an assembly for the temperature control of the sample holder by circulating liquid from the thermostat. The temperature of the liquid was maintained constant (± 0.02 K) by circulating coolant from the MK-70 ultracryostat (Germany). The details are given elsewhere.[18] The refractometer was calibrated using doubly distilled water.[31] Our refractive indices are accurate up to ± 0.0001.

Calculations of Derived Parameters. Apparent molar volumes (ϕ_V) of solutes in water at 298.15 K have been calculated by using the following equation

$$\phi_V = \frac{M_2}{\rho} + \left[\frac{1000(\rho_1 - \rho)}{m\rho\rho_1}\right] \tag{1}$$

where M_2 is the molar mass of the solute; m is the molality of the solution; and ρ and ρ_1 are the densities of solution and solvent, respectively.

Excess molar volumes (V^E) of solution for the binary mixtures of aqueous solutions of the solutes have been computed using the expression

$$V^E = \left[\frac{x_1M_1 + x_2M_2}{\rho}\right] - \left[\left(\frac{x_1M_1}{\rho_1}\right) + \left(\frac{x_2M_2}{\rho_2}\right)\right] \tag{2}$$

where M_1 and M_2 are the molar masses of the pure solvent and solute, respectively; x_1 and x_2 are the mole fractions of solvent and solute, respectively; and ρ, ρ_1, and ρ_2 are the densities of solution, pure solvent, and pure solute, respectively.

Isentropic compressibilities (β_S) of the solutions have been estimated using the Newton−Laplace equation, assuming the absorption of acoustic wave is negligible

$$\beta_S = \frac{1}{\rho u^2} \tag{3}$$

where ρ is the density of solution and u is speed of sound in solution.

The deviation in the isentropic compressibility ($\Delta\beta_S$) has also been computed using the relation given as below

$$\Delta\beta_S = \beta_S - [\phi_1\beta_{S_1} + \phi_2\beta_{S_2}] \tag{4}$$

where β_S, β_{S_1}, and β_{S_2} represent the values of isentropic compressibility for solution, pure solvent, and pure solute, respectively. ϕ_1 and ϕ_2 are the volume fractions of solvent and solute in the solution, respectively, where $\phi_i = (x_i\bar{V}_i^0)/V_{id}$, x_i, and $\bar{V}_i^0$ are the mole fraction and partial molar volume of the constituent i, respectively, and $V_{id} = \sum x_i\bar{V}_i^0$.

The apparent molar isentropic compressibilities (ϕ_{KS}) of the solute in water have been computed using the following equation

$$\phi_{KS} = \left[\frac{1000(\rho_1\beta_S - \rho\beta_{S_1})}{m\rho\rho_1}\right] + \left[\frac{M_2\beta_S}{\rho}\right] \tag{5}$$

where ρ and ρ_1 are the densities of solution and solvent, respectively; m is the molality of the solution; M_2 is the molar mass of the solute; and β_S and β_{S_1} represent the values of isentropic compressibility of solution and pure solvent, respectively.

Table 2. Density (ρ), Speed of Sound (u), Refractive Index (n_D), Isentropic Compressibility (β_S) of Aqueous Solutions of Solute, Apparent Molar Volume of Solutes (ϕ_V), Apparent Molar Isentropic Compressibility (ϕ_{KS}) of Solutes in Aqueous Solutions, Excess Molar Volume (V^E), Deviation in Isentropic Compressibility ($\Delta\beta_S$), Deviation in Refractive Index (Δn_D), Apparent Molar Refraction ($[R]_{1,2}$), and Deviation in Molar Refraction ($\Delta[R]_{1,2}$) of Aqueous Solutions of Glycol Ethers at T/K = 298.15

