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Volumetric and Acoustic Properties for Binary Mixtures of Dipropylene Glycol Monopropyl Ether with Alkylamines at Temperatures Between 288.15 K and 308.15 K

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Abstract The densities, ρ, and speeds of sound, u, have been measured as a function of composition for binary liquid mixtures of dipropylene glycol monopropyl ether (DPGMPE) with n-butylamine (BA), dibutylamine (DBA), and tributylamine (TBA) at (288.15, 293.15, 298.15, 303.15, and 308.15) K and atmospheric pressure using an Anton Paar DSA-5000 instrument. The ρ and u values were used to calculate excess molar volumes, V^{E}, deviations from the ideal behavior of the thermal expansion coefficient, α^{E}, and the isentropic compressibilities, $\Delta\kappa_S$. Moreover, the apparent molar volume $\bar{V}^0_{\phi,i}$, and apparent molar compressibility $\bar{K}^0_{\phi,i}$, of the components have been calculated at infinite dilution. The Jouyban–Acree model is used to correlate the experimental values of density and ultrasonic speed at different temperatures.

Keywords n-Alkylamines · Density · Dipropylene glycol monopropyl ether · Ultrasonic (sound) speed

1 Introduction

Alkoxyalkanols represent a very interesting class of substances due to the presence of –O- and –OH groups in the same molecule, which allow self-association via inter- and intra-molecular hydrogen bonds. Different spectroscopic techniques have been used to investigate the existence of intramolecular hydrogen bonds [1–6], which are present at all conditions, even in the vapor phase. In contrast, intermolecular H-bonds appear at higher concentrations of the alkoxyalkanols in the liquid state. Alkoxyalkanols are

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used in the chemical industry as a solvent for oils and petrol and also used as cleaning agents and as fuel additives. Amines are used to make azo-dyes, many drugs, and medicines. Therefore, the treatment of this class of mixtures is a particular challenge to describe interaction behavior occurring in the solution.

As a part of our ongoing program of research on thermodynamic and acoustic properties of binary liquid mixtures containing alkoxyethanols or alkoxypropanols, we have recently reported measurements of excess molar volumes, viscosities, and speeds of sound of binary mixtures of alkoxyalkanols with *n*-alkylamines [7–10], 1-alkanols [11–16], or amides [17]. As a part of this continuing work, the experimental results for the densities, ρ, and speeds of sound, u, are reported in this article for the binary systems of dipropylene glycol monopropyl ether (DPGMPE) with *n*-butylamine, dibutylamine, and tributylamine over the complete composition range and those of the pure liquid components at atmospheric pressure and temperatures of (288.15, 293.15, 298.15, 303.15, and 308.15) K. The experimental values of $\rho$and u were used to calculate the excess molar volume, V^{E}, deviations in isentropic compressibilities, $\Delta\kappa_S$, and the apparent molar volume and apparent molar compressibility, $\bar{V}^0_{\phi,i}$ and $\bar{K}^0_{\phi,i}$, of the components at infinite dilution.

2 Experimental

Butylamine (BA), dibutylamine (DBA), and tributylamine (TBA) (all analytical reagent grade products from S.D. Fine Chemicals, India) were stored over sodium hydroxide pellets for several days and fractionally distilled twice [18,19]. The middle fraction of the distillate was used. The DPGMPE ($C_3H_7(OC_3H_6)_2OH$) (Aldrich, Germany, purity >99.1 %) was used without further purification. Before use, all the liquids were stored in dark bottles over 0.4-nm molecular sieves to reduce water content, and were partially degassed with a vacuum pump under a nitrogen atmosphere. The estimated purities determined by gas chromatographic analysis were better than 99.5 mol % for all the liquid samples. The water content, measured for each sample by Karl–Fisher titration, was always found to be less than 0.002 mass %. Furthermore, the purities of the final samples were checked by comparing the densities and speeds of sound at the desired temperatures with their corresponding values available in the literature [19–24], and are presented in Table 1.

The densities, ρ, and speeds of sound, u, of both pure liquids and of their binary mixtures were simultaneously, and automatically measured, using an Anton Paar DSA 5000 instrument thermostated within $\pm 1 \times 10^{-2}$ K. The calibration of the apparatus was carried out with deionized double-distilled water, *n*-hexane, *n*-heptane, *n*-octane, cyclohexane, benzene, and toluene at the desired temperatures. The sensitivity of the instrument corresponded to a precision in density and speed-of-sound measurements of $1 \times 10^{-6}\ \mathrm{g \cdot cm^{-3}}$ and $1 \times 10^{-2}\ \mathrm{m \cdot s^{-1}}$, respectively. The reproducibility of the density and speed-of-sound estimates was found to be within $\pm 5 \times 10^{-6}\ \mathrm{g \cdot cm^{-3}}$ and $\pm 5 \times 10^{-2}\ \mathrm{m \cdot s^{-1}}$, respectively.

The binary mixtures were prepared by mass and were kept in special airtight stoppered glass bottles to avoid evaporation. The weighings were done with an electronic balance with a precision of ±0.01 mg. The probable error in the mole fraction was

Table 1 Thermodynamic properties for pure components

	T (K)	$\rho^* \times 10^{-3}\,(\mathrm{kg \cdot m^{-3}})$		$\alpha_P^* \times 10^{-3}\,(\mathrm{K^{-1}})$	$\kappa_S\,(\mathrm{TPa^{-1}})$	$u\,(\mathrm{m \cdot s^{-1}})$	
		Exptl.	Lit.			Exptl.	Lit.
Dipropylene glycol mono-propyl ether	288.15	0.924348			634.023	1306.26	
	293.15	0.919954			655.344	1287.90	
	298.15	0.915522	0.9164 [20]	0.977	677.339	1269.88	
	303.15	0.911078			700.580	1251.68	
	308.15	0.906642	0.9075 [20]		724.255	1234.06	
Butylamine	288.15	0.742865		1.280	801.358	1296.08	
	293.15	0.738096	0.73869 [21]	1.304	835.311	1273.56	
	298.15	0.733301	0.73359 [21] 0.73323 [22]	1.320	872.822	1249.96	1250 [22] 1249.8 [23]
	303.15	0.728477	0.72849 [21] 0.72842 [22]	1.337	908.944	1228.92	1227 [22]
	308.15	0.723625	0.72339 [21] 0.72364 [22]	1.360	950.059	1205.72	1204 [22]
Dibutylamine	288.15	0.766115		1.020	785.317	1289.23	
	293.15	0.762022		1.180	814.306	1269.47	
	298.15	0.757784	0.75572 [19] 0.75770 [24]	1.200	849.059	1246.69	1248 [22]
	303.15	0.752284		1250	883.368	1226.70	1227 [22]
	308.15	0.748170		1.330	917.501	1206.97	1206 [22]
Tributylamine	288.15	0.781336		0.949	775.049	1285.04	
	293.15	0.777632		0.959	801.871	1266.37	
	298.15	0.773913	0.77391 [22]	0.967	830.098	1247.64	1246.9 [23]
	303.15	0.770183	0.77046 [22]	0.975	859.640	1228.98	
	308.15	0.766440	0.76695 [22]	0.984	890.532	1210.42	

estimated to be less than $\pm 1 \times 10^{-4}$. All the molar quantities were based on the IUPAC relative atomic mass Table [25].

3 Results and Discussion

The experimental results of the density, ρ, and speed-of-sound, u, measurements of binary mixtures of DPGMPE with BA, DBA, and TBA, over the whole composition range expressed as the mole fraction, x_1, of DPGMPE ($0 \leq x_1 \leq 1$) at different temperatures are listed in Table 2.

