Densities, Excess Molar Volumes, Speeds of Sound, and Isothermal Compressibilities for 2-(2-Hexyloxyethoxy)ethanol + *n*-Alkylamine at Temperatures Between 288.15 K and 308.15 K

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The densities, ρ, and the speeds of sound, u, for binary liquid mixtures containing 2-(2-hexyloxyethoxy)ethanol (C_6E_2) and *n*-butylamine (BA), dibutylamine (DBA), and tributylamine (TBA) have been measured as a function of composition using a vibrating tube densimeter and sound analyzer Anton-Paar model DSA-5000 at temperatures (288.15, 293.15, 298.15, 303.15, and 308.15) K and atmospheric pressure. The ρ and u values were used to calculate excess molar volumes, V_m^E, and excess molar isentropic compressibility, $K_{S,m}^E$, respectively. The V_m^E and $K_{S,m}^E$ values are shown to be negative for all the mixtures over the entire composition range. Both V_m^E and $K_{S,m}^E$ become more negative for all the mixtures with the rise of temperature. The change of V_m^E and $K_{S,m}^E$ with composition and temperature are discussed with a view to understand molecular interactions present in alkoxyethanol-alkylamine mixtures.

1. Introduction

As part of our program of research on thermodynamic, acoustic, and transport properties of binary mixtures containing alkoxyethanols or alkoxypropanols, we have recently reported excess molar volumes, viscosities, and speeds of sound measurements of binary mixtures of alkoxypropanols either with n-alkylamines,[1–3] 1-alkanols[4–7] or amides.[8] As a part of this continuing work, the experimental results are reported in this paper for the binary systems of 2-(2-hexyloxyethoxy)ethanol (C_6E_2) with *n*-butylamine, dibutylamine, and tributylamine at temperatures from 288.15 K to 308.15 K. The liquids were selected on the basis of their industrial use.[9–12] The aim of this work is to provide a set of data for the characterization of molecular interactions of the oxygen (−O−) and hydroxyl (−OH) groups of alkoxyethanol with the amine (−NH) group of alkylamines and to study the composition-temperature dependent behavior of these mixtures.

2. Experimental Section

Materials. Butylamine (BA), dibutylamine (DBA), and tributylamine (TBA), C_6E_2 are the same as used before.[13,14] Prior to experimental measurements, all 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 purities of liquids were checked by comparing the densities, and speeds of sound at the desired temperatures with their corresponding literature values.[15–21] This comparison is given in Table 1 and the experimental values are in good agreement with those from the literature. Also given in Table 1 are the measured and literature values of those quantities that were required in the estimation of $K_{S,m}$ and $K_{S,m}^E$.

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Apparatus and Procedure. The densities, ρ, and speeds of sound, u, of both pure liquids and of the mixtures were simultaneously and automatically measured using an Anton Paar DSA-5000 instrument.[14] The sensitivity of the instrument corresponded to a precision in density and speed of sound measurements of $1 \cdot 10^{-6}$ g·cm^{-3} and $1 \cdot 10^{-2}$ m·s^{-1}. The reproducibility of the density and speed of sound estimates was found to be within $\pm$ $5 \cdot 10^{-6}$ g·cm^{-3} and $\pm$ $5 \cdot 10^{-2}$ m·s^{-1}, respectively.

The 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 precision of $\pm$ 0.01 mg. The probable error in the mole fraction was estimated to be less than $\pm$ $1 \cdot 10^{-4}$. Conversion to molar quantities was based on the relative atomic mass table of 1985 issued by I.U.P.A.C.[22]

3. Results and Discussion

The experimental results of the density, ρ, and speed of sound, u, measurements of binary mixture of C_6E_2 with *n*-butylamine, dibutylamine and tributylamine with C_6E_2 as a common component over the whole composition range expressed as mole fractions, x_1 of C_6E_2 ($0 \le x_1 \le 1$) at different temperatures are listed in Table 2.

The excess molar volumes, V_m^E, were calculated by using the following relation

$$V^E = \sum_{i=1} x_i M_i (\rho^{-1} - \rho_i^{*-1}) \quad (1)$$

where ρ is the density and x_i and M_i are the mole fraction and molar mass of component i in the mixture, respectively.

