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Molar Volume, Refractive Index, and Isentropic Compressibility at 298.15 K for 1-Butanol + Ethanol + 2-Methoxy-2-methylpropane

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Densities, refractive indices, and isentropic compressibility were measured for the system 1-butanol + ethanol + 2-methoxy-2-methylpropane at 298.15 K and atmospheric pressure. The derived excess molar volumes, deviations of molar refractions from the mole fraction average, and deviations of isentropic compressibility from the volume freaction average were correlated by the Redlich−Kister polynomial.

Introduction

The ether most commonly added to gasoline is 2-methoxy-2-methylpropane (MTBE) and vapor−liquid equilibrium (VLE) of this compound with ethanol shows an azeotrope with a minimum boiling point (Arce et al., 1996; Gmehling and Bölts, 1996). 1-Butanol could be used as an entrainer in extractive distillation to obtain MTBE with high purity. That is why we have accordingly undertaken a study of the thermodynamics of 1-butanol + ethanol + 2-methoxy-2-methylpropane mixtures. In this work, we have determined the excess molar volumes and the deviations of their molar refractions and isentropic compressibilities from the mole fraction or volume fraction average, respectively. These parameters were estimated from the densities and refractive indices of the system at 298.15 K and atmospheric pressure and from the speed of sound. Previous measurements have been made for 1-butanol + ethanol (Browstow and Lu, 1974; Fukuchi et al., 1983; Pflug and Benson, 1968) and 1-butanol + MTBE (Riggio et al., 1995), but no measurements appear to have been made on the ternary system.

Experimental Section

Materials. Ethanol was supplied by Merck with nominal purity >99.8 mass %; 2-methoxy-2-methylpropane and 1-butanol were supplied by Aldrich and had nominal purities >99.8 and >99.9 mass %, respectively. Water contents of the 1-butanol, ethanol, and MTBE (determined with a Metrohm 737 KF coulometer) were 0.1, 0.08, and 0.03 mass %, respectively.

Apparatus and Procedure. The mixtures were prepared by mass using a Mettler AE 240 balance that measured to within ±0.0001 g. The densities and speeds of sound of the mixtures were measured to within ±0.0001 $g\cdot cm^{-3}$ and ±1 $m\cdot s^{-1}$, respectively, in an Anton Paar DSA-48 densimeter and sound analyzer calibrated with air and water. Refractive indices were measured to within ±0.0001 in an ATAGO RX-1000 refractometer. A Hetotherm thermostat was used to maintain the temperature at (298.15 ± 0.02) K.

Table 1 lists the densities, speeds of sound, and refractive indices measured for the pure components, together with published values for these parameters (Aminabhavi et al., 1993; Daubert and Danner, 1989; Riddick et al., 1986).

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Results

With respect to the binary and ternary systems, the measured densities, speeds of sound, and refractive indices are listed in Tables 2 and 3. Data for ethanol + MTBE mixtures have been already published (Arce et al., 1997). The corresponding excess molar volumes are included in these tables, which were calculated using the expression

$$V^{E} = V - \sum_i x_i V_i \qquad (1)$$

where V is the molar volume of the mixture and V_i and x_i are the molar volume and mole fraction, respectively, of component i; R are the molar refractions, which were calculated using the Lorentz−Lorenz equation

$$R = \frac{n_D^{\,2} - 1}{n_D^{\,2} + 2} V \qquad (2)$$

with n_D and V being the refractive indices and the molar volume, respectively; and ΔR the molar refraction changes of mixing, which were obtained from

$$\Delta R = R - \sum_i x_i R_i \qquad (3)$$

with R_i being the molar refraction of pure component i, x_i being the mole fraction of the ith component, and R being the molar refraction of the mixture.