x_2	ρ / $kg \cdot m^{-3}$	u / $m \cdot s^{-1}$	n_D	$10^6 \cdot \phi_V$ / $m^3 \cdot mol^{-1}$	$10^{11} \cdot \beta_S$ / $N^{-1} \cdot m^2$	$10^{15} \cdot \phi_{KS}$ / $N^{-1} \cdot m^5 \cdot mol^{-1}$	$10^6 \cdot V^E$ / $m^3 \cdot mol^{-1}$	$10^{11} \cdot \Delta\beta_S$ / $N^{-1} \cdot m^2$	Δn_D	$10^6 \cdot [R]_{1,2}$ / $m^3 \cdot mol^{-1}$	$10^6 \cdot \Delta[R]_{1,2}$ / $m^3 \cdot mol^{-1}$
						EGIPE					
0.0000	997.05	1496.7	1.3325	107.9	44.77	−13.61	0.00	0.00	0.0000	3.7116	0.0000
0.0050	996.27	1524.5	1.3355	107.3	43.19	−10.96	−0.04	−2.29	0.0008	3.8342	−0.5997
0.0075	996.00	1530.2	1.3376	107.1	42.88	0.35	−0.06	−2.94	0.0019	3.9018	−0.8797
0.0100	995.77	1541.8	1.3396	106.9	42.25	−0.15	−0.09	−3.91	0.0028	3.9693	−1.1555
0.0251	995.00	1593.3	1.3476	106.1	39.59	5.62	−0.24	−8.42	0.0051	4.3364	−2.6856
0.0500	993.52	1620.9	1.3590	106.0	38.31	18.41	−0.49	−12.27	0.0085	4.9460	−4.7052
0.0748	989.70	1610.9	1.3675	106.9	38.94	28.56	−0.66	−13.74	0.0104	5.5558	−6.2441
0.0999	984.98	1588.8	1.3737	107.7	40.22	35.90	−0.80	−14.23	0.0111	6.1675	−7.4458
0.1504	974.25	1532.7	1.3845	109.3	43.69	46.64	−0.97	−13.56	0.0133	7.4409	−9.0361
0.1986	965.16	1493.9	1.3890	110.3	46.43	52.41	−1.08	−12.84	0.0115	8.6063	−9.9291
0.3015	949.21	1435.8	1.3975	111.8	51.10	59.80	−1.18	−11.17	0.0107	11.1713	−10.4374
0.4033	937.08	1391.8	1.4019	112.9	55.09	64.93	−1.17	−9.15	0.0090	13.7044	−9.9151
0.4960	928.29	1355.1	1.4047	113.6	58.66	69.17	−1.09	−6.86	0.0078	16.0261	−8.9135
0.6005	920.17	1331.8	1.4054	114.2	61.27	71.95	−0.94	−5.35	0.0051	18.5916	−7.4701
0.7027	913.59	1313.6	1.4065	114.7	63.43	74.18	−0.75	−4.02	0.0036	21.1397	−5.7684
0.7957	908.56	1297.5	1.4070	115.1	65.38	76.19	−0.54	−2.68	0.0022	23.4501	−4.0774
0.9034	903.69	1283.7	1.4071	115.4	67.15	77.96	−0.28	−1.48	0.0005	26.1090	−2.0055
1.0000	899.69	1268.6	1.4079	115.8	69.06	88.86	0.00	0.00	0.0000	28.5514	0.0000
						EGMBE					
0.0000	997.05	1496.7	1.3325	124.6	44.77	−12.97	0.00	0.00	0.0000	3.7116	0.0000
0.0050	995.62	1521.8	1.3361	123.8	43.37	3.08	−0.04	−2.08	0.0008	3.8577	−0.8409
0.0075	995.08	1536.0	1.3393	123.5	42.59	0.51	−0.06	−3.19	0.0026	3.9457	−1.2256
0.0124	994.34	1552.6	1.3421	122.8	41.72	7.25	−0.11	−4.68	0.0029	4.0822	−1.9777
0.0249	990.15	1538.3	1.3490	124.3	42.68	38.17	−0.19	−5.15	0.0038	4.4454	−3.6788
0.0502	980.52	1506.7	1.3601	126.3	44.93	57.23	−0.28	−5.30	0.0050	5.1879	−6.3958
0.0748	971.93	1481.8	1.3688	127.4	46.86	64.32	−0.34	−5.22	0.0060	5.9187	−8.3420
0.1187	960.53	1455.1	1.3803	128.1	49.17	68.89	−0.45	−5.43	0.0071	7.2166	−10.6863
0.1486	954.18	1442.7	1.3867	128.5	50.35	70.47	−0.51	−5.57	0.0080	8.1117	−11.6966
0.1993	945.30	1424.9	1.3931	129.0	52.10	72.52	−0.59	−5.56	0.0071	9.5904	−12.7429
0.2969	932.71	1398.2	1.4041	129.7	54.84	75.41	−0.67	−5.12	0.0086	12.5238	−13.1249
0.4011	923.10	1376.0	1.4075	130.2	57.22	77.84	−0.70	−4.33	0.0055	15.5341	−12.4027
0.4975	916.23	1361.8	1.4109	130.6	58.85	79.42	−0.66	−3.72	0.0046	18.3815	−11.0356
0.5981	910.49	1346.7	1.4130	131.0	60.56	81.22	−0.58	−2.81	0.0034	21.3400	−9.2255
0.7004	905.80	1335.5	1.4155	131.2	61.90	82.56	−0.48	−2.09	0.0033	24.4003	−7.0606