The isentropic compressibility, κ_S, and the molar isentropic compressibility, $K_{S,m}$ have been calculated from the relations[23]

$$\kappa_S = (\rho \cdot u^2)^{-1} = V(M \cdot u^2)^{-1} \quad (2)$$

$$K_{S,m} = -(\delta V_m / \delta P)_s = V_m \kappa_S = \Sigma x_i M_i / (\rho u)^2 \quad (3)$$

The excess molar quantities were calculated from

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Table 1. Thermodynamic Parameter for Pure Components

	T	$\rho^* \cdot 10^{-3}$/kg·m^{-3}		$\alpha_P^* \cdot 10^{-3}$	$C_{P,m}^*$	u/m·s^{-1}	
	K	expt	lit	K^{-1}	J·mol^{-1}·K^{-1}	expt	lit
C_6E_2	288.15	0.936901		0.856[a]	417.94[b]	1405.17	
	293.15	0.932917		0.859[a]	420.62[b]	1388.08	
	298.15	0.928926		0.867[a]	423.30[b]	1371.14	
	303.15	0.924931		0.868[a]	426.00[b]	1354.30	
	308.15	0.920933		0.872[a]	428.78[b]	1337.65	
butylamine	288.15	0.742865		1.280[a]	185[b]	1296.08	
	293.15	0.738096	0.73869[17]	1.304[a]	187[b]	1273.56	
	298.15	0.733301	0.73359[17]	1.320[a]	189[b]	1249.96	1.250[18]
			0.73323[18]				1249.8[19]
	303.15	0.728477	0.72849[17]	1.337[a]	191[b]	1228.92	1227[18]
			0.72842[18]				
	308.15	0.723625	0.72339[17]	1.360[a]	192[b]	1205.72	1204[18]
			0.72364[18]				
dibutylamine	288.15	0.766115		1.02[a]	298[b]	1289.23	
	293.15	0.762022		1.180[a]	300[b]	1269.47	
	298.15	0.757784	0.75572[15]	1.200[a]	302[b]	1246.69	1248[18]
			0.75770[20]				
	303.15	0.752284		1.250[a]	304[b]	1226.70	1227[18]
	308.15	0.748170		1.330[a]	306[b]	1206.97	1206[18]
tributylamine	288.15	0.781336		0.949[a]	388[b]	1285.04	
	293.15	0.777632		0.959[a]	390[b]	1266.37	
	298.15	0.773913	0.77391[18]	0.967[a]	392[b]	1247.64	1246.9[19]
	303.15	0.770183	0.77046[18]	0.975[a]	396[b]	1228.98	
	308.15	0.766440	0.76695[18]	0.984[a]	400[b]	1210.42	

[a] Derived from our measured densities. [b] Experimental heat capacity from literature[15,16,19] and computed from group additivity.[21,22]

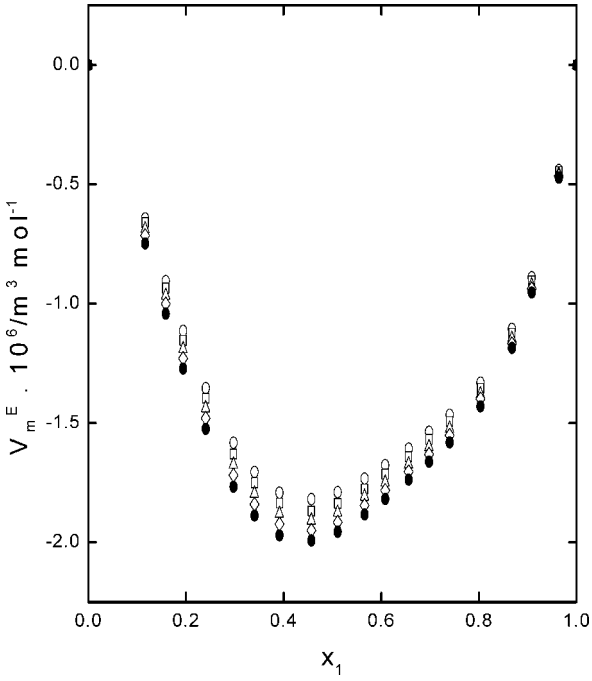

Figure 1. Excess molar volumes V_m^E for the mixture C_6E_2 (1) + butylamine (2). ○, 288.15 K; , 293.15 K; Δ, 298.15 K; ◇, 303.15 K; ●, 308.15 K.