The excess molar volumes, V^{E}, were calculated by using the following relation:

$$V^{\mathrm{E}} = \sum_i x_i M_i \left(\rho^{-1} - \rho_i^{*-1}\right) \tag{1}$$

where ρ and ρ_i^* are the density of the mixture and density of pure component i, respectively; x_i and M_i are the mole fraction and molar mass of component i in the mixture, respectively.

The isentropic compressibilities, κ_S have been calculated from the relation

$$\kappa_S = -V^{-1}\left(\delta V/\delta P\right)_S = \left(\rho u^2\right)^{-1} \tag{2}$$

The deviations of the isentropic compressibility, $\Delta\kappa_S$, and excess molar thermal expansivities, $\alpha^*_{p,i}$ were calculated from experimental u and ρ values, as follows:

$$\Delta\kappa_S = \kappa_S - \kappa_S^{\mathrm{id}} \tag{3}$$

where [26–28]

$$\kappa_S^{\mathrm{id}} = \sum \left(\kappa^*_{S,i}\right)\varphi_i \tag{4}$$

$$\alpha^{\mathrm{E}} = \alpha - \sum \varphi_i \alpha^*_{p,i} \tag{5}$$

where φ_i is the volume fraction and $\alpha^*_{p,i}$ is the isobaric expansivity.

The calculated values of V^{E} and $\Delta\kappa_S$ of the binary mixtures, at each temperature, were fitted to a Redlich–Kister polynomial eq.:

$$F(x) = x_1 x_2 \sum_{i=0} a_i \left(x_1 - x_2\right)^i \tag{6}$$

where if $F(x)$ $(\Delta\kappa_S)$ is the deviation in isentropic compressibility, the composition is in volume fraction, and if $F(x)$ $\left(V^{\mathrm{E}}\right)$ is the excess molar volume, the composition is in mole fraction. Values of coefficients a_i of Eq. 6, evaluated by using the method of least squares with all points weighted equally along with the standard deviations, $\sigma(F(x))$are listed in Table 3. Experimental results for V^{E} are plotted against x_i and

Table 2 Values of densities, ρ, and speeds of sound, u, of binary mixtures as a function of mole fraction, x_1, of DPGMPE at different temperatures

x_1	$\rho \times 10^{-3}$ (kg · m^{-3})					u (m · s^{-1})				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
DPGMPE (1) + butylamine (2)										
0.1119	0.781657	0.776972	0.772263	0.767527	0.762757	1299.12	1277.85	1256.72	1235.48	1214.24
0.1625	0.797600	0.792951	0.788269	0.783562	0.778830	1301.83	1280.99	1260.17	1239.50	1218.71
0.2153	0.812620	0.807989	0.803334	0.798647	0.793946	1304.26	1283.87	1263.36	1243.18	1222.78
0.2414	0.819423	0.814802	0.810159	0.805481	0.800782	1305.36	1285.15	1264.79	1244.81	1224.60
0.3043	0.834258	0.829657	0.825032	0.820381	0.815702	1307.67	1287.87	1267.80	1248.27	1228.41
0.3500	0.843808	0.839220	0.834611	0.829962	0.825299	1309.06	1289.49	1269.68	1250.35	1230.71
0.4162	0.856225	0.851642	0.847040	0.842409	0.837754	1310.65	1291.39	1271.86	1252.77	1233.42
0.4519	0.862336	0.857764	0.853179	0.848559	0.843907	1311.32	1292.18	1272.80	1253.82	1234.61
0.4946	0.869260	0.864701	0.860118	0.855511	0.850865	1311.93	1292.93	1273.70	1254.83	1235.73
0.5470	0.877184	0.872638	0.868071	0.863480	0.858867	1312.39	1293.55	1274.54	1255.75	1236.84
0.5938	0.883779	0.879250	0.874697	0.870123	0.865527	1312.59	1293.84	1274.99	1256.28	1237.45
0.6567	0.891947	0.887440	0.882917	0.878364	0.873793	1312.51	1293.87	1275.22	1256.60	1237.89
0.6997	0.897047	0.892555	0.888041	0.883505	0.878950	1312.24	1293.66	1275.11	1256.55	1237.96
0.7665	0.904096	0.899635	0.895150	0.890644	0.886119	1311.48	1293.00	1274.59	1256.14	1237.70
0.8068	0.907864	0.903410	0.898940	0.894449	0.889942	1310.82	1292.35	1274.06	1255.71	1237.37
0.8466	0.911283	0.906844	0.902376	0.897903	0.893412	1309.98	1291.59	1273.38	1255.15	1236.91
0.9038	0.915886	0.911457	0.906999	0.902546	0.898065	1308.62	1290.26	1272.14	1254.15	1236.10
0.9528	0.919898	0.915486	0.911061	0.906607	0.902137	1307.16	1288.89	1270.88	1253.12	1235.23
DPGMPE (1) + dibutylamine (2)										
0.1230	0.792458	0.788243	0.783999	0.779716	0.775539	1291.15	1271.60	1252.04	1232.14	1212.22
0.1582	0.798993	0.794736	0.790406	0.786126	0.781949	1293.18	1273.25	1253.68	1233.94	1214.00
0.1913	0.804954	0.800654	0.796281	0.791940	0.787756	1294.88	1274.68	1255.10	1235.52	1215.55
0.2541	0.815956	0.811610	0.807160	0.802761	0.798492	1297.60	1277.10	1257.47	1238.13	1218.12
0.3195	0.827100	0.822710	0.818293	0.813903	0.809461	1299.77	1279.28	1259.58	1240.41	1220.40
0.3623	0.834198	0.829803	0.825410	0.821074	0.816520	1300.90	1280.48	1260.76	1241.67	1221.68
0.4155	0.842790	0.838381	0.834053	0.829782	0.825107	1301.98	1281.80	1262.04	1243.03	1223.06
0.4543	0.848877	0.844447	0.840151	0.835948	0.831190	1302.61	1282.65	1262.86	1243.90	1223.90
0.4974	0.855422	0.850980	0.846721	0.842533	0.837752	1303.15	1283.49	1263.65	1244.74	1224.74