0.7937	902.23	1323.4	1.4167	131.5	63.29	84.07	−0.36	−1.15	0.0027	27.1621	−4.9496
0.9107	898.33	1313.2	1.4168	131.7	64.55	85.39	−0.16	−0.34	0.0009	30.5646	−2.2044
1.0000	895.79	1308.7	1.4171	131.9	65.18	95.95	0.00	0.00	0.0000	33.1822	0.0000
						DEGMBE					
0.0000	997.05	1496.7	1.3325	163.7	44.77	−40.52	0.00	0.00	0.0000	3.7116	0.0000
0.0010	997.04	1507.7	1.3339	162.8	44.12	−45.57	−0.01	−0.76	0.0005	3.7559	−0.3222
0.0020	997.13	1513.9	1.3356	161.9	43.76	−20.52	−0.02	−1.23	0.0014	3.8026	−0.6283
0.0050	997.60	1526.2	1.3380	160.6	43.03	6.09	−0.05	−2.26	0.0013	3.9151	−1.5454
0.0075	998.10	1544.6	1.3415	160.0	41.99	0.20	−0.08	−3.55	0.0028	4.0252	−2.2571
0.0098	998.56	1555.2	1.3426	159.7	41.41	4.41	−0.11	−4.36	0.0021	4.1061	−2.9201
0.0125	999.03	1564.8	1.3460	159.5	40.88	9.37	−0.14	−5.14	0.0035	4.2244	−3.6373
0.0250	999.25	1571.7	1.3551	160.8	40.51	35.01	−0.25	−6.54	0.0043	4.7167	−6.5990
0.0252	999.25	1566.2	1.3563	160.8	40.80	37.73	−0.25	−6.27	0.0054	4.7374	−6.6289
0.0502	996.46	1546.2	1.3703	163.0	41.98	58.85	−0.39	−6.72	0.0064	5.7367	−11.0361
0.0753	993.48	1532.8	1.3800	164.1	42.84	66.00	−0.50	−7.07	0.0063	6.7322	−14.1046
0.1012	990.25	1515.2	1.3886	164.9	43.99	71.27	−0.58	−6.90	0.0071	7.7812	−16.2998
0.1511	985.47	1494.4	1.3995	165.8	45.44	76.01	−0.74	−6.81	0.0071	9.7813	−18.8283
0.2014	980.54	1481.7	1.4072	166.7	46.45	78.61	−0.81	−6.75	0.0072	11.8194	−19.9498
0.3030	973.56	1453.3	1.4152	167.6	48.63	83.13	−0.93	−5.80	0.0053	15.8800	−20.0012
0.4021	967.48	1422.4	1.4203	168.5	51.09	87.78	−0.88	−4.09	0.0045	19.8924	−18.4697
0.4939	964.01	1409.6	1.4229	168.9	52.21	89.56	−0.88	−3.45	0.0033	23.5683	−16.3821
0.6049	959.05	1395.2	1.4251	169.6	53.57	91.89	−0.65	−2.51	0.0021	28.0668	−13.2698
0.7064	956.27	1384.0	1.4269	170.0	54.59	93.53	−0.51	−1.76	0.0017	32.1819	−10.0979
0.7988	954.15	1379.2	1.4276	170.2	55.10	94.26	−0.37	−1.46	0.0007	35.8897	−7.0683
0.8948	952.06	1369.6	1.4284	170.5	55.99	95.71	−0.18	−0.74	0.0001	39.7713	−3.7611
1.0000	950.41	1360.0	1.4295	170.7	56.89	102.17	0.00	0.00	0.0000	44.0491	0.0000
						DEGDME					
0.0000	997.05	1496.7	1.3325	133.8	44.77	−9.57	0.00	0.00	0.0000	3.7116	0.0000
0.0048	997.40	1524.3	1.3360	133.3	43.15	−1.25	−0.05	−2.32	0.0010	3.8616	−0.9329
0.0075	997.77	1533.3	1.3373	132.7	42.63	4.98	−0.08	−3.22	0.0009	3.9391	−1.4366
0.0124	998.61	1554.0	1.3420	132.1	41.47	7.23	−0.13	−5.04	0.0032	4.1055	−2.2773
0.0356	1003.00	1608.2	1.3517	130.9	38.55	19.94	−0.43	−10.60	0.0034	4.7722	−5.6771
0.0502	1004.83	1651.8	1.3620	130.9	36.47	19.33	−0.60	−14.04	0.0088	5.2637	−7.2804
0.0748	1006.15	1665.5	1.3718	131.3	35.83	27.05	−0.87	−16.58	0.0118	6.0292	−9.4410
0.0998	1005.85	1655.5	1.3787	132.0	36.28	34.01	−1.09	−17.70	0.0131	6.7976	−11.0698
0.1500	1000.60	1603.8	1.3883	133.7	38.85	45.89	−1.38	−17.48	0.0142	8.3633	−13.1351
0.1996	993.39	1550.5	1.3935	135.3	41.87	54.56	−1.52	−16.14	0.0133	9.9097	−14.1697
0.3008	979.90	1469.5	1.3996	137.7	47.26	66.10	−1.58	−13.05	0.0112	13.0896	−14.5177
0.4062	968.57	1408.2	1.4020	139.3	52.06	74.45	−1.47	−9.75	0.0081	16.3907	−13.5409