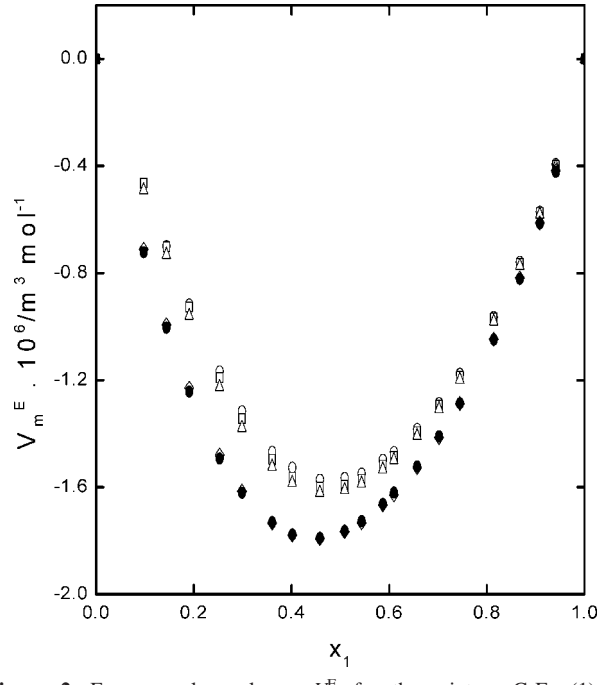

Figure 2. Excess molar volumes V_m^E for the mixture C_6E_2 (1) + dibutylamine (2). ○, 288.15 K; , 293.15 K; Δ, 298.15 K; ◇, 303.15 K; ●, 308.15 K.

$$K_{S,m}^E = K_{S,m} - K_{S,m}^{id} \quad (4)$$

where[24,25]

$$K_{S,m}^{id} = \Sigma x_i [K_{S,i}^* - TA_{P,i}^* \{(\Sigma x_i A_{P,i}^* / \Sigma x_i C_{P,i}^*) - (A_{P,i}^* / C_{P,i}^*)\}] \quad (5)$$

where $A_{P,i}^*$ is the product of the molar volume, V_i^*, and the isobaric expansivity, $\alpha_{P,i}^*$, $C_{P,i}^*$, is the isobaric molar heat capacity, and $K_{S,i}^*$ are properties of the pure liquid component i.

For each mixture, the values of V_m^E and $K_{S,m}^E$ were fitted by an equation of the type

$$F(x) = x_1 x_2 \sum_{i=0}^{k} a_i (x_1 - x_2)^i \quad (6)$$

The values of coefficients a_i were evaluated by using the methods of least-squares with all points weighted equally. The values of coefficients a_i for all the mixtures are listed in Table 3.

Figures 1 to 6 show V_m^E and $K_{S,m}^E$ data for the three mixtures at five temperatures. For each of the mixtures studied, V_m^E and $K_{S,m}^E$ are negative over the whole mole fraction range at all five temperatures and the minimum is shifted toward ether rich region. In fact, we observe similar characteristics for V_m^E as in

Table 2. Densities, ρ, and Ultrasonic Speeds, u, of Binary Mixtures As a Function of Mole Fraction, x_1, of C_6E_2 at Different Temperatures