Table 2 continued

x_1	$\rho \times 10^{-3}$ (kg · m^{-3})					u (m · s^{-1})				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.5491	0.863031	0.858573	0.854308	0.850092	0.845325	1303.62	1284.32	1264.48	1245.60	1225.62
0.6027	0.870648	0.866201	0.861902	0.857606	0.852894	1303.98	1285.06	1265.22	1246.36	1226.39
0.6687	0.879855	0.875389	0.871011	0.866586	0.861990	1304.29	1285.77	1266.00	1247.14	1227.17
0.6962	0.883669	0.879204	0.874806	0.870321	0.865762	1304.37	1286.00	1266.30	1247.42	1227.47
0.7504	0.891256	0.886786	0.882325	0.877785	0.873301	1304.58	1286.42	1266.84	1247.96	1228.01
0.7997	0.898225	0.893775	0.889311	0.884771	0.880284	1304.82	1286.70	1267.30	1248.42	1228.47
0.8657	0.907649	0.903208	0.898805	0.894377	0.889838	1305.27	1287.04	1267.92	1249.04	1229.09
0.8963	0.911923	0.907504	0.903128	0.898789	0.894205	1305.55	1287.15	1268.20	1249.32	1229.37
0.9589	0.920017	0.915650	0.911300	0.907024	0.902428	1306.33	1287.39	1268.83	1249.97	1230.00
DPGMPE (1) + tributylamine (2)										
0.1122	0.794307	0.790425	0.786535	0.782639	0.778737	1282.28	1263.03	1243.91	1225.01	1206.30
0.1665	0.800621	0.796684	0.792738	0.788781	0.784827	1282.31	1262.78	1243.67	1224.80	1206.08
0.2084	0.805602	0.801628	0.797652	0.793661	0.789668	1282.33	1262.86	1243.75	1224.89	1206.16
0.2538	0.811153	0.807130	0.803115	0.799083	0.795057	1282.38	1263.20	1244.09	1225.27	1206.53
0.3130	0.818607	0.814554	0.810479	0.806404	0.802329	1282.55	1263.95	1244.88	1226.12	1207.40
0.3558	0.824155	0.820060	0.815967	0.811844	0.807734	1282.77	1264.75	1245.72	1226.97	1208.26
0.4148	0.832006	0.827869	0.823733	0.819576	0.815399	1283.28	1266.18	1247.16	1228.46	1209.78
0.4472	0.836410	0.832261	0.828099	0.823913	0.819704	1283.67	1267.07	1248.07	1229.40	1210.75
0.5008	0.843844	0.839656	0.835454	0.831251	0.826995	1284.53	1268.80	1249.82	1231.18	1212.59
0.5451	0.850136	0.845938	0.841706	0.837473	0.833167	1285.47	1270.35	1251.45	1232.84	1214.29
0.6077	0.859289	0.855060	0.850786	0.846555	0.842165	1287.17	1272.80	1253.99	1235.42	1216.98
0.6566	0.866709	0.862460	0.858150	0.853912	0.849462	1288.88	1274.89	1256.15	1237.65	1219.26
0.7143	0.875786	0.871507	0.867175	0.862905	0.858430	1291.34	1277.50	1258.85	1240.42	1222.14
0.7514	0.881851	0.877544	0.873195	0.868910	0.864430	1293.23	1279.26	1260.69	1242.29	1224.10
0.8016	0.890298	0.885967	0.881597	0.877285	0.872799	1296.14	1281.68	1263.25	1244.95	1226.85
0.8560	0.897740	0.895367	0.890981	0.886630	0.882174	1299.88	1284.40	1266.12	1247.92	1229.96
0.9040	0.908188	0.903800	0.899400	0.894989	0.890566	1303.68	1286.85	1268.68	1250.60	1232.77
0.9553	0.917078	0.912677	0.908256	0.903826	0.899408	1308.26	1289.48	1271.50	1253.55	1235.87

Table 3 Coefficients, a_i, of Eq. 6 and standard deviations, σ, for the binary mixtures at different temperatures

Property	T (K)	a_0	a_1	a_2	a_3	a_4	σ
DPGMPE (1) + butylamine (2)							
$V^E \times 10^6$ ($m^3 \cdot mol^{-1}$)	288.15	−5.0076	1.2964	−3.7184	−0.3283	10.1069	0.0011
	293.15	−5.0673	1.3781	−3.8878	−0.2404	10.4267	0.0014
	298.15	−5.1537	1.4454	−4.0162	−0.2689	10.5824	0.0014
	303.15	−5.2362	1.5114	−4.1437	−0.2973	10.7071	0.0013
	308.15	−5.3048	1.5875	−4.3467	−0.2926	11.0166	0.0015
$\Delta\kappa_S$ (TPa^{-1})	288.15	−96.3085	−20.5937	−6.5389	−46.9707	68.6414	0.276
	293.15	−106.4013	−22.0423	−4.9802	−49.1707	68.6237	0.270
	298.15	−118.6198	−18.6685	−10.4700	−43.8009	64.7495	0.272
	303.15	−127.6557	−21.4592	4.3286	−55.3575	55.9826	0.155
	308.15	−139.9212	−18.1537	4.2269	−54.8333	61.4404	0.171
$\alpha^E \times 10^5$(K^{-1})	298.15	−8.4085	1.6382	−11.2155	−11.2538	27.4444	0.356
DPGMPE (1) + dibutylamine (2)							
$V^E \times 10^6$ ($m^3 \cdot mol^{-1}$)	288.15	−4.8214	3.6127	2.2651	−0.6460	−10.5973	0.0013
	293.15	−4.7148	3.6265	2.4412	−0.5697	−10.9172	0.0009
	298.15	−4.8545	3.4738	3.7694	−0.2031	−13.2266	0.0014
	303.15	−5.6392	3.9712	5.3142	1.2509	−17.9980	0.0012
	308.15	−5.2841	4.0016	3.4140	1.6111	−15.5365	0.0013
$\Delta\kappa_S$(TPa^{-1})	288.15	−70.0845	32.4097	27.0649	−41.6016	−16.8125	0.224
	293.15	−69.3953	19.5958	19.1691	−21.3632	−14.2602	0.106
	298.15	−77.2241	30.8610	17.6473	1.1512	−38.6648	0.064
	303.15	−85.9410	36.6481	22.8377	9.9147	−43.6756	0.140
	308.15	−81.5196	41.5442	7.5222	27.0965	10.5912	0.483
$\alpha^E \times 10^5$(K^{-1})	298.15	−9.2031	0.0995	17.3018	26.8700	−55.4509	0.220
DPGMPE (1) + tributylamine (2)							
$V^E \times 10^6$ ($m^3 \cdot mol^{-1}$)	288.15	1.3548	0.1926	0.3640	−1.9858	−4.7319	0.0010

Table 3 continued

Property	T (K)	a_0	a_1	a_2	a_3	a_4	σ
	293.15	1.5619	0.0854	0.3660	−1.8970	−4.4391	0.0014
	298.15	1.7492	0.0600	0.4325	−2.0589	−4.4410	0.0013
	303.15	1.9325	−0.1252	0.2343	−2.0091	−4.0547	0.0013
	308.15	2.1755	0.0959	0.4965	−2.4701	−4.4526	0.0014
$\Delta\kappa_S(\mathrm{TPa}^{-1})$	288.15	23.3657	−2.6051	−1.8165	−69.0587	−57.8161	0.561
	293.15	9.0774	−34.2413	12.8051	−46.6143	−41.8010	0.341
	298.15	12.7016	−37.0977	13.5485	−54.0105	−41.4846	0.386
	303.15	14.8526	−40.1247	14.9190	−62.2609	−44.1944	0.451
	308.15	18.8640	−42.5739	16.2353	−68.2737	−47.0146	0.482
$\alpha^{\mathrm{E}} \times 10^5(\mathrm{K}^{-1})$	1.9180	−0.5614	0.2141	−2.1092	−4.4018		0.039

values for $\Delta\kappa_S$ are plotted against φ_i at various temperatures along with the smoothed V^E and $\Delta\kappa_S$ values by using Eq. 5 as in Figs. 1, 2, 3,4, 5, and 6. There are no literature values of ρ or u for these mixtures available for comparison. The calculated values of V^E and $\Delta\kappa_S$ of the binary mixtures are presented in the Appendix.

The volume or theoretical expansion coefficient α of the mixture at a particular composition was calculated for our systems by adding the contributions of the expansivities of each component in the mixtures;

$$\alpha = \frac{1}{\rho}\left[\frac{\delta\rho}{\delta T}\right] \tag{7}$$

In Fig. 7, α plots are shown for different mixtures at 298.15 K. We note that a sharp decrease in α is shown for the DPGMPE + BA or DBA systems as the concentration of ether increases.