The speeds of sound through the mixtures (u) and the corresponding densities (ρ) were used to calculate isentropic compressibilities (κ_s) using the equation

$$\kappa_s = u^{-2}\rho^{-1} \qquad (4)$$

and the isentropic compressibility changes of mixing ($\Delta\kappa_s$) were obtained using the expression

$$\Delta\kappa_s = \kappa_s - \sum_i \phi_i \kappa_{si} \qquad (5)$$

where κ_s and κ_{si} are the isentropic compressibilities of the mixture and component i, respectively, and ϕ_i is the volume

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Table 1. Densities ρ and Refractive Indices n_D of the Pure Components and Speeds of Sound u through Them at 298.15 K and Atmospheric Pressure

	ρ/(g·cm^{-3})		n_D		u/(m·s^{-1})	
component	exptl	lit.	exptl	lit.	exptl	lit.
1-butanol	0.8060	0.80575[a]	1.3975	1.39741[a]	1241	1240[b]
ethanol	0.7851	0.78493[a]	1.3592	1.35941[a]	1143	1145[b]
MTBE	0.7356	0.73528[c]	1.3666	1.36630[c]	1037	not found

[a] Riddick et al., 1986. [b] Aminabhavi, 1993. [c] Daubert and Danner, 1989.

Table 2. Densities ρ, Speeds of Sound u, Isentropic Compressibilities κ_s, Refractive Indices n_D, Excess Molar Volumes V^E, and Deviations $\Delta\kappa_s$ and ΔR for Mixtures of 1-Butanol (1) + Ethanol (2) and 1-Butanol (1) + MTBE (2) at 298.15 K and Atmospheric Pressure

x_1	ρ/(g·cm^{-3})	u/(m·s^{-1})	κ_s/TPa^{-1}	n_D	V^E/(cm^3·mol^{-1})	$\Delta\kappa_s$/TPa^{-1}	ΔR/(cm^3·mol^{-1})
			1-Butanol (1) + Ethanol (2)				
0.0584	0.7869	1152	957	1.3626	0.004	0	0.001
0.1205	0.7887	1160	943	1.3659	0.007	1	0.002
0.1641	0.7899	1165	933	1.3681	0.009	0	0.003
0.2231	0.7915	1172	920	1.3710	0.011	0	0.003
0.2413	0.7919	1174	916	1.3719	0.011	0	0.004
0.3053	0.7935	1181	903	1.3747	0.012	−0	0.004
0.3423	0.7944	1185	896	1.3763	0.012	−1	0.005
0.4017	0.7957	1191	885	1.3788	0.011	−1	0.005
0.4377	0.7965	1195	879	1.3802	0.011	−1	0.005
0.4945	0.7976	1201	870	1.3823	0.010	−1	0.005
0.5236	0.7982	1203	865	1.3834	0.009	−2	0.005
0.5605	0.7989	1207	859	1.3847	0.009	−2	0.005
0.6255	0.8002	1213	850	1.3869	0.007	−2	0.005
0.6608	0.8008	1216	845	1.3880	0.006	−2	0.004
0.7084	0.8016	1220	838	1.3895	0.005	−2	0.004
0.7544	0.8024	1224	833	1.3909	0.003	−2	0.004
0.8127	0.8033	1228	825	1.3926	0.002	−1	0.003
0.8710	0.8042	1232	819	1.3942	0.001	−1	0.002
0.9413	0.8052	1237	811	1.3960	0.000	−0	0.001
			1-Butanol (1) + MTBE (2)				
0.0691	0.7407	1052	1220	1.3688	−0.205	−20	−0.011
0.1202	0.7444	1063	1189	1.3705	−0.331	−31	−0.015
0.1603	0.7473	1071	1167	1.3718	−0.416	−39	−0.016
0.2019	0.7503	1079	1144	1.3731	−0.493	−46	−0.017
0.2784	0.7558	1094	1105	1.3756	−0.603	−55	−0.017
0.3170	0.7585	1102	1086	1.3768	−0.645	−58	−0.016
0.3764	0.7627	1113	1057	1.3787	−0.693	−61	−0.015
0.4007	0.7644	1118	1046	1.3795	−0.706	−63	−0.015
0.4544	0.7682	1129	1021	1.3812	−0.725	−64	−0.015
0.5061	0.7719	1139	998	1.3828	−0.728	−64	−0.016
0.5527	0.7752	1149	977	1.3842	−0.719	−64	−0.017
0.5842	0.7774	1156	963	1.3852	−0.706	−63	−0.018
0.6524	0.7822	1170	934	1.3872	−0.660	−59	−0.020
0.6865	0.7846	1177	920	1.3882	−0.628	−57	−0.021
0.7453	0.7887	1190	896	1.3900	−0.557	−51	−0.023
0.8322	0.7948	1208	862	1.3925	−0.414	−38	−0.022
0.8652	0.7970	1215	851	1.3935	−0.348	−32	−0.021
0.9031	0.7996	1222	837	1.3946	−0.263	−24	−0.018
0.9531	0.8029	1232	821	1.3961	−0.136	−12	−0.010