	$\rho \cdot 10^{-3}$/kg·m^{-3}					u/m·s^{-1}				
x_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
					C_6E_2 (1) + butylamine (2)					
0.1165	0.788964	0.784340	0.779774	0.775186	0.770602	1322.97	1302.22	1281.50	1260.84	1240.34
0.1587	0.803568	0.799044	0.794520	0.790000	0.785486	1332.27	1311.99	1291.67	1271.49	1251.29
0.1943	0.814988	0.810552	0.806078	0.801606	0.797118	1339.54	1319.68	1299.68	1279.79	1259.85
0.2403	0.828571	0.824179	0.819765	0.815333	0.810880	1348.18	1328.73	1309.11	1289.63	1270.00
0.2974	0.843631	0.839297	0.834939	0.830566	0.826173	1357.78	1338.75	1319.61	1300.56	1281.34
0.3403	0.853769	0.849458	0.845129	0.840783	0.836421	1364.21	1345.54	1326.64	1307.87	1288.93
0.3916	0.864666	0.860386	0.856092	0.851780	0.847457	1371.09	1352.63	1334.08	1315.65	1297.07
0.4568	0.876866	0.872668	0.868407	0.864132	0.859843	1378.62	1360.44	1342.25	1324.16	1306.00
0.5105	0.885772	0.881607	0.877388	0.873158	0.868903	1383.90	1365.88	1348.00	1330.09	1312.26
0.5658	0.894125	0.889991	0.885768	0.881571	0.877355	1388.52	1370.68	1353.01	1335.29	1317.75
0.6081	0.900031	0.895896	0.891728	0.887547	0.883355	1391.55	1373.87	1356.27	1338.75	1321.25
0.6559	0.906287	0.902162	0.898025	0.893876	0.889716	1394.49	1376.94	1359.49	1342.08	1324.90
0.6977	0.911396	0.907303	0.903194	0.899074	0.894942	1396.68	1379.21	1361.85	1344.55	1327.53
0.7402	0.916322	0.912248	0.908162	0.904067	0.899964	1398.57	1381.20	1363.91	1346.71	1329.80
0.8029	0.923000	0.918949	0.914893	0.910805	0.906755	1400.81	1383.52	1366.38	1349.31	1332.55
0.8675	0.928954	0.924929	0.920910	0.916890	0.912833	1402.53	1385.39	1368.32	1351.36	1334.62
0.9082	0.932126	0.928121	0.924113	0.920095	0.916070	1403.37	1386.30	1369.33	1352.42	1335.69
0.9635	0.935472	0.931477	0.927494	0.923492	0.919484	1404.26	1387.36	1370.39	1353.58	1336.81
					C_6E_2 (1) + dibutylamine (2)					
0.0974	0.787886	0.783751	0.779614	0.775187	0.771079	1301.44	1282.29	1262.90	1243.68	1225.28
0.1441	0.798114	0.793988	0.789859	0.785667	0.781545	1308.12	1289.17	1270.55	1250.97	1232.23
0.1908	0.808103	0.804000	0.799885	0.795785	0.791662	1314.80	1295.95	1277.82	1258.20	1239.18
0.2530	0.820984	0.816934	0.812825	0.808731	0.804600	1323.47	1304.81	1287.05	1267.67	1248.65
0.2986	0.830092	0.826059	0.821948	0.817838	0.813671	1329.78	1311.25	1293.60	1274.45	1255.53
0.3606	0.842000	0.837970	0.833834	0.829692	0.825520	1338.03	1319.71	1302.12	1283.44	1264.84
0.4018	0.849578	0.845585	0.841423	0.837274	0.833078	1343.36	1325.25	1307.60	1289.30	1270.93
0.4584	0.859635	0.855602	0.851460	0.847291	0.843095	1350.40	1332.64	1314.96	1297.05	1279.08
0.5091	0.868247	0.864218	0.860074	0.855914	0.851724	1356.69	1339.01	1321.35	1303.79	1286.18