From Figs. 1, 2, and 3, it can be seen that the V^E values are negative over the entire mole fraction range at all temperatures investigated for each binary system under study, except for DPGMPE + TBA, which exhibits positive V^E values at lower DPGMPE concentrations that become negative in the ether-rich region. This contrasts

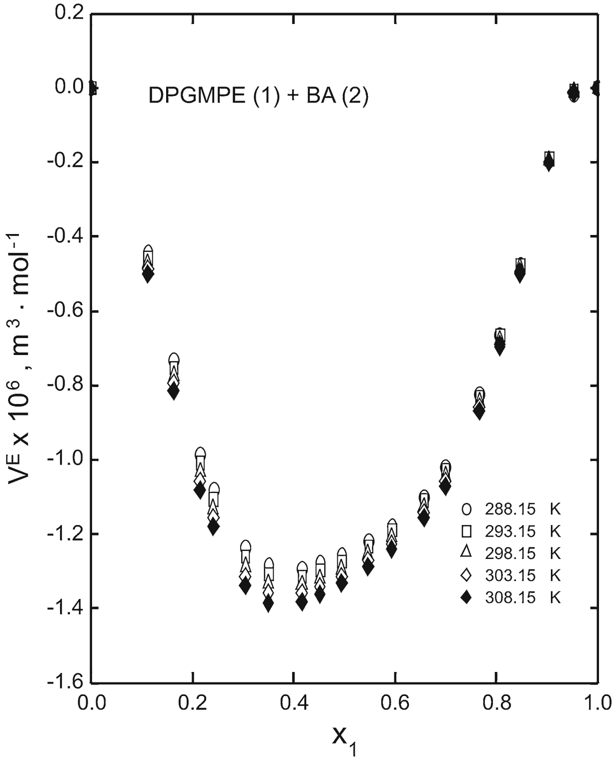

Fig. 1 Excess molar volumes, V^E for the DPGMPE (1) + butylamine (2) mixture at different temperatures

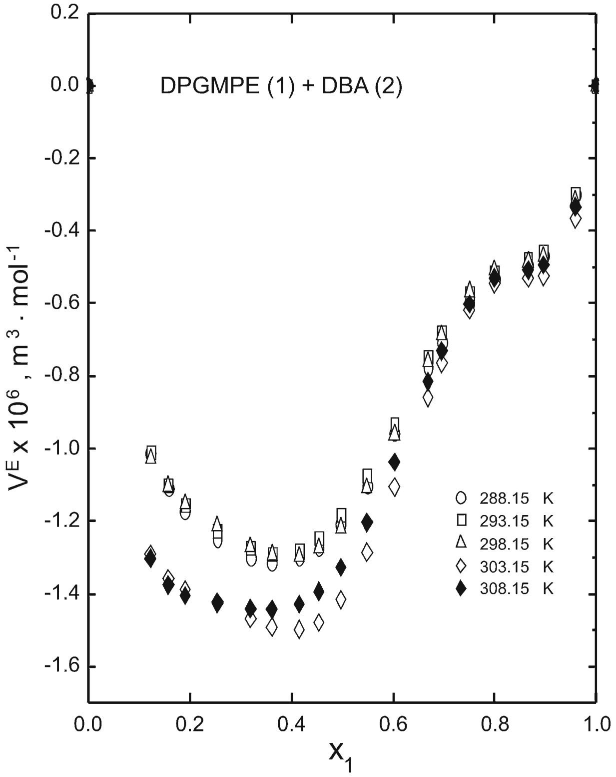

Fig. 2 Excess molar volumes, V^E for the DPGMPE (1) + dibutylamine (2) mixture at different temperatures

with the behavior of positive V^E results for dipropylene glycol monomethyl ether or dipropylene glycol monobutyl ether + tributylamine [9,10] but is consistent with that of the negative V^E values for butylamine, or DBA and dipropylene glycol monomethyl ether, dipropylene glycol monobutyl ether or dipropylene glycol *tert*-butyl ether + *n*-alkanol [14–16,29]. Moreover, their behavior may be compared with the V^E results for mixtures of DPGMME or DPGMBE with alkylamines [9,10] as shown in Fig. 8 with the alkyl chain length of the alkoxypropanol at $x_1 = 0.5$: the values of V^E decrease at each temperature with an increase of the alkyl chain length in dipropylene glycol monomethyl ether as in BA or TBA, and this becomes less negative from dipropylene glycol monomethyl ether to dipropylene glycol monobutyl ether with DBA.

The magnitude of the V^E values follows the sequence at lower temperatures: BA $<$ DBA $<$ TBA, but at higher temperatures the sequence is: DBA $<$ BA $<$ TBA. This suggests that there is an expansion in volume of the mixtures at lower temperatures as we move from butylamine to TBA. Furthermore, the behavior of V^E for DPGMPE + BA or DBA may be compared with the negative H^E results for (a primary or a secondary amine + an alkanol) [30–33], which can be considered as evidence for complex formation through the strong molecular interactions between the –NH group of the amine molecule and the –OH group of the alkanol molecules. The mixing

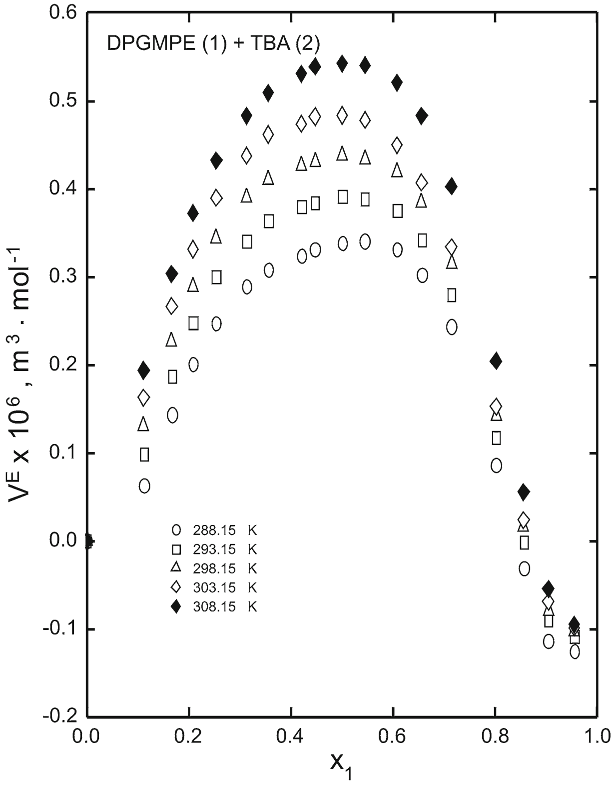

Fig. 3 Excess molar volumes, V^E for the DPGMPE (1) + tributylamine (2) mixture at different temperatures

of ether with the amines would induce mutual dissociation of the hydrogen-bonded structures, that is, O–H–O and N–H–N present in the pure liquids with subsequent formation of new N–H–O bonds between the proton acceptor nitrogen atom (with two unshared pair of electrons) of the amine and the hydrogen atom of the –OH group of the ether molecule. Equally important is the formation of an H-bond of the type N–H–O between the hydrogen atoms of the –NH group of amine and oxygen atom(s) of the –OH group(s) of ether molecules, leading to a contraction in volume, which should result in negative V^E values. A similar effect is observed for H^E (straight chain ether + butylamine or dibutylamine) [34,35]; with secondary amine H^E much more positive indicating less association than that between the primary butylamine and the ether. In the case of DPGMPE + TBA mixtures (Fig. 3), the positive V^E values at intermediate compositions can be considered as being due to the breaking up of associated structures present in the pure liquids leading to an expansion in volume; the decrease in V^E values at lower and higher concentrations of DPGMPE can be considered as being due to the formation of hydrogen bonds, leading to negative V^E values.