Table 3. Densities ρ, Speeds of Sound u, Isentropic Compressibilities κ_s, Refractive Indices n_D, Excess Molar Volumes V^E, and Deviations $\Delta\kappa_s$ and ΔR for Mixtures of 1-Butanol (1) + Ethanol (2) + MTBE (3) at 298.15 K and Atmospheric Pressure

x_1	x_2	ρ/(g·cm^{-3})	u/(m·s^{-1})	κ_s/TPa^{-1}	n_D	V^E/(cm^3·mol^{-1})	$\Delta\kappa_s$/TPa^{-1}	ΔR/(cm^3·mol^{-1})
0.1010	0.8990	0.7882	1157	948	1.3650	0.007	1	0.002
0.0906	0.8067	0.7811	1143	980	1.3662	−0.202	−25	−0.016
0.0786	0.6998	0.7735	1127	1017	1.3672	−0.383	−43	−0.026
0.0697	0.6208	0.7684	1115	1046	1.3676	−0.477	−48	−0.029
0.0621	0.5527	0.7642	1105	1071	1.3679	−0.529	−50	−0.029
0.0495	0.4407	0.7577	1090	1110	1.3680	−0.559	−49	−0.026
0.0466	0.4152	0.7563	1087	1119	1.3680	−0.556	−48	−0.025
0.0292	0.2603	0.7481	1069	1170	1.3677	−0.461	−39	−0.017
0.0204	0.1814	0.7442	1059	1197	1.3675	−0.361	−31	−0.012
0.0120	0.1065	0.7406	1050	1224	1.3671	−0.234	−20	−0.007
0.2094	0.7906	0.7913	1171	922	1.3704	0.010	0	0.003
0.1849	0.6983	0.7832	1153	960	1.3707	−0.244	−28	−0.013
0.1637	0.6179	0.7766	1138	994	1.3707	−0.412	−44	−0.024
0.1442	0.5446	0.7710	1124	1026	1.3706	−0.520	−51	−0.030
0.1269	0.4792	0.7662	1112	1055	1.3704	−0.581	−53	−0.034
0.1066	0.4023	0.7608	1099	1088	1.3700	−0.609	−53	−0.035
0.0854	0.3223	0.7554	1087	1121	1.3695	−0.589	−50	−0.033
0.0649	0.2449	0.7504	1075	1153	1.3689	−0.522	−44	−0.029
0.0500	0.1887	0.7469	1066	1177	1.3684	−0.444	−37	−0.024
0.0279	0.1054	0.7418	1054	1214	1.3677	−0.284	−24	−0.015

Table 3 (Continued)