0.5435	0.873936	0.869891	0.865751	0.861602	0.857401	1360.77	1343.21	1325.65	1308.19	1290.82
0.5870	0.880891	0.876847	0.872720	0.868580	0.864406	1365.80	1348.42	1330.88	1313.63	1296.48
0.6097	0.884459	0.880409	0.876278	0.872165	0.867972	1368.36	1351.05	1333.56	1316.39	1299.36
0.6572	0.891716	0.887648	0.883547	0.879461	0.875302	1373.58	1356.39	1339.05	1321.98	1305.05
0.7023	0.898412	0.894343	0.890259	0.886201	0.882052	1378.35	1361.27	1344.12	1327.00	1310.12
0.7451	0.904586	0.900530	0.896459	0.892397	0.888281	1382.71	1365.70	1348.64	1331.58	1314.55
0.8144	0.914195	0.910151	0.906103	0.902062	0.897980	1389.39	1372.42	1355.55	1338.46	1320.88
0.8675	0.921188	0.917168	0.913137	0.909095	0.905045	1394.19	1377.16	1360.35	1343.32	1325.02
0.9083	0.926329	0.922317	0.918300	0.914270	0.910231	1397.68	1380.58	1363.70	1346.75	1327.65
0.9414	0.930322	0.926329	0.922323	0.918281	0.914269	1400.38	1383.16	1366.18	1349.33	1329.46
					C_6E_2 (1) + tributylamine (2)					
0.1168	0.797427	0.793696	0.789759	0.785812	0.782055	1294.00	1274.86	1255.97	1237.35	1219.04
0.1659	0.804490	0.800716	0.796756	0.792800	0.788975	1298.17	1279.15	1260.30	1241.70	1223.38
0.2092	0.810815	0.806993	0.803027	0.799078	0.795200	1302.06	1283.10	1264.34	1245.84	1227.47
0.2589	0.818158	0.814291	0.810334	0.806371	0.802458	1306.80	1287.92	1269.25	1250.78	1232.48
0.3155	0.826596	0.822690	0.818738	0.814795	0.810825	1312.56	1293.77	1275.16	1256.82	1238.57
0.3564	0.832755	0.828829	0.824867	0.820924	0.816944	1316.92	1298.20	1279.68	1261.41	1243.23
0.3830	0.836791	0.832842	0.828881	0.824930	0.820957	1319.83	1301.19	1282.70	1264.51	1246.36
0.4544	0.847683	0.843729	0.839761	0.835783	0.831821	1328.05	1309.53	1291.20	1273.20	1255.19
0.5108	0.856418	0.852463	0.848467	0.844479	0.840528	1334.89	1316.53	1298.27	1280.40	1262.57
0.5568	0.863630	0.859691	0.855668	0.851665	0.847701	1340.69	1322.44	1304.29	1286.52	1268.83
0.6107	0.872206	0.868299	0.864217	0.860198	0.856228	1347.74	1329.59	1311.58	1293.98	1276.38
0.6683	0.881543	0.877641	0.873531	0.869475	0.865486	1355.56	1337.54	1319.68	1302.25	1284.77
0.6924	0.885505	0.881587	0.877468	0.873410	0.869396	1358.91	1340.94	1323 0.16	1305.80	1288.36
0.7544	0.895808	0.891893	0.887746	0.883674	0.879592	1367.76	1349.97	1332.33	1315.15	1297.81
0.7963	0.902873	0.898935	0.894791	0.890730	0.886574	1373.92	1356.22	1338.74	1321.64	1304.36
0.8580	0.913335	0.909366	0.905245	0.901206	0.896983	1383.21	1365.72	1348.35	1331.46	1314.21
0.9079	0.921784	0.917773	0.913700	0.909674	0.905434	1390.94	1373.58	1356.38	1339.62	1322.35
0.9553	0.929676	0.925674	0.921630	0.917622	0.913460	1398.43	1381.26	1364.20	1347.56	1330.18