There is a systematic increase in V^E for DPGMPE + TBA and a decrease in V^E for DPGMPE + BA with a rise in temperature. In the case of DPGMPE + DBA, there

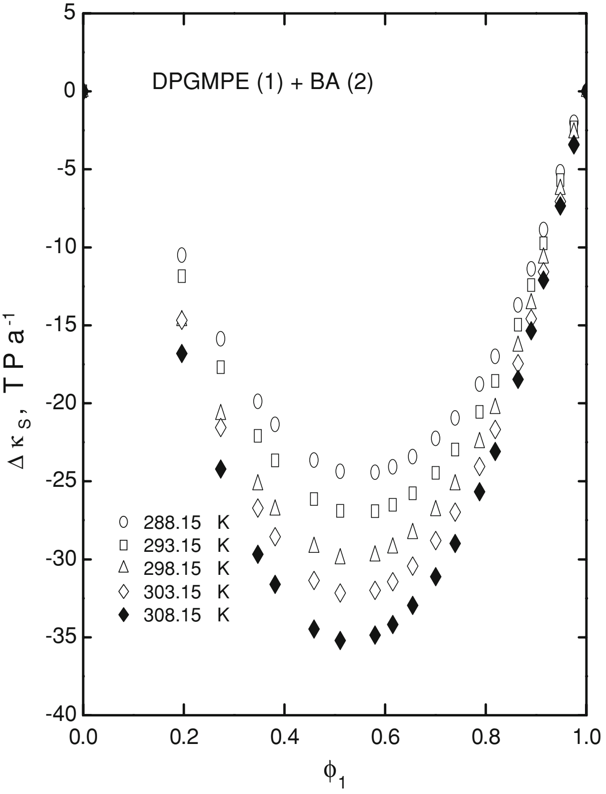

Fig. 4 Variation of deviations in isentropic compressibility, $\Delta\kappa_S$ for the DPGMPE (1)+butylamine (2) mixture at different temperatures

is no systematic variation of V^{E} with a rise in temperature. A comparison of data at different temperatures reveals that the temperature coefficient $\left(\delta V^{\mathrm{E}}/\delta T\right)_P$ is positive for TBA and negative for BA, indicating there is a breaking of H-bonded species formed between DPGMPE+TBA mixtures, which results in an expansion in volume with a rise in temperature. In the case of DPGMPE+BA mixtures, it is attributed to the formation of H-bonded associated species formed between dissimilar molecules with a rise in temperature, resulting in a contraction in volume of the mixture, and hence negative V^{E} values. The values of the excess thermal coefficient, α^{E} (not shown) are found to be negative over the whole composition range for {DPGMPE+butylamine (BA) or dibutylamine (DBA)} at all the investigated temperatures. In general, negative α^{E} values indicate the presence of strong interactions between the components in the mixtures [36]. The trends observed in α^{E} values for the present mixtures suggest the formation of H-bonding between dissimilar molecules which is stronger in {DPGMPE + butylamine (BA) or dibutylamine (DBA)}. The increasing negative α^{E} and $\left(\delta V^{\mathrm{E}}/\delta T\right)_P$ values for {DPGMPE + butylamine (BA) or dibutylamine (DBA)} with an increase in temperature indicate that more destruction of order during mixing [37] contributes negatively to $\left(\delta V^{\mathrm{E}}/\delta T\right)_P$ and α^{E}.

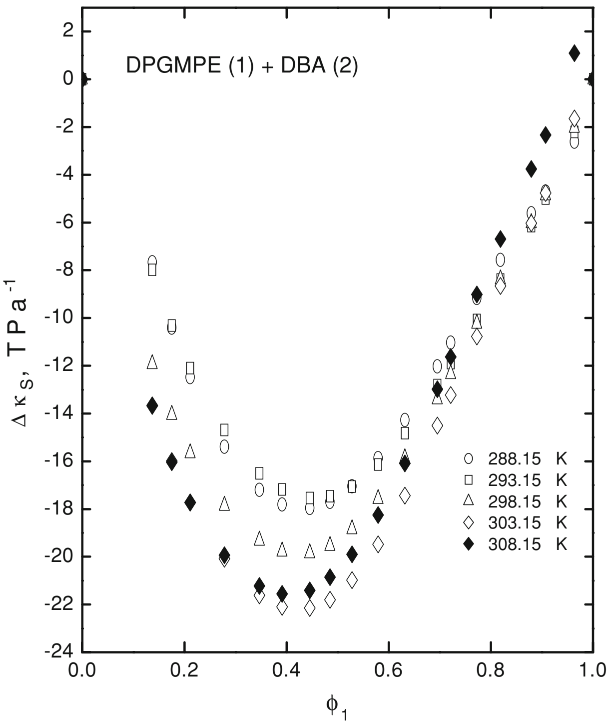

Fig. 5 Variation of deviations in isentropic compressibility, $\Delta\kappa_S$ for the DPGMPE (1) + dibutylamine (2) mixture at different temperatures

From Figs. 4, 5, and 6 it is seen that $\Delta\kappa_S$ is negative for butylamine (BA) and DBA, and positive to negative for TBA over the whole mole fraction range and at all the investigated temperatures. The values of $\Delta\kappa_S$ for all the three mixtures decrease from TBA to butylamine. Moreover, the behavior of the excess molar volume seems to be consistent with a minimum value of $\Delta\kappa_S$ with DPGMPE with butylamine (BA). Negative values of $\Delta\kappa_S$ mean that the mixture is less compressible than the corresponding ideal mixture, suggesting that there may be strong intermolecular hydrogen bonding with butylamine (BA) and DBA. As the ether is added to amines, thereby causing a breakdown of self-associated ether, or both contributing to a denser packing of all the molecules through hydrogen bonding, the speed of sound increases and $\Delta\kappa_S$ decreases.

The apparent molar volume $\left(V_{\varphi,1}\right)$ and the apparent molar compressibility $(K_{\varphi,1})$ properties of a solute component ether (1) in alkylamine, defined in terms of mole fraction concentration units, are calculated from the following relations [38,39]:

$$V_{\varphi,1} = V_1^* + \frac{V^{\mathrm{E}}}{x_1} \tag{8}$$

$$K_{\varphi,1} = K_{\varphi,1}^* + \frac{\left(\Delta\kappa_{S,m}\right)}{x_1} \tag{9}$$

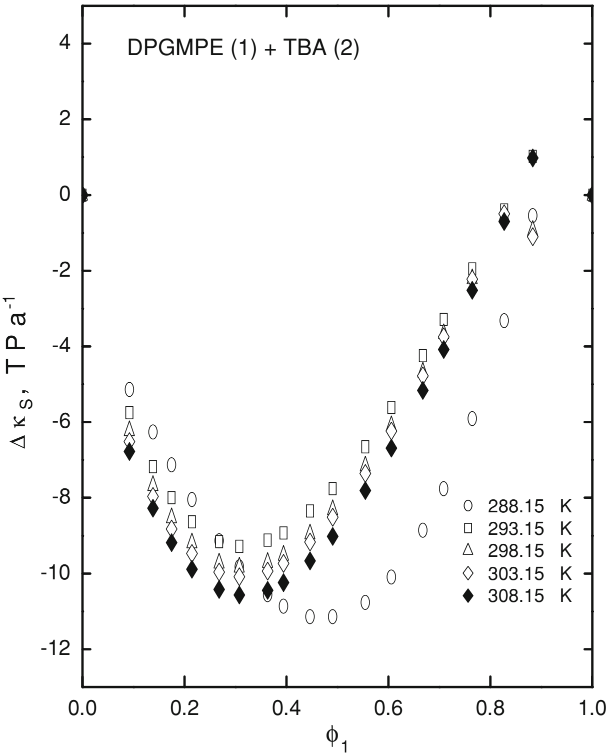

Fig. 6 Variation of deviations in isentropic compressibility, $\Delta\kappa_S$ for the DPGMPE (1) + tributylamine (2) mixture at different temperatures

where $K^*_{\varphi,1}$ is the molar isentropic compressibility $\left(\kappa^*_{S,m,1}\right)$. Simple graphical extrapolation of $V_{\varphi,1}$ and $K_{\varphi,1}$ to $x_1 = 0$ ($x_2 = 1$) and of $V_{\varphi,2}$ and $K_{\varphi,2}$ to $x_2 = 0$ ($x_1 = 1$) gives values of $V^0_{\varphi,1}$ or $V^0_{\varphi,2}$ and $K^0_{\varphi,1}$or $K^0_{\varphi,2}$ at infinite dilution, represented by $\bar{V}^0_{\varphi,1}$ or $\bar{V}^0_{\varphi,2}$ and $\bar{K}^0_{\varphi,1}$ or $\bar{K}^0_{\varphi,2}$.