x_1	x_2	ρ/(g·cm^{-3})	u/(m·s^{-1})	κ_s/TPa^{-1}	n_D	V^E/(cm^3·mol^{-1})	$\Delta\kappa_s$/TPa^{-1}	ΔR/(cm^3·mol^{-1})
0.2854	0.7146	0.7932	1180	906	1.3738	0.012	−0	0.004
0.2539	0.6357	0.7855	1162	943	1.3736	−0.236	−27	−0.009
0.2278	0.5703	0.7795	1148	973	1.3732	−0.394	−43	−0.017
0.1965	0.4921	0.7727	1131	1012	1.3727	−0.529	−53	−0.024
0.1730	0.4332	0.7678	1118	1042	1.3721	−0.591	−55	−0.028
0.1424	0.3566	0.7616	1102	1081	1.3714	−0.620	−55	−0.030
0.1200	0.3005	0.7572	1092	1108	1.3707	−0.606	−52	−0.029
0.0913	0.2285	0.7518	1079	1143	1.3698	−0.541	−46	−0.026
0.0696	0.1742	0.7478	1069	1170	1.3691	−0.459	−39	−0.022
0.0441	0.1103	0.7432	1058	1203	1.3682	−0.325	−28	−0.016
0.3957	0.6043	0.7956	1191	886	1.3785	0.011	−1	0.005
0.3473	0.5304	0.7863	1172	924	1.3773	−0.257	−32	−0.008
0.3148	0.4807	0.7817	1157	955	1.3766	−0.412	−37	−0.015
0.2750	0.4199	0.7752	1140	992	1.3756	−0.549	−45	−0.021
0.2385	0.3642	0.7695	1125	1027	1.3746	−0.618	−49	−0.024
0.2042	0.3119	0.7642	1112	1060	1.3735	−0.635	−49	−0.026
0.1628	0.2486	0.7580	1096	1101	1.3722	−0.608	−47	−0.026
0.1288	0.1967	0.7531	1084	1135	1.3710	−0.553	−42	−0.023
0.0956	0.1459	0.7485	1072	1168	1.3700	−0.472	−36	−0.020
0.0452	0.0691	0.7418	1054	1219	1.3683	−0.287	−22	−0.011
0.4999	0.5001	0.7979	1202	868	1.3825	0.010	−1	0.005
0.4436	0.4438	0.7901	1183	905	1.3810	−0.260	−31	−0.005
0.4019	0.4021	0.7844	1168	935	1.3798	−0.407	−45	−0.011
0.3455	0.3457	0.7769	1148	977	1.3781	−0.547	−57	−0.018
0.3063	0.3064	0.7718	1134	1008	1.3768	−0.609	−60	−0.020
0.2502	0.2503	0.7648	1114	1054	1.3751	−0.649	−59	−0.022
0.2094	0.2094	0.7599	1100	1087	1.3738	−0.642	−55	−0.022
0.1655	0.1656	0.7547	1087	1122	1.3724	−0.600	−49	−0.020
0.1340	0.1340	0.7511	1077	1148	1.3713	−0.543	−43	−0.018
0.0666	0.0666	0.7433	1057	1203	1.3690	−0.340	−27	−0.011
0.5425	0.4575	0.7987	1205	862	1.3839	0.009	−2	0.005
0.4762	0.4016	0.7904	1182	905	1.3824	−0.279	−30	0.007
0.4261	0.3594	0.7842	1166	937	1.3810	−0.440	−46	0.006
0.3692	0.3114	0.7773	1148	976	1.3793	−0.569	−57	0.001
0.3272	0.2760	0.7723	1135	1005	1.3779	−0.628	−61	−0.002
0.2740	0.2311	0.7661	1118	1045	1.3762	−0.660	−62	−0.008
0.2271	0.1915	0.7607	1103	1080	1.3746	−0.649	−58	−0.012
0.1664	0.1403	0.7539	1085	1126	1.3724	−0.576	−50	−0.015
0.1291	0.1089	0.7498	1075	1155	1.3711	−0.498	−43	−0.015
0.0784	0.0661	0.7442	1060	1195	1.3694	−0.346	−30	−0.012
0.6780	0.3220	0.8012	1217	842	1.3885	0.006	−2	0.004
0.5919	0.2811	0.7925	1193	887	1.3859	−0.303	−31	−0.007
0.5368	0.2549	0.7869	1177	917	1.3842	−0.445	−45	−0.013
0.4677	0.2221	0.7800	1159	955	1.3820	−0.571	−57	−0.018
0.4081	0.1938	0.7741	1142	990	1.3801	−0.640	−61	−0.021
0.3452	0.1639	0.7680	1125	1029	1.3780	−0.673	−62	−0.023
0.2770	0.1316	0.7615	1107	1072	1.3759	−0.664	−58	−0.022
0.2141	0.1017	0.7556	1091	1112	1.3738	−0.610	−52	−0.020
0.1661	0.0789	0.7512	1079	1143	1.3723	−0.536	−46	−0.018
0.0792	0.0376	0.7431	1058	1203	1.3694	−0.318	−28	−0.010
0.7924	0.2076	0.8030	1226	829	1.3919	0.003	−1	0.003
0.6938	0.1818	0.7944	1201	873	1.3887	−0.295	−29	−0.014
0.6262	0.1641	0.7887	1184	904	1.3868	−0.473	−45	−0.020
0.5663	0.1484	0.7836	1170	932	1.3849	−0.586	−55	−0.022
0.4745	0.1243	0.7757	1148	978	1.3820	−0.684	−63	−0.025
0.4029	0.1055	0.7696	1131	1017	1.3797	−0.714	−64	−0.027
0.3382	0.0886	0.7641	1115	1053	1.3777	−0.713	−62	−0.029
0.2658	0.0696	0.7581	1098	1094	1.3753	−0.673	−56	−0.030
0.1890	0.0495	0.7517	1081	1139	1.3728	−0.567	−47	−0.030
0.1135	0.0297	0.7452	1064	1186	1.3702	−0.381	−33	−0.025
0.8760	0.1240	0.8043	1232	819	1.3941	0.001	−1	0.002
0.7649	0.1083	0.7956	1205	865	1.3912	−0.298	−29	0.011
0.6804	0.0963	0.7891	1187	900	1.3888	−0.492	−46	0.013
0.6068	0.0859	0.7834	1170	932	1.3866	−0.607	−56	0.011
0.5151	0.0729	0.7761	1150	974	1.3837	−0.686	−63	0.006
0.4369	0.0618	0.7700	1132	1013	1.3811	−0.712	−64	0.001
0.3709	0.0525	0.7649	1117	1047	1.3790	−0.705	−62	−0.003
0.2914	0.0412	0.7587	1100	1089	1.3764	−0.659	−57	−0.008
0.2095	0.0297	0.7523	1083	1134	1.3736	−0.552	−47	−0.010
0.1233	0.0175	0.7454	1064	1184	1.3707	−0.362	−33	−0.010