Table 2. Continued

x_2	ρ / $kg \cdot m^{-3}$	u / $m \cdot s^{-1}$	n_D	$10^6 \cdot \phi_V$ / $m^3 \cdot mol^{-1}$	$10^{11} \cdot \beta_S$ / $N^{-1} \cdot m^2$	$10^{15} \cdot \phi_{KS}$ / $N^{-1} \cdot m^5 \cdot mol^{-1}$	$10^6 \cdot V^E$ / $m^3 \cdot mol^{-1}$	$10^{11} \cdot \Delta\beta_S$ / $N^{-1} \cdot m^2$	Δn_D	$10^6 \cdot [R]_{1,2}$ / $m^3 \cdot mol^{-1}$	$10^6 \cdot \Delta[R]_{1,2}$ / $m^3 \cdot mol^{-1}$
DEGDME											
0.5031	960.57	1372.3	1.4036	140.4	55.28	79.47	−1.29	−7.50	0.0062	19.4485	−11.9641
0.6025	954.28	1346.0	1.4040	141.1	57.84	83.19	−1.08	−5.67	0.0040	22.5530	−9.9857
0.7027	949.26	1334.0	1.4046	141.7	59.20	85.00	−0.84	−4.88	0.0026	25.7045	−7.7056
0.8052	944.94	1306.0	1.4051	142.2	62.05	89.00	−0.56	−2.49	0.0014	28.9390	−5.1745
0.9035	941.39	1294.4	1.4056	142.6	63.40	90.79	−0.26	−1.49	0.0006	32.0564	−2.6038
1.0000	938.78	1278.4	1.4060	142.9	65.18	99.14	0.00	0.00	0.0000	35.1065	0.0000
PGMME											
0.0000	997.05	1496.7	1.3325	93.5	44.77	−14.66	0.00	0.00	0.0000	3.7116	0.0000
0.0025	996.69	1509.3	1.3336	93.0	44.04	−11.77	−0.01	−1.03	0.0002	3.7613	−0.2089
0.0075	996.36	1523.6	1.3363	92.1	43.24	2.95	−0.05	−2.43	0.0012	3.8653	−0.6062
0.0122	996.29	1540.9	1.3385	91.6	42.27	1.88	−0.08	−3.93	0.0019	3.9595	−0.9652
0.0248	996.55	1570.7	1.3435	90.8	40.67	7.73	−0.19	−6.87	0.0030	4.2042	−1.8557
0.0502	996.91	1628.4	1.3545	90.4	37.83	10.47	−0.40	−12.07	0.0073	4.7237	−3.3245
0.0748	996.80	1645.5	1.3618	90.5	37.05	16.24	−0.59	−14.76	0.0091	5.2061	−4.4612
0.0988	995.92	1642.7	1.3683	90.7	37.21	21.26	−0.76	−16.20	0.0110	5.6860	−5.3339
0.1499	990.48	1624.5	1.3780	91.7	38.26	28.38	−1.00	−17.89	0.0128	6.7092	−6.6351
0.2002	984.07	1574.5	1.3848	92.5	40.99	35.20	−1.17	−17.24	0.0136	7.7243	−7.3803
0.3000	970.87	1508.2	1.3917	94.0	45.28	42.76	−1.32	−15.93	0.0120	9.7177	−7.9074
0.3991	959.07	1440.0	1.3964	95.0	50.28	49.29	−1.32	−12.96	0.0108	11.7336	−7.6147
0.5039	948.69	1400.9	1.3981	95.9	53.71	53.10	−1.23	−11.10	0.0080	13.8298	−6.8409
0.6039	940.41	1357.3	1.3998	96.5	57.72	57.26	−1.09	−8.22	0.0065	15.8622	−5.7639
0.7019	933.32	1337.3	1.4003	97.1	59.91	59.33	−0.89	−6.90	0.0045	17.8372	−4.5282
0.7947	927.28	1304.7	1.4007	97.5	63.35	62.66	−0.65	−4.13	0.0030	19.7212	−3.2151
0.8920	921.69	1280.9	1.4009	98.0	66.13	65.24	−0.35	−1.95	0.0015	21.6985	−1.7359
1.0000	916.31	1261.1	1.4010	98.4	68.62	73.67	0.00	0.00	0.0000	23.8956	0.0000
DPGMME											
0.0000	997.05	1496.7	1.3325	145.8	44.77	−0.99	0.00	0.00	0.0000	3.7116	0.0000
0.0050	998.00	1524.7	1.3370	145.0	43.10	2.07	−0.05	−2.34	0.0011	3.8891	−1.2146
0.0075	998.58	1537.4	1.3400	144.7	42.37	3.83	−0.08	−3.39	0.0025	3.9865	−1.7782
0.0136	1000.11	1561.7	1.3444	144.2	41.00	9.28	−0.16	−5.50	0.0031	4.1954	−3.0926
0.0250	1002.97	1607.0	1.3529	143.6	38.61	12.02	−0.31	−9.12	0.0054	4.5972	−5.2384
0.0500	1007.24	1643.5	1.3683	143.7	36.76	25.24	−0.61	−13.15	0.0097	5.4846	−8.8755
0.0743	1008.35	1640.0	1.3791	144.5	36.87	35.45	−0.85	−14.67	0.0122	6.3470	−11.4059
0.0994	1007.66	1613.0	1.3871	145.3	38.14	44.58	−1.04	−14.74	0.0134	7.2341	−13.3044
0.1480	1002.34	1570.4	1.3970	147.3	40.45	55.09	−1.26	−14.37	0.0135	8.9573	−15.6016
0.1966	996.47	1522.7	1.4033	148.8	43.28	63.28	−1.39	−12.93	0.0128	10.6868	−16.7364
0.2934	986.17	1464.9	1.4106	150.8	47.25	72.32	−1.49	−10.79	0.0108	14.1406	−17.0800
0.3986	977.22	1420.0	1.4148	152.2	50.75	78.87	−1.45	−8.54	0.0087	17.9065	−15.8973
0.4950	970.81	1386.1	1.4167	153.2	53.61	83.74	−1.33	−6.45	0.0066	21.3459	−14.0753
0.5790	966.24	1367.8	1.4178	153.8	55.32	86.46	−1.18	−5.26	0.0051	24.3475	−12.1301
0.6984	960.80	1342.6	1.4189	154.5	57.74	90.24	−0.91	−3.39	0.0034	28.6266	−8.9889
0.8177	956.29	1322.4	1.4192	155.1	59.80	93.38	−0.57	−1.74	0.0016	32.8829	−5.5866
0.8873	954.06	1315.2	1.4194	155.4	60.60	94.55	−0.36	−1.14	0.0009	35.3739	−3.5024
1.0000	950.96	1302.3	1.4199	155.8	62.00	101.61	0.00	0.00	0.0000	39.4297	0.0000

The deviations in the refractive index (Δn_D) of solution are estimated by using the following expression

$$\Delta n_D = n_D - [\phi_1 n_{D_1} + \phi_2 n_{D_2}] \tag{6}$$

where n_D, n_{D_1}, and n_{D_2} represent the values of refractive indices for the solution, pure solvent, and pure solute, respectively. ϕ_1 and ϕ_2 are the volume fractions of solvent and solute in the solution, respectively.