Partial molar volumes and partial molar isentropic compressibilities at infinite dilution and at different temperatures are listed in Tables 4 and 5. All of these $\bar{V}^0_{\varphi,1}$ values for DPGMPE in various alkylamines are smaller than the corresponding V^*_1 values of pure DPGMPE except for TBA. Thus, the difference in values $\left(= \bar{V}^0_{\varphi,1} - V^*_1\right)$ became more negative with increasing temperature, suggesting that the molar volumes of each component in the mixture are less than their respective molar volumes in the pure state, i.e., there is contraction in volume of the mixture on mixing DPGMPE/BA/DBA. This may be attributed to the presence of significant interactions between DPGMPE and BA or DBA molecules. For the DPGMPE + TBA system, the differences in values at each temperature are positive suggesting an expansion in volume. This further supports the trends observed in V^{E} values for these binary systems that are attributed to similar types of interactions as discussed above for the V^{E} values.

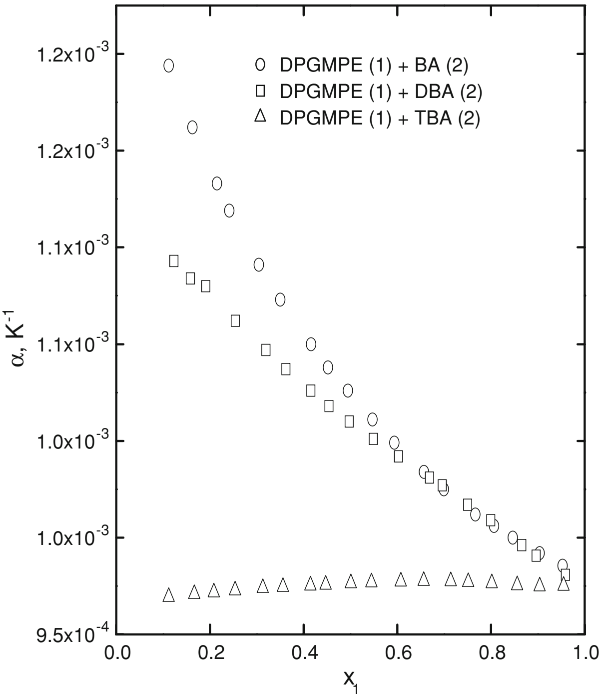

Fig. 7 Variation of thermal expansivity α for binary mixtures of DPGMPE (1) + alkylamines (2) at 298.15 K

We also observe that the values of $V^0_{\varphi,2}$ are smaller than the corresponding molar volumes V^*_2 of alkylamine with DPGMPE, for which the values of $\bar{V}^0_{\varphi,2}$ are lower. Thus, the differences in values $\left(\bar{V}^0_{\varphi,2} - V^*_2\right)$ are negative for all alkylamines at all temperatures. The negative values suggest that the interactions between dissimilar molecules exceed the structure breaking effect between similar molecules. These interactions are relatively strong between DPGMPE and BA, as suggested from V^{E} data.

Furthermore, a close perusal of Table 4 indicates that the differences between $\bar{K}^0_{\varphi,1}$ and $K^*_{\varphi,1}$ are large and negative for DPGMPE + BA or DBA as compared with DPGMPE + TBA for which the differences in values are positive evidenced that the strength of interactions in these mixtures follows the order TBA $<$ DBA $<$ BA. Again, the difference between $\bar{K}^0_{\varphi,2}$ and $K^*_{\varphi,2}$ (Table 5) are large and negative for lower alkylamines. This is consistent with the view that larger association between the DPGMPE and BA or DBA molecules takes place, thereby creating conditions of highly efficient packing and hence relatively lower compressibilities.

Recently, Jouyban and Acree [40,41] proposed a model for correlating the density and viscosity of liquid mixtures at various temperatures. The application of the model could be extended for calculating the density and ultrasonic speed of binary mixtures at various temperatures. The proposed eq. is

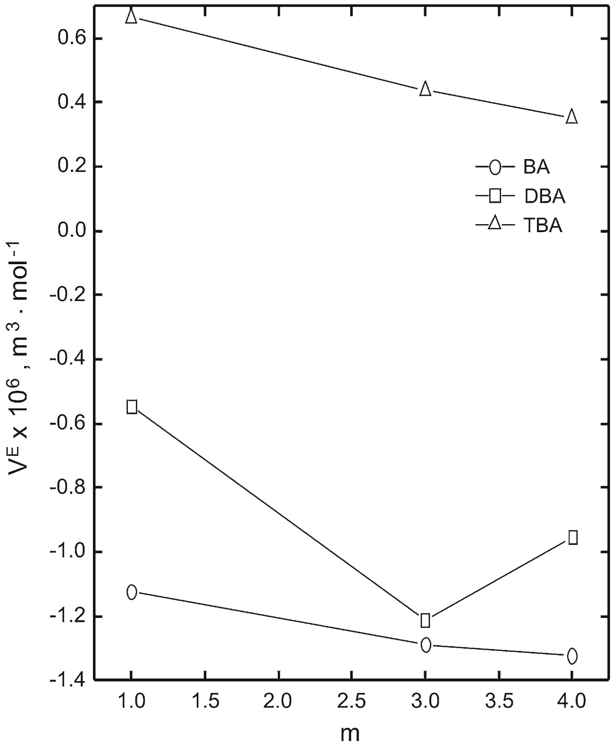

Fig. 8 Experimental excess molar volumes, V^E at equimolar concentration for $x_1(C_mH_{2m+1}\{(OCH_3)_2\}OH)+x_2C_4H_9NH_2$ (○); $(C_4H_9)_2NH$ (□); $(C_4H_9)_3N$ (Δ) at 298.15 K

Table 4 Values of V_1^*, $\bar{V}^0_{\varphi,1}$, and $\overline{K}^0_{\varphi,1}$ for (DPGMPE + *n*-alkylamine) systems at different temperatures

	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
DPGMPE (1) + butylamine (2)					
$V_1^* \times 10^6 (m^3 \cdot mol^{-1})$	190.691	191.592	192.520	193.459	194.405
$K^*_{S,m,1} (mm^3 \cdot mol^{-1} \cdot MPa^{-1})$	120.903	125.565	130.408	135.540	140.806
$\overline{K}^0_{\varphi,1} (mm^3 \cdot mol^{-1} \cdot MPa^{-1})$	106.473	109.259	110.206	115.237	117.470
$\bar{V}^0_{\varphi,1} \times 10^6 (m^3 \cdot mol^{-1})$	186.457	187.186	187.954	188.725	189.515
DPGMPE (1) + dibutylamine (2)					
$V_1^* \times 10^6 (m^3 \cdot mol^{-1})$	190.691	191.592	192.520	193.459	194.405
$K^*_{S,m,1} (mm^3 \cdot mol^{-1} \cdot MPa^{-1})$	120.903	125.565	130.408	135.540	140.806
$\overline{K}^0_{\varphi,1} (mm^3 \cdot mol^{-1} \cdot MPa^{-1})$	101.578	104.391	100.988	99.952	104.138
$\bar{V}^0_{\varphi,1} \times 10^6 (m^3 \cdot mol^{-1})$	180.072	181.017	181.743	179.758	180.409
DPGMPE (1) + tributylamine (2)					
$V_1^* \times 10^6 (m^3 \cdot mol^{-1})$	190.691	191.592	192.520	193.459	194.405
$K^*_{S,m,1} (mm^3 \cdot mol^{-1} \cdot MPa^{-1})$	120.903	125.565	130.408	135.540	140.806
$\overline{K}^0_{\varphi,1} (mm^3 \cdot mol^{-1} \cdot MPa^{-1})$	130.244	139.611	145.439	154.258	162.00
$\bar{V}^0_{\varphi,1} \times 10^6 (m^3 \cdot mol^{-1})$	190.815	192.134	193.768	195.053	196.329