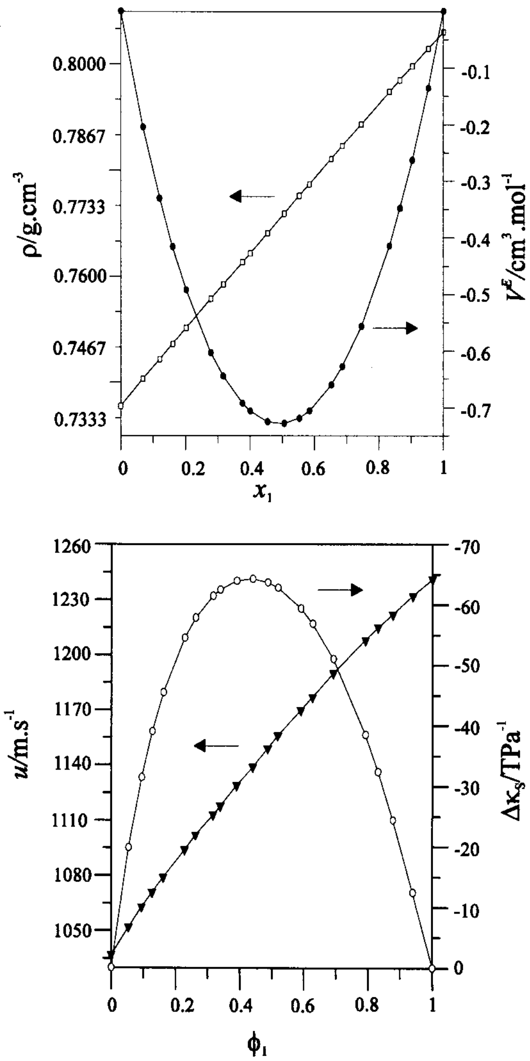

Figure 1. Physical properties and changes of mixing for the 1-butanol (1) + MTBE (2) system at 298.15 K and atmospheric pressure.