The molar refractions ($[R]_{1,2}$) of the solution have been calculated using the following expression

$$[R]_{1,2} = \left[\frac{n_D^2 - 1}{n_D^2 + 2}\right]\left[\frac{x_1 M_1 + x_2 M_2}{\rho}\right] \tag{7}$$

where n_D and ρ represent the values of refractive index and density of the solution; M_1 and M_2 are the molar masses of the pure solvent and pure solute, respectively; and x_1 and x_2 are the mole fractions of the solvent and solute, respectively.

The deviations in the molar refraction ($\Delta[R]_{1,2}$) have been computed using the following expression

$$\Delta[R]_{1,2} = [R]_{1,2} - (\phi_1[R]_1 + \phi_2[R]_2) \tag{8}$$

where $[R]_{1,2}$ is molar refraction of solution; ϕ_1 and ϕ_2 are the volume fractions of solvent and solute in the solution, respectively; and $[R]_1$ and $[R]_2$ are molar refractions of pure solvent and solute, respectively.

The values of these parameters at different concentrations for aqueous binary mixtures of all the glycol ethers studied in the present work are listed in Table 2.

The limiting apparent molar volumes of solute (ϕ_V^0) and the limiting apparent molar isentropic compressibilities (ϕ_{KS}^0) of the solutes in aqueous solutions at $T = 298.15$ K have been obtained by smooth extrapolation of $\phi_V - x_2$ and $\phi_{KS} - x_2$ curves, respectively, to the zero concentration. The limiting excess partial molar volumes of solute ($\bar{V}_2^{0E}$) were calculated using the following relation

$$\bar{V}_2^{0E} = \phi_V^0 - V_2^0 \tag{8a}$$

where ϕ_V^0 and V_2^0 are the limiting apparent molar volume of solute and the molar volume of pure solute, respectively. Table 3 lists the values of ϕ_V^0, ϕ_{KS}^0, and $\bar{V}_2^{0E}$.

The error in β_S values is of the order of $\pm 0.05 \cdot 10^{-11}$ $m^2 \cdot N^{-1}$. The errors at the lowest concentration studied for the derived parameters are of the order of $\pm 0.1 \cdot 10^{-6}$ $m^3 \cdot mol^{-1}$ and $\pm 1 \cdot 10^{-15}$ $m^5 \cdot N^{-1} \cdot mol^{-1}$ for ϕ_V and ϕ_{KS}, respectively.

The composition dependence of excess molar volume, deviation in the isentropic compressibility, deviation in the refractive

Table 3. Limiting Apparent Molar Volumes of Solute (ϕ_V^0), Limiting Excess Partial Molar Volumes of Solute ($\bar{V}_2^{0E}$), and Limiting Apparent Molar Isentropic Compressibility of Solutes (ϕ_{KS}^0) in Aqueous Solutions at T/K = 298.15

name of compound	$10^6 \cdot \phi_V^0$ / $m^3 \cdot mol^{-1}$	$10^6 \cdot \bar{V}_2^{0E}$ / $m^3 \cdot mol^{-1}$	$10^{15} \cdot \phi_{KS}^0$ / $N^{-1} \cdot m^5 \cdot mol^{-1}$
EGIPE	107.91	−7.84	−13.61
EGMBE	124.60	−10.32	−12.97
DEGMBE	163.70	−6.99	−40.52
DEGDME	133.83	−9.09	−9.57
PGMME	93.55	−4.82	−14.66
DPGMME	145.82	−10.02	−0.99

index, and deviation in molar refraction for aqueous solutions have been fitted by a general curve fitting method for binary mixtures using the Redlich−Kister-type equation

$$F(x) = x_2(1 - x_2)\sum_{i=0}^{n} A_i(1 - 2x_2)^i \tag{9}$$

where $F(x)$ refers to the values of V^E, $\Delta\beta_S$, Δn_D, or $\Delta[R]_{1,2}$. The coefficient A_i is the polynomial coefficient tabulated by using the least-squares fit method computed using MAPLE software, whereas for $\Delta\beta_S$, Δn_D, $\Delta[R]_{1,2}$ instead of x_2, ϕ_2 values have been used. These coefficients for aqueous solutions of different glycol ethers are listed in Table 4.

The calculated values of molar refractions ($[R]_{1,\ 2}$) of the solution were fitted by the equation of straight line. These coefficients are also given in Table 4.

The values of the standard deviation (σ) were obtained from the expression

$$\sigma = \{\sum (F(x)_{exp} - F(x)_{calcd})^2/(k/n)\}^{1/2} \tag{10}$$

where k is the number of experimental points excluding the end points and n is the order of polynomial equation.