the mixture of alkoxypropanol with amines[1,2] but with a marked decrease in the values of V_m^E here. However, the V_m^E is less negative for the system (C_6E_2 + tributylamine). Furthermore, similar negative V_m^E results were found for butylamine + n-alkanol at a temperature of 298.15 K in the literatures,[26–28] indicating the dominance of strong specific interactions between components and the free volume effect when mixture is formed. The following effects can be considered: (i) contraction due to molar volume and free volume[29] differences of unlike molecules, and (ii) hydrogen bond formation between amine and ether through N-----H−O and N−H-----O, producing a negative contribution to V_m^E.

The V_m^E values were found to decrease at all five temperatures in the order TBA > DBA > BA. This behavior may be compared with the negative H_m^E results for (a primary or secondary amine + an alkanol):[26,30–32] strong molecular interactions between the −NH group of the amine molecule and the −OH group of the alkanol molecules. The observed very large negative values of V_m^E than the free volume contribution indicate the predominance of an energetically favored[33,34] cross bonding N-----H−O compared to N−H-----O over the hydrogen bonding between like molecule. A similar effect can be observed on H_m^E for (butylamine or dibutylamine + straight chain ether:[35,36] the secondary amine H_m^E is much more positive indicating less

Table 3. Coefficients, a_i, from Equation 6 and Standard Deviation, σ, for the Binary Mixtures at Different Temperatures

property	T/K	a_0	a_1	a_2	a_3	a_4	$\sigma(F(x))$
			C_6E_2 (1) + Butylamine (2)				
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	288.15	−7.1977	1.3921	−1.5648	−6.768		0.0022
	293.15	−7.3799	1.4987	−1.5768	−6.7052	−0.0057	0.0019
	298.15	−7.5338	1.6034	−1.6959	−6.8291		0.0017
	303.15	−7.6961	1.7061	−1.8093	−6.8957		0.0017
	308.15	−7.8574	1.7980	−1.9912	−6.8994	0.0647	0.0017
$K^E_{S,m}$ /mm$^3 \cdot$mol$^{-1} \cdot$MPa^{-1}	288.15	−30.2975	9.0462	−4.411	−7.5091	3.3908	0.008
	293.15	−33.0407	10.3761	−4.9681	−8.1326	3.3318	0.010
	298.15	−35.9665	11.6387	−5.5278	−7.3390	2.0469	0.006
	303.15	−38.6104	12.6647	−5.9293	−8.4012	2.6687	0.011
	308.15	−42.3007	13.8726	−7.1265	−6.7093	1.6611	0.006
			C_6E_2 (1) + Dibutylamine (2)				
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	288.15	−6.2794	0.5506	0.3530	−2.3009		0.0018
	293.15	−6.3853	0.7174	0.5018	−2.6541		0.0018
	298.15	−6.4580	0.8116	0.2696	−2.6028	0.0862	0.0017
	303.15	−7.0935	1.4106	−0.8400	−1.4339	−0.1458	0.0013
	308.15	−7.0757	1.4862	−1.0217	−1.4735	−0.8470	0.0012
$K^E_{S,m}$ /mm$^3 \cdot$mol$^{-1} \cdot$MPa^{-1}	288.15	−24.3014	3.4194	−0.0824	−5.2475	3.9554	0.013
	293.15	−27.8268	3.8266	−1.009	−4.9548	4.2259	0.012
	298.15	−33.7795	6.6956	−4.3846	−2.8518	4.7625	0.012
	303.15	−31.8693	6.004	−2.9923	1.4018	−0.7875	0.021
	308.15	−43.8278	5.4196	−4.7723	11.9647	5.0756	0.101
			C_6E_2 (1) + Tributylamine (2)				
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	288.15	−1.6943	−0.1372	−0.5138	−2.6243	−0.1503	0.0011
	293.15	−1.6091	−0.3991	−0.7353	−2.0966		0.0013
	298.15	−1.4869	−0.2745	−0.1971	−2.5703	−0.1159	0.0010
	303.15	−1.3835	−0.2201	0.0991	−0.30947		0.0018
	308.15	−1.3131	−0.4822	0.4739	−1.9720	−0.0794	0.0013
$K^E_{S,m}$ /mm$^3 \cdot$mol$^{-1} \cdot$MPa^{-1}	288.15	−12.3069	−2.6133	0.0008	−3.5550	−0.9633	0.007
	293.15	−12.6079	−3.4315	0.1188	−3.8935	−0.1089	0.013
	298.15	−12.8469	−3.6709	0.8882	−5.3500	0.5471	0.014
	303.15	−13.4479	−4.2421	1.6433	−6.8648	0.0306	0.021
	308.15	−13.9721	−5.4892	2.0883	−3.5216	0.6934	0.005