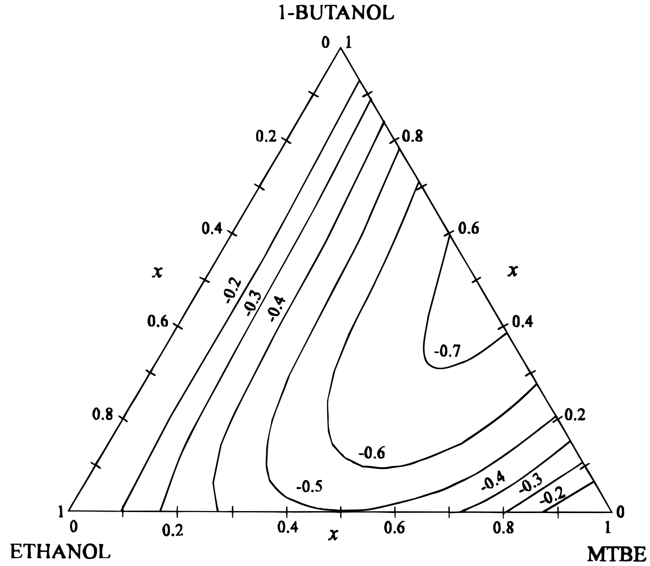

Figure 2. Excess molar volume isolines for 1-butanol (1) + ethanol (2) + MTBE (3) at 298.15 K and atmospheric pressure (system compositions in mole fraction).

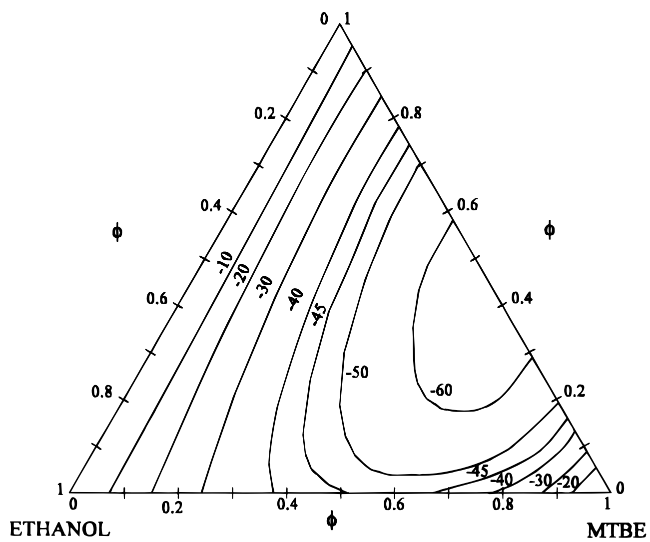

Figure 3. Isentropic compressibility changes of mixing isolines for 1-butanol (1) + ethanol (2) + MTBE (3) mixtures at 298.15 K and atmospheric pressure (system compositions in volume fractions).

fraction of component i in the mixture as given by

$$\phi_i = x_i V_i / \sum_j x_j V_j \quad (6)$$

where j refers to all components of the mixture.

With respect to the binary system 1-butanol + MTBE, the measured values of ρ, u, V^E, and $\Delta\kappa_s$ are plotted against the composition of 1-butanol in Figure 1. Excess molar volumes obtained for this system are lower (minimum at -0.73 $cm^3 \cdot mol^{-1}$) than previously published by Riggio et al. (1995) (minimum at -0.86 $cm^3 \cdot mol^{-1}$). Unfortunately we could not find in the literature other comparative data. The behavior of the V^E and ΔR vs composition for this binary mixture reveals that a symmetric regular solution model could be applied.

For the ternary system, Figure 2 shows excess volume isolines (system compositions in mole fractions, x_i), and Figure 3, isolines for the isentropic compressibility changes of mixing (system compositions in volume fraction, ϕ_i).

Correlation

The V^E, ΔR, and $\Delta\kappa_s$ data were correlated with the composition data by means of the Redlich−Kister polynomial (1948), which for binary mixtures is

$$\Delta M = x_i x_j \sum_n A_n (x_i - x_j)^n \quad (7)$$

where ΔM is V^E or ΔR and x_i is the mole fraction of component i in the mixture or ΔM is $\Delta\kappa_s$ and x_i is the volume fraction of component i in the mixture, A_n is the polynomial coefficient, and n is the number of the polynomial coefficient. For ternary systems the corresponding equation is

$$\Delta M_{123} = \Delta M_{12} + \Delta M_{23} + \Delta M_{13} + x_1 x_2 x_3 (A + B(x_1 - x_2) + C(x_2 - x_3) + D(x_1 - x_3) + E(x_1 - x_2)^2 + F(x_2 - x_3)^2 + G(x_1 - x_3)^2 + ...) \quad (8)$$

where ΔM_{123} is V^E, $\Delta\kappa_s$, or ΔR, x_i is the mole fraction or volume fraction of component i, according to the parameter being correlated (as previously indicated), and ΔM_{ij} is the