The average absolute deviation (AAD) was obtained by using the equation

$$AAD = \sum \{[F(x)_{exp} - F(x)_{calcd}]/F(x)_{exp}\}/k \tag{11}$$

The values of the coefficient (A_i), standard deviation (σ), and average absolute deviation (AAD) are listed in Table 4.

Results and Discussion

Figure 1 shows the variation of excess molar volume (V^E) of the solution against the mole fraction of the solute (x_2) over the entire concentration range for aqueous solutions of all the glycol ethers. It is observed from the above figure that the values of excess molar volume of the solution for aqueous solutions of all the glycol ethers over the entire mole fraction range are negative. The close scrutiny of the above figure reveals that V^E decreases sharply with concentration of the solute, goes through a minimum, and then increases less sharply with further increase in the concentration of the solute for all the aqueous systems. The minimum for all the systems lies in the water-rich region. The magnitude of minimum in excess molar volume (V^E) of

Table 4. Estimated Values of Coefficient (A_i), Standard Deviation (σ), and Absolute Average Deviation (AAD) for the Various Properties of Aqueous Solutions at T/K = 298.15

properties	A_0	A_1	A_2	A_3	A_4	σ	AAD
			EGIPE				
$10^6 \cdot V^E/m^3 \cdot mol^{-1}$	−4.3427	−2.2346	−1.3198	−1.8976	−1.9059	0.025	−0.0600
$10^{11} \cdot \Delta\beta_S/N^{-1} \cdot m^2$	−55.281	−14.554	−23.9065	10.8542	7.6873	0.3733	0.0143
Δn_D	0.0491	−0.0079	0.0084	−0.0009	−0.0062	0.0016	0.3676
$10^6 \cdot \Delta[R]_{1,2}/m^3 \cdot mol^{-1}$	−35.654	23.4812	−14.4246	22.7806	−19.3639	0.1309	−0.0104
			EGMBE				
$10^6 \cdot V^E/m^3 \cdot mol^{-1}$	−2.6729	−0.9093	0.17981	−1.1443	−2.13382	0.0367	0.0549
$10^{11} \cdot \Delta\beta_S/N^{-1} \cdot m^2$	−21.808	5.5582	−18.2677	−27.563	−24.1397	0.3167	−0.0522
Δn_D	0.0295	−0.0161	0.0043	0.0137	0.0229	0.0011	−0.1216
$10^6 \cdot \Delta[R]_{1,2}/m^3 \cdot mol^{-1}$	−44.217	29.7496	−18.4683	34.1041	−30.6346	0.1804	−0.0097
			DEGMBE				
$10^6 \cdot V^E/m^3 \cdot mol^{-1}$	−3.3677	−2.0642	0.6870	−1.2434	−3.7887	0.0457	0.0111
$10^{11} \cdot \Delta\beta_S/N^{-1} \cdot m^2$	−27.841	1.0124	−25.0555	−16.369	−5.6559	0.3798	0.0245
Δn_D	0.0279	−0.0076	0.0182	0.0127	−0.0091	0.0007	−0.3889
$10^6 \cdot \Delta[R]_{1,2}/m^3 \cdot mol^{-1}$	−64.867	46.1882	−34.9755	65.6456	−54.9706	0.6274	−0.022
			DEGDME				
$10^6 \cdot V^E/m^3 \cdot mol^{-1}$	−5.1408	−4.0982	−4.8069	−2.2833	2.1027	0.0396	−0.0717
$10^{11} \cdot \Delta\beta_S/N^{-1} \cdot m^2$	−73.586	−5.404	55.4552	14.6318	−69.8278	1.196	0.0325
Δn_D	0.0565	−0.027	−0.0449	0.0122	0.0325	0.0015	−0.2521
$10^6 \cdot \Delta[R]_{1,2}/m^3 \cdot mol^{-1}$	−48.364	32.7983	−20.1675	41.5655	−38.595	0.2896	−0.004
			PGMME				
$10^6 \cdot V^E/m^3 \cdot mol^{-1}$	−4.9127	−2.4386	−2.855	−0.832	2.1437	0.0289	−0.0919
$10^{11} \cdot \Delta\beta_S/N^{-1} \cdot m^2$	−70.886	−0.0559	−5.7154	18.184	−8.5331	0.5203	0.0025
Δn_D	0.0524	−0.0149	−0.0066	−0.007	0.0006	0.0005	−0.0382
$10^6 \cdot \Delta[R]_{1,2}/m^3 \cdot mol^{-1}$	−27.47	17.6142	−10.8907	13.9454	−10.9482	0.0611	−0.02
			DPGMME				
$10^6 \cdot V^E/m^3 \cdot mol^{-1}$	−5.2771	−3.0553	−2.5674	−2.8161	−1.1849	0.0336	−0.0588
$10^{11} \cdot \Delta\beta_S/N^{-1} \cdot m^2$	−59.225	−7.1658	−12.0283	15.1812	5.2769	0.2088	−0.0008
Δn_D	0.0542	−0.0145	−0.0119	−0.0069	0.0123	0.0004	−0.1151
$10^6 \cdot \Delta[R]_{1,2}/m^3 \cdot mol^{-1}$	−56.258	39.0166	−22.1469	52.1379	−53.1355	0.3992	−0.0053
			Linear Fitting Coefficients of $[R]_{1,2}$				
EGIPE	3.711	24.80				0.0172	−0.00044
EGMBE	3.711	29.50				0.0275	0.00071
DEGMBE	3.711	40.29				0.0228	0.00015
DEGDME	3.711	31.32				0.0011	−0.00266
PGMME	3.711	20.14				0.0232	−0.00106
DPGMME	3.711	35.67				0.0276	−0.00116