Table 5 Values of V_2^*, $\overline{V}^0_{\varphi,2}$, and $\overline{K}^0_{\varphi,2}$ for (DPGMPE + *n*-alkylamine) systems at different temperatures

	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
DPGMPE (1) + butylamine (2)					
$V_2^* \times 10^6$ $(\mathrm{m^3 \cdot mol^{-1}})$	98.451	99.090	99.738	100.399	101.072
$K^*_{S,m,2}$$(\mathrm{mm^3 \cdot mol^{-1} \cdot MPa^{-1}})$	78.890	82.770	87.050	91.260	96.080
$\overline{K}^0_{\varphi,2}$ $(\mathrm{mm^3 \cdot mol^{-1} \cdot MPa^{-1}})$	70.808	73.585	76.456	80.920	82.135
$\overline{V}^0_{\varphi,2} \times 10^6$$(\mathrm{m^3 \cdot mol^{-1}})$	97.892	99.303	99.277	99.841	100.561
DPGMPE (1) + dibutylamine (2)					
$V_2^* \times 10^6$ $(\mathrm{m^3 \cdot mol^{-1}})$	168.703	169.609	170.557	171.804	172.749
$K^*_{S,m,2}$$(\mathrm{mm^3 \cdot mol^{-1} \cdot MPa^{-1}})$	132.490	138.110	144.810	151.770	158.500
$\overline{K}^0_{\varphi,2}$ $(\mathrm{mm^3 \cdot mol^{-1} \cdot MPa^{-1}})$	132.148	137.567	143.408	149.861	156.404
$\overline{V}^0_{\varphi,2} \times 10^6$$(\mathrm{m^3 \cdot mol^{-1}})$	168.015	168.907	170.076	170.724	171.640
DPGMPE (1) + tributylamine (2)					
$V_2^* \times 10^6$ $(\mathrm{m^3 \cdot mol^{-1}})$	237.226	238.356	239.501	240.661	241.836
$K^*_{S,m,2}$$(\mathrm{mm^3 \cdot mol^{-1} \cdot MPa^{-1}})$	183.860	191.130	198.810	206.880	215.360
$\overline{K}^0_{\varphi,2}$ $(\mathrm{mm^3 \cdot mol^{-1} \cdot MPa^{-1}})$	184.250	192.002	199.890	208.167	216.791
$\overline{V}^0_{\varphi,2} \times 10^6$$(\mathrm{m^3 \cdot mol^{-1}})$	237.107	238.256	239.423	240.660	271.792

$$\ln y_{m,T} = f_1 \ln y_{1,T} + f_2 \ln y_{2,T} + f_1 f_2 \sum \left[\frac{A_j \left(f_1 - f_2\right)^j}{T} \right] \tag{10}$$

where $y_{m,T}$, $y_{1,T}$, and $y_{2,T}$ are the density, or speed of sound of the mixture and pure liquids 1 and 2 at temperature T, respectively, f_1 and f_2 are the volume fraction, and A_j's are the model constants.

In order to perform a numerical comparison of the prediction capability of the model, the experimental density and ultrasonic speed data at different temperatures were fitted with the model and the average percentage deviation (*APD*) was calculated as

$$APD = \frac{100}{N} \sum \left[\left(\frac{\left| y_{\mathrm{exptl}} - y_{\mathrm{calcd}} \right|}{y_{\mathrm{exptl}}} \right) \right] \tag{11}$$

where N represents the number of experimental data in each data set. The optimum number of constants A_j, in each case, was determined from an examination of the *APD* values.

The Jouyban–Acree model was applied to ultrasonic speed measurements by Hasan et al. [42], We also extended the Jouyban–Acree model (Eq. 10) to the ultrasonic speeds of the present liquid mixtures with f as the mole fraction and again apply Eq. 11 to test the prediction capability of the model. The A'_js, calculated from the least-squares analysis, are presented in Table 6 along with the *APD*s. The *APD* values show the reliability of the proposed model for calculating the density and ultrasonic speed of binary mixtures at various temperatures. Hence, the proposed model provides rea-

Table 6 Parameters of Jouyban–Acree model and average percentage deviation (*APD*) for densities and speeds of sound for (DPGMPE + *n*-alkylamine) systems at different temperatures

		T (K)	A_0	A_1	A_2	*APD*
DPGMPE (1) + butylamine (2)	$\rho \times 10^{-3}$ (kg · m^{-3})	288.15	56.74957	−23.7233	5.795419	0.0538
		293.15	58.34686	−24.5454	5.540137	0.0411
		298.15	59.58737	−25.0879	6.371352	0.0440
		303.15	61.03852	−25.7656	6.710716	0.0423
		308.15	62.49109	−26.4940	7.021531	0.0423
	u (m · s^{-1})	288.15	9.672058	1.343817	−4.03342	0.0001
		293.15	11.32482	0.853399	−4.00755	0.0004
		298.15	13.19732	−0.45308	−2.51845	0.0003
		303.15	14.37761	−0.1203	−3.3621	0.0189
		308.15	16.20765	−1.75116	−2.82016	0.0014
DPGMPE (1) + dibutylamine (2)	$\rho \times 10^{-3}$ (kg · m^{-3})	288.15	18.96882	−7.29198	5.609804	0.0490
		293.15	19.12154	−7.39076	5.732803	0.0402
		298.15	19.57185	−7.41368	5.544921	0.0420
		303.15	21.05128	−9.16567	7.141441	0.0432
		308.15	20.88686	−9.54078	8.311552	0.0306
	u (m · s^{-1})	288.15	4.957656	−0.72026	−5.23072	0.0004
		293.15	4.589245	−0.726067	−4.54235	0.0000
		298.15	5.235984	−2.61397	−2.03385	0.0009
		303.15	5.647162	−3.25879	−3.54015	0.0003
		308.15	4.66332	−5.16339	−6.83895	0.0008
DPGMPE (1) + tributylamine (2)	$\rho \times 10^{-3}$ (kg · m^{-3})	288.15	−8.39137	−0.52937	1.352214	0.0411
		293.15	−8.89191	0.146168	2.896866	0.0325
		298.15	−9.28547	0.216495	2.871619	0.0316
		303.15	−9.66537	0.429425	2.928542	0.0286
		308.15	−10.5424	2.13496	8.311727	0.0401
	u (m · s^{-1})	288.15	−10.1944	1.644387	8.26589	0.0013
		293.15	−7.84818	7.126027	2.940581	0.0009
		298.15	−8.65648	7.898814	3.40123	0.0009
		303.15	−9.15511	8.523495	3.604664	0.0010
		308.15	−9.95665	8.909275	3.746285	0.0050

sonably accurate calculations for the density and ultrasonic speed of binary liquid mixtures at various temperatures.