Table 4. Polynomial Coefficients (A_n) and Standard Deviations (σ) Obtained for the Fits of Equation 7 to the V^E−, $\Delta\kappa_s$−, ΔR−Composition Data for the Binary Systems (for $\Delta\kappa_s$, System Compositions Were in Volume Fraction, ϕ_i)

property	A_0	A_1	A_2	A_3	σ
		1-Butanol (1) + Ethanol (2)			
V^E/(cm^3·mol^{-1})	0.0396	−0.0404			0.001
$\Delta\kappa_s$/TPa^{-1}	−3.52	−10.87			0.1
ΔR/(cm^3·mol^{-1})	0.0199	−0.0273			0.001
		Ethanol (1) + MTBE (2)			
V^E/(cm^3·mol^{-1})	−1.9854	0.0379		−0.5576	0.001
$\Delta\kappa_s$/TPa^{-1}	−179.4	46.5	−56.4	53.9	0.2
ΔR/(cm^3·mol^{-1})	−0.0864	−0.0377			0.001
		1-Butanol (1) + MTBE (2)			
V^E/(cm^3·mol^{-1})	−2.9144	0.0886	−0.2546		0.001
$\Delta\kappa_s$/TPa^{-1}	−253.5	53.1	−63.2	52.6	0.1
ΔR/(cm^3·mol^{-1})	−0.0627	−0.0314	−0.1725		0.001

Table 5. Polynomial Coefficients and Standard Deviations (σ) Obtained for the Fits of Equation 8 to the V^E−, $\Delta\kappa_s$−, and ΔR−Composition Data for the Ternary System 1-Butanol (1) + Ethanol (2) + MTBE (3) (for $\Delta\kappa_s$, System Compositions Were in Volume Fraction, ϕ_i)

property	A	B	C	D	σ
V^E/(cm^3·mol^{-1})	−1.3233	1.1970	0.5744	1.7714	0.013
$\Delta\kappa_s$/TPa^{-1}	62.77	21.41	−97.73	−76.32	1.36
ΔR/(cm^3·mol^{-1})	−0.0599	0.6412	−0.067	0.5741	0.005

value of the Redlich−Kister coefficient for the same property, as obtained by fitting the Redlich−Kister polynomial to the data for the binary system (i, j).

Equations 7 and 8 were fitted to the appropriate parameter−composition data for the binary and ternary systems by least-squares regression, applying Fisher's F-test to establish the number of coefficients. These coefficients and their mean standard deviations are listed in Table 4 for the binary systems (data for the ethanol + MTBE system were taken from Arce et al., 1997) and in Table 5 for the ternary system. The mean standard deviation was calculated by the usual equation

$$\sigma = \left[\frac{1}{n-1}(\Delta M_{\text{expt}} - \Delta M_{\text{calc}})^2\right]^{1/2}$$

where n is the number of points.

Conclusions

Molar volumes, molar refractions, and isentropic compressibilities were evaluated at 298.15 K and atmospheric pressure for ternary mixtures of 1-butanol + ethanol + MTBE from measurements of their density and refractive index and the speed of sound through them. The effects of mixing on these properties were determined.

Excess molar volumes were negative and relatively large except for binary 1-butanol + ethanol mixtures, which had small positive V^E. The minimum (around −0.73 cm^3·mol^{-1}) corresponds to mixtures with 1-butanol and MTBE of around 0.5 mole fraction. Molar refraction changes of mixing were negative, becoming positive in the nearness to 1-butanol + ethanol mixtures. Isentropic compressibility changes of mixing were negative, reaching −64 TPa^{-1} for some mixtures with 1-butanol and MTBE.

In all cases the data were satisfactorily correlated with the composition data by the Redlich−Kister polynomial.

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