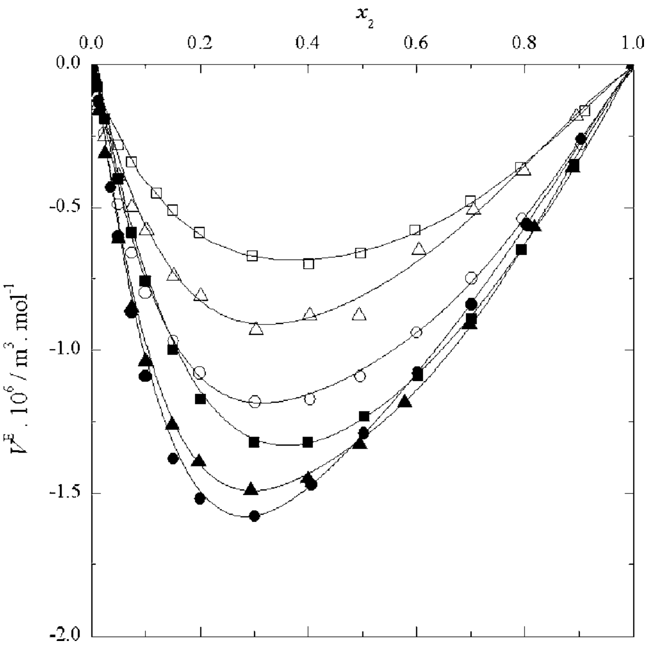

Figure 1. Plot of excess molar volume of solutions (V^E) with mole fraction of solute (x_2) for aqueous systems at 298.15 K: □−□, EGMBE; ■−■, PGMME; Δ−Δ, DEGMBE; ▲−▲, DPGMME; ○−○, EGIPE; ●−●, DEGDME.

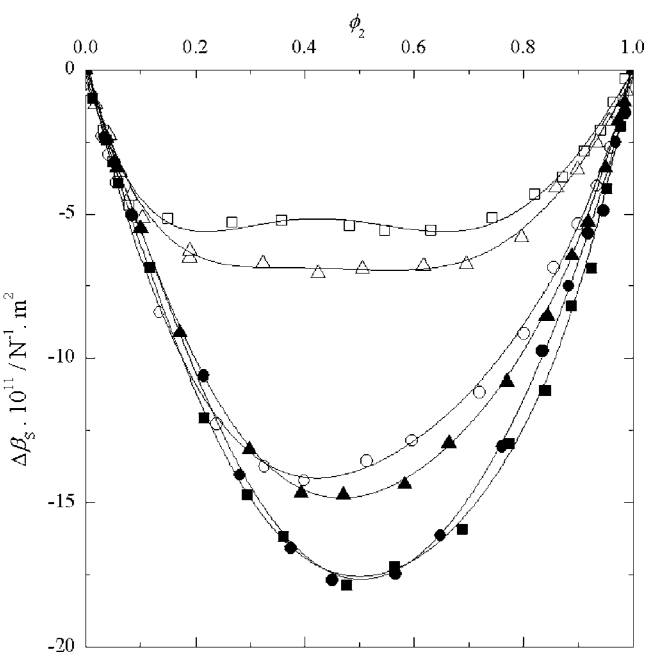

Figure 2. Plot of deviation in isentropic compressibility of solutions ($\Delta\beta_S$) with volume fraction of solute (ϕ_2) for aqueous systems at 298.15 K: □−□, EGMBE; ■−■, PGMME; Δ−Δ, DEGMBE; ▲−▲, DPGMME; ○−○, EGIPE; ●−●, DEGDME.

the solution increases in the order of EGMBE < DEGMBE < EGIPE < PGMME < DPGMME < DEGDME. It is observed from the above figure that the minimum in the excess molar volume of the solution shifts to lower concentration for diethers in comparison to their respective monoethers. The minimum in the curve suggests that strong hydrogen bonding interactions are taking place at that concentration of the solute.

Figure 2 represents the variation of deviation of isentropic compressibility ($\Delta\beta_S$) as a function of volume fraction of solute (ϕ_2) for all the systems of aqueous solutions of glycol ethers at 298.15 K. It is observed from the above figure that the values