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Appendix

Table A 1 Excess molar volume, V^{E}, and deviations in isentropic compressibility, $\Delta\kappa_S$, of binary mixtures as a function of mole fraction, x_1, of DPGMPE at different temperatures

x_1	$V^{\mathrm{E}} \times 10^6(\mathrm{m}^3 \cdot \mathrm{mol}^{-1})$					$\Delta\kappa_S$ (TPa^{-1})				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	288.15 K	293.15*K*	298.15 K	303.15 K	308.15 K
DPGMPE (1) + butylamine (2)										
0.1119	−0.4410	−0.4566	−0.4712	−0.4863	−0.5001	−10.504	−11.857	−14.689	−14.674	−16.801
0.1625	−0.7314	−0.7526	−0.7726	−0.7933	−0.8138	−15.863	−17.680	−20.707	−21.559	−24.210
0.2153	−0.9842	−1.0076	−1.0320	−1.0559	−1.0811	−19.881	−22.080	−25.222	−26.708	−29.672
0.2414	−1.0810	−1.1052	−1.1311	−1.1560	−1.1810	−21.353	−23.669	−26.840	−28.546	−31.612
0.3043	−1.2349	−1.2593	−1.2861	−1.3131	−1.3386	−23.634	−26.143	−29.237	−31.356	−34.468
0.3500	−1.2830	−1.3063	−1.3333	−1.3582	−1.3835	−24.359	−26.893	−29.959	−32.151	−35.204
0.4162	−1.2933	−1.3122	−1.3361	−1.3593	−1.3807	−24.416	−26.918	−29.789	−31.988	−34.860
0.4519	−1.2774	−1.2950	−1.3191	−1.3413	−1.3602	−24.075	−26.505	−29.273	−31.437	−34.179
0.4946	−1.2563	−1.2722	−1.2934	−1.3145	−1.3305	−23.424	−25.761	−28.355	−30.441	−32.951
0.5470	−1.2194	−1.2326	−1.2524	−1.2718	−1.2887	−22.253	−24.452	−26.881	−28.797	−31.110
0.5938	−1.1773	−1.1889	−1.2074	−1.2257	−1.2412	−20.948	−22.971	−25.210	−26.977	−28.989
0.6567	−1.0991	−1.1083	−1.1267	−1.1431	−1.1569	−18.773	−20.543	−22.518	−24.051	−25.675
0.6997	−1.0199	−1.0273	−1.0433	−1.0587	−1.0709	−16.995	−18.571	−20.316	−21.680	−23.079
0.7665	−0.8248	−0.8301	−0.8447	−0.8586	−0.8668	−13.686	−14.966	−16.343	−17.450	−18.455
0.8068	−0.6629	−0.6644	−0.6774	−0.6894	−0.6979	−11.380	−12.415	−13.601	−14.580	−15.356
0.8466	−0.4762	−0.4752	−0.4838	−0.4944	−0.5008	−8.869	−9.735	−10.682	−11.554	−12.103
0.9038	−0.1966	−0.1899	−0.1935	−0.2009	−0.2015	−5.162	−5.694	−6.280	−7.067	−7.339
0.9528	−0.0152	−0.0060	−0.0109	−0.0126	−0.0095	−1.980	−2.304	−2.647	−3.384	−3.436
DPGMPE (1) + dibutylamine (2)										
0.1230	−1.0146	−1.0089	−1.0276	−1.2906	−1.3028	−7.661	−7.986	−11.921	−13.660	−13.686
0.1582	−1.1108	−1.0998	−1.1028	−1.3596	−1.3758	−10.402	−10.315	−14.044	−15.996	−16.037
0.1913	−1.1720	−1.1548	−1.1511	−1.3876	−1.4055	−12.482	−12.096	−15.654	−17.726	−17.736
0.2541	−1.2496	−1.2279	−1.2120	−1.4222	−1.4270	−15.391	−14.690	−17.857	−20.084	−19.935
0.3195	−1.3013	−1.2750	−1.2703	−1.4680	−1.4405	−17.200	−16.503	−19.314	−21.613	−21.227
0.3623	−1.3141	−1.2897	−1.2924	−1.4919	−1.4436	−17.811	−17.182	−19.771	−22.094	−21.560
0.4155	−1.3036	−1.2792	−1.2981	−1.4990	−1.4285	−17.954	−17.541	−19.847	−22.143	−21.413

Table A1 continued

x_1	$V^E \times 10^6 (m^3 \cdot mol^{-1})$					$\Delta\kappa_S$ (TPa^{-1})				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	288.15 K	293.15 *K*	298.15 K	303.15 K	308.15 K
0.4543	−1.2735	−1.2465	−1.2736	−1.4794	−1.3936	−17.702	−17.467	−19.536	−21.801	−20.858
0.4974	−1.2096	−1.1818	−1.2177	−1.4155	−1.3272	−17.050	−17.061	−18.836	−20.983	−19.895
0.5491	−1.1029	−1.0733	−1.1086	−1.2863	−1.2034	−15.862	−16.141	−17.570	−19.477	−18.255
0.6027	−0.9587	−0.9325	−0.9610	−1.1065	−1.0371	−14.269	−14.830	−15.865	−17.446	−16.084
0.6687	−0.7769	−0.7479	−0.7597	−0.8587	−0.8160	−12.029	−12.815	−13.397	−14.510	−12.978
0.6962	−0.7065	−0.6782	−0.6859	−0.7643	−0.7302	−11.028	−11.891	−12.339	−13.226	−11.635
0.7504	−0.5993	−0.5711	−0.5658	−0.6173	−0.6010	−9.161	−10.081	−10.224	−10.768	−9.013
0.79970	−0.5350	−0.5122	−0.5068	−0.5449	−0.5298	−7.561	−8.393	−8.380	−8.657	−6.694
0.8657	−0.4958	−0.4769	−0.4854	−0.5297	−0.5067	−5.611	−6.149	−6.018	−6.031	−3.759
0.8963	−0.4685	−0.4548	−0.4694	−0.5243	−0.4931	−4.707	−5.000	−4.850	−4.767	−2.333
0.9589	−0.2991	−0.2966	−0.3161	−0.3660	−0.3336	−2.608	−2.205	−2.019	−1.644	1.099
DPGMPE (1) + tributylamine (2)										
0.1122	0.0636	0.0990	0.1315	0.1630	0.1935	3.633	4.713	5.670	6.479	7.271
0.1665	0.1438	0.1867	0.2262	0.2665	0.3035	4.066	5.555	6.603	7.478	8.815
0.2084	0.2012	0.2479	0.2887	0.3314	0.3726	4.466	5.920	7.020	7.932	8.965
0.2538	0.2468	0.2997	0.3434	0.3896	0.4323	4.887	6.037	7.184	8.086	9.193
0.3130	0.2896	0.3407	0.3900	0.4372	0.4829	5.390	5.866	7.014	7.861	9.000
0.3558	0.3085	0.3634	0.4097	0.4618	0.5093	5.695	5.515	6.613	7.467	8.623
0.4148	0.3244	0.3800	0.4263	0.4761	0.5307	5.984	4.752	5.840	6.611	7.770
0.4472	0.3309	0.3839	0.4307	0.4818	0.5386	6.083	4.277	5.341	6.069	7.201
0.5008	0.3384	0.3918	0.4386	0.4835	0.5421	6.115	3.339	4.366	5.011	6.071
0.5451	0.3406	0.3885	0.4341	0.4779	0.5405	5.993	2.533	3.449	4.027	5.047
0.6077	0.3312	0.3755	0.4190	0.4497	0.5208	5.573	1.305	2.088	2.532	3.430
0.6566	0.3024	0.3424	0.3847	0.4073	0.4827	4.938	0.272	0.952	1.249	2.091
0.7143	0.2435	0.2798	0.3153	0.3338	0.4025	3.854	−0.9424	−0.407	−0.246	0.423
0.7514	0.1830	0.2187	0.2502	0.3645	0.3259	2.896	−1.739	−1.318	−1.231	−0.705
0.8016	0.0859	0.1172	0.1427	0.1526	0.2040	1.370	−2.732	−2.496	−2.571	−2.195
0.8560	−0.0311	−0.0011	0.0163	0.0235	0.0556	−0.6529	−3.684	−3.642	−3.863	−3.720
0.9040	−0.1136	−0.0896	−0.0792	−0.0688	−0.0543	−2.6157	−4.293	−4.381	−4.734	−4.789
0.9553	−0.1255	−0.1078	−0.1028	−0.0983	−0.0949	−4.681	−4.449	−4.725	−5.233	−5.462

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