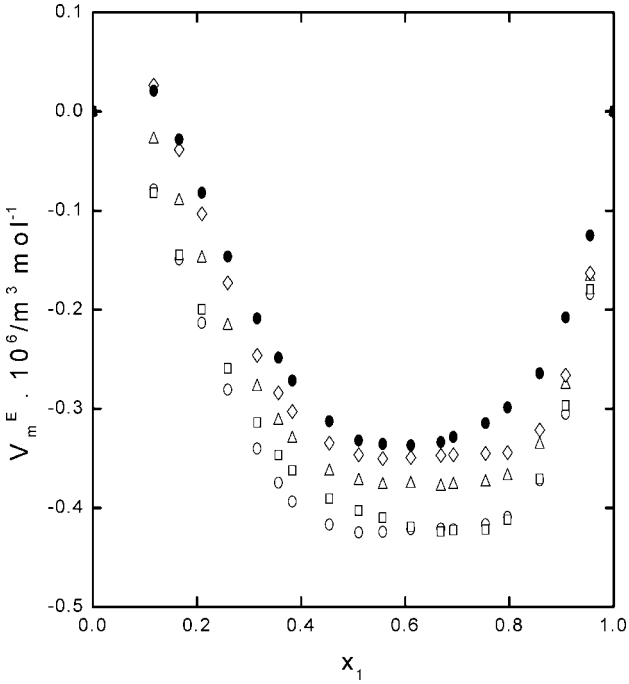

Figure 3. Excess molar volumes V^E_m for the mixture C_6E_2 (1) + tributylamine (2). ○, 288.15 K; , 293.15 K; Δ, 298.15 K; ◇, 303.15 K; ●, 308.15 K.

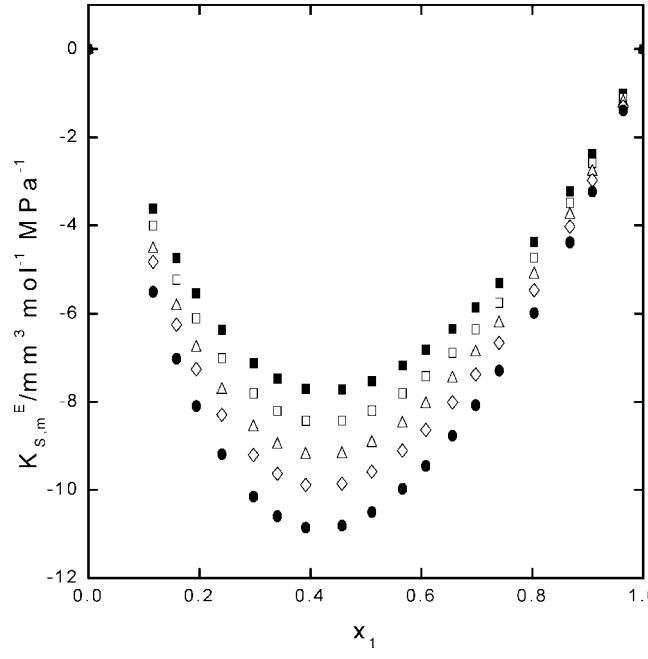

Figure 4. . Excess molar isentropic compressibility $K^E_{S,m}$ for the mixture C_6E_2 (1) + *n*-butylamine (2). ○, 288.15 K; , 293.15 K; Δ, 298.15 K; ◇, 303.15 K; ●, 308.15 K.

association than between the butylamine and the ether. The values of V^E_m decrease with an increase in temperature of the mixtures C_6E_2 + butylamine and dibutylamine (Figures 1 and 2) whereas the V^E_m values for C_6E_2 + tributylamine (Figure 3) increase with an increase in temperature. The decrease in V^E_m is attributed to the formation of hydrogen bonds between unlike molecules with increasing temperature, leading to closer packing of molecules in a contraction in volume of the mixture, and hence more negative V^E_m values. In the case of C_6E_2 + tributylamine mixtures, the increase in V^E_m values can be considered as being due to the breaking of H-bonded associated species formed between unlike molecules with a rise in temperature, leading to an expansion in volume, hence resulting in an increase in the V^E_m values.