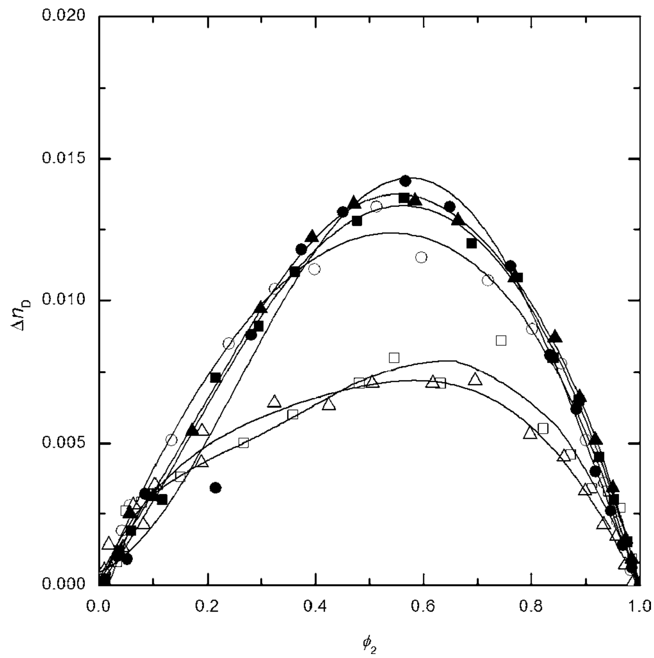

Figure 3. Plot of deviation in the refractive indices (Δn_D) with volume fraction of solute (ϕ_2) for aqueous systems at 298.15 K: □−□, EGMBE; ■−■, PGMME; Δ−Δ, DEGMBE; ▲−▲, DPGMME; ○−○, EGIPE; ●−●, DEGDME.

of $\Delta\beta_S$ of the solution are all negative. The negative magnitude of the values increases with increase in volume fraction of solute initially, and it reaches to maximum and then decreases with further increase in volume fraction of the solute. Thus, a plot of $\Delta\beta_S - \phi_2$ shows a minimum in the curve. The minimum in the curve suggests maximum hydrogen bonding at the volume fraction of the minimum. From the comparison of Figure 1 and Figure 2 it is observed that the behavior of $V^E - x_2$ curves and $\Delta\beta_S - \phi_2$ curves is similar.

In Figure 3 are plotted the deviations in refractive index (Δn_D) as a function of volume fraction of solute (ϕ_2) for all the binary mixtures. It is observed that all of the values of Δn_D are positive over the entire volume fraction range. The Δn_D values increase with an increase in the volume fraction of the solute initially, go through maximum, and then decrease with further increase in volume fraction of the solute. This behavior is exactly opposite to the behavior of $V^E - x_2$ curves.

Figure 4 represents the variation of deviation of molar refraction ($\Delta[R]_{1,2}$) as a function of volume fraction of the solute (ϕ_2) at 298.15 K for all binary aqueous systems. It is observed from the above that the values of $\Delta[R]_{1,2}$ are all negative for all the aqueous systems. It is seen from the above figure that $\Delta[R]_{1,2}$ decreases with increase in volume fraction of the solute, goes through a minima, and then increases with further increase in volume fraction of the solute. The minimum in the curve suggests the extensive hydrogen bonding at that volume fraction.

In Table 3 are listed the values of limiting apparent molar volume of solutes (ϕ_V^0) in water, limiting excess partial molar volume ($\bar{V}_2^{0E}$) of the solutes, and apparent molar isentropic compressibilities of solutes (ϕ_{KS}^0) in water at 298.15 K for all the aqueous systems of glycol ethers studied in this work. It is observed from the above table that ϕ_V^0 values for all the solutes are positive, whereas $\bar{V}_2^{0E}$ are negative for all the solutes. It is also seen from the above table that all the values of ϕ_{KS}^0 are negative for all the solutes. The order of increase in the magnitude is as follows: DPGMME < DEGDME < EGMBE < EGIPE < PGMME < DEGMBE. This indicates that glycol ethers

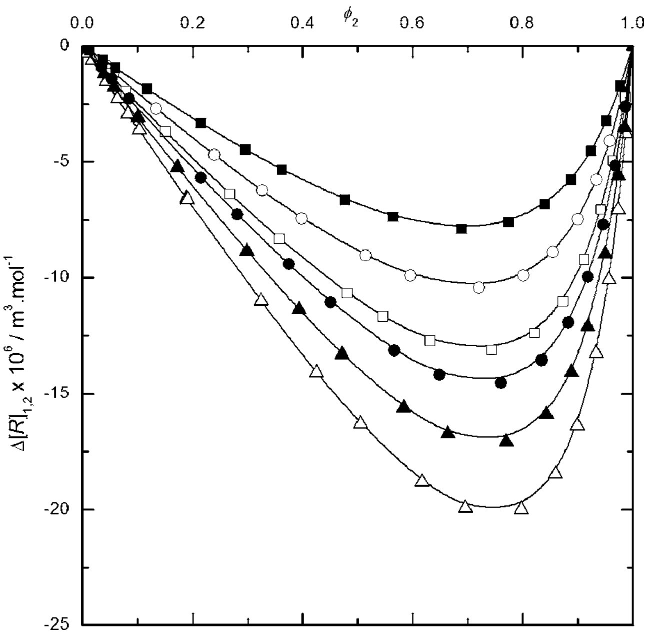

Figure 4. Plot of deviation in the molar refraction ($\Delta[R]_{1,2}$) with volume fraction of solute (ϕ_2) for aqueous systems at 298.15 K: □—□, EGMBE; ■—■, PGMME; Δ—Δ, DEGMBE; ▲—▲, DPGMME; ○—○, EGIPE; ●—●, DEGDME.

get accommodated in the vicinity of the structured lattice of the water on dissolution[18] and give protection to an ordered compact structure like aggregates.

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