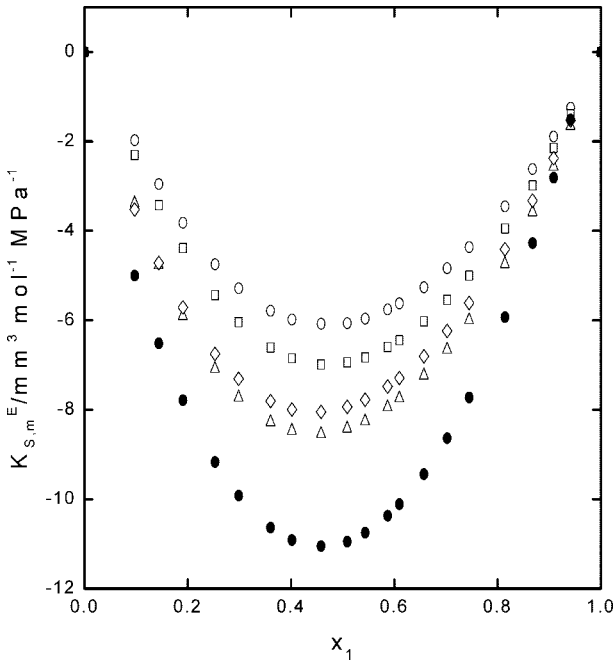

Figure 5. Excess molar isentropic compressibility $K_{S,m}^{E}$ for the mixture C_6E_2 (1) + dibutylamine (2). ○, 288.15 K; , 293.15 K; Δ, 298.15 K; ◇, 303.15 K; ●, 308.15 K.

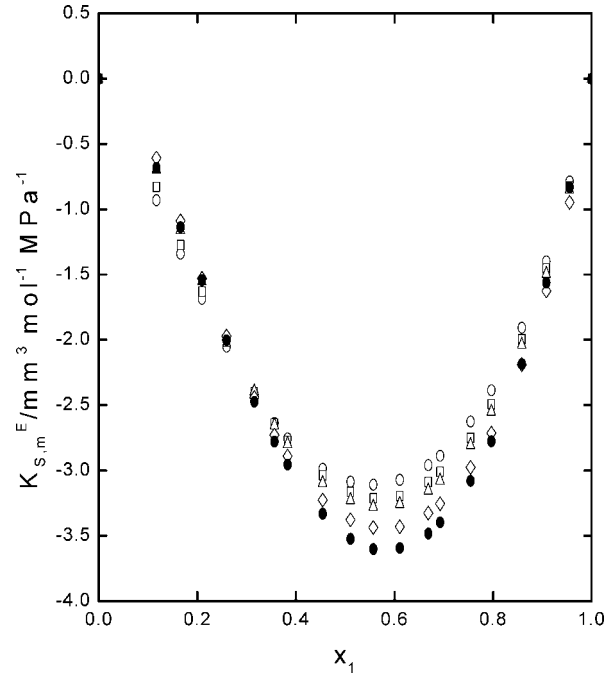

Figure 6. Excess molar isentropic compressibility $K_{S,m}^{E}$ for the mixture C_6E_2 (1) + tributylamine (2). ○, 288.15 K; , 293.15 K; Δ, 298.15 K; ◇, 303.15 K; ●, 308.15 K.

For all the mixtures studied, $K_{S,m}^{E}$ is negative over the whole mole fraction range at all five temperatures and shows a minimum in the sequence butylamine > dibutylamine > tributylamine. The behavior of excess molar volumes seems to be consistent with a minimum value of $K_{S,m}^{E}$ of C_6E_2 with n-alkylamines. Negative values of $K_{S,m}^{E}$ mean that the mixture is less compressible than the corresponding ideal mixture, suggesting that there may be strong intermolecular hydrogen bonding as in case of butylamine as compared to DBA and TBA. As the ether is added to amines thereby causing a breakdown of self-associated ether, or both contribute to a denser packing of all the molecules through hydrogen bonding, the speed of sound increases and $K_{S,m}^{E}$ decreases. As the temperature is lowered, the packing effect in all three mixtures is also lowered due to the decrease in the intermolecular interaction in C_6E_2 + alkylamine, hence resulting in an increase in $K_{S,m}^{E}$ values (Figures 4 to 6).

Supporting Information Available:

Table of excess molar volume and excess molar isentropic compressibility of binary mixtures as a function of mole fraction of C_6E_2 at different temperatures. This material is available free of charge via the Internet at http://pubs.acs.org.

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