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Thermodynamic Properties of Dodecane + 1-Butanol and + 2-Butanol Systems

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Densities and speeds of sound in the temperature interval (278.15 to 318.15) K and heat capacities per unit volume from (283.15 to 318.15) K were determined for the dodecane + 1-butanol and + 2-butanol systems. Measurements were made over the whole composition range and at atmospheric pressure. From these data, excess molar volumes, excess isentropic compressibilities, and excess isobaric molar heat capacities were calculated. Results were critically compared with literature data of systems containing these butanol isomers. Differences between both systems were ascribed to the different association ability of the two alcohols.

Introduction

Thermophysical properties of systems containing alkanols have been extensively analyzed from both theoretical and experimental points of view.[1−9] Several papers dealt with the study of the thermodynamic behavior of systems each containing a different alkanol isomer and the same solvent. It was found that the value of such properties varies with the position of the hydroxyl group in the alkyl chain of the alkanol. The explanation of these results was related to a decrease of the association capability when the hydroxyl group is located in a nonprimary position. The steric hindrance over the hydroxyl group was found to be responsible for this fact rather than the debilitation of the H-bond energy.[7]

In this work, a comparative study of the thermodynamic behavior of dodecane + 1-butanol and + 2-butanol systems is presented. To this end, densities and speeds of sound in the temperature interval (278.15 to 318.15) K and isobaric heat capacities per unit volume at (283.15 to 318.15) K were measured at atmospheric pressure and over the whole composition range. From these data, excess volumes, excess isentropic compressibilities, and excess isobaric molar heat capacities were calculated. The difference between both systems was discussed in terms of the above-commented considerations.

Experimental Section

Materials. 1-Butanol, heptane, and octane (calibration liquids) were purchased from Fluka, whereas 2-butanol and dodecane were purchased from from Aldrich. In all cases, the purity was checked by gas chromatography analysis (GC) obtaining 1-butanol (99.8 mol %), heptane (99.60 mol %), octane (99.50 mol %), 2-butanol (99.45 mol %), and dodecane (99.20 mol %). Liquids were degassed and passed through 0.4-nm molecular sieves prior to use. The mole fractions of the mixtures were obtained by weighing on a Mettler Balance AE-240 with a sensitivity of ±0.00001 g.

Apparatus and Procedure. The densimeter and sound analyzer DSA-48 from Anton Paar was used to measure the density ρ and speed of sound u. MilliQ water and octane were used as calibration fluids, taking their values from the literature.[10−13] Data were obtained with steps between consecutive measurements of 5 K using an automated method described elsewhere.[14] The uncertainty in ρ and u was estimated to be $\pm 0.00010\ \mathrm{g{\cdot}cm^{-3}}$ and $\pm 0.1\ \mathrm{m{\cdot}s^{-1}}$, respectively.[15]

Isobaric heat capacities per unit volume C_pV^{-1} were measured with a Setaram micro DSC II differential scanning calorimeter using the scanning method with a rate of $0.25\ \mathrm{K{\cdot}min^{-1}}$. Isobaric molar heat capacities $C_{p,\mathrm{m}}$ were obtained from these data, density values, and molar mass. 1-Butanol and heptane were used as calibration liquids; their $C_{p,\mathrm{m}}$ values were taken from the literature.[16] The uncertainty in $C_{p,\mathrm{m}}$ was estimated to be $\pm 0.15\ \mathrm{J{\cdot}K^{-1}{\cdot}mol^{-1}}$.[15] A more detailed explanation of the measurement method is available elsewhere.[17]

Results and Discussion

The properties for pure liquids and values from the literature are presented in Table 1. Good agreement between the two sets of data can be observed. The data for the mixtures are listed in Tables 2−4. The values obtained for each property were fit to the polynomials

$$Y = \sum_{i=0}^{n} A_i x^i \tag{1}$$

where Y denotes $\rho/(\mathrm{g{\cdot}cm^{-3}})$, $u/(\mathrm{m{\cdot}s^{-1}})$, or $C_{p,\mathrm{m}}/(\mathrm{J{\cdot}mol^{-1}{\cdot}K^{-1}})$ and x is the mole fraction of the alkanol. The dependence of A_i coefficients against temperature is given by the next polynomial

$$A_i = \sum_{j=0}^{2} A_{ij} 10^{-j} (T/K - T_0/K)^j \tag{2}$$

where T_0 = 273.15 K. The A_{ij} coefficients were obtained from the simultaneous fit of the experimental data against T and x. Their values are listed in Table 5. The optimum number of parameters was determined using a previously reported methodology.[20]

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Table 1. Properties of Pure Liquids at Temperature *T*

	ρ/(g·cm^{-3})		$C_{p,\mathrm{m}}$/(J·mol^{-1}·K^{-1})		u/(m·s^{-1})	
T/K	this work	literature[11]	this work	literature[16]	this work	literature
			n-Dodecane			
278.15	0.75955	0.75980			1357.14	
288.15	0.75236	0.75255	371.38	371.21	1317.27	
298.15	0.74517	0.74528	375.94	375.94	1278.20	1278[18]
308.15	0.73795	0.73799	380.92	381.13	1239.82	
318.15	0.73069	0.73066	386.31	386.54	1201.88	
			1-Butanol			
278.15	0.82111	0.82078		164.73	1308.19	
288.15	0.81349	0.81328		170.60	1273.51	
298.15	0.80586	0.80569		177.12	1239.29	
308.15	0.79804	0.79798		180.64	1205.50	1203[19]
318.15	0.79007	0.79011		192.27	1172.12	
			2-Butanol			
278.15	0.81803				1282.95	
288.15	0.81025		186.96	186.39	1247.03	
298.15	0.80219	0.80255	197.23	196.81	1210.98	1212[18]
308.15	0.79375	0.79392	207.51	207.57	1174.53	
318.15	0.78486	0.78492	217.75	218.22	1137.61	

Table 2. Densities of Mixtures at Several Temperatures

	ρ/(g·cm^{-3})						
x	278.15 K	283.15 K	288.15 K	298.15 K	308.15 K	313.15 K	318.15 K
			x(1-Butanol) + (1 − x)(Dodecane)				
0.02749	0.76000	0.75637	0.75275	0.74549	0.73818	0.73451	0.73080
0.08048	0.76112	0.75747	0.75382	0.74651	0.73913	0.73541	0.73165
0.19017	0.76386	0.76019	0.75650	0.74912	0.74167	0.73790	0.73412
0.28812	0.76684	0.76316	0.75947	0.75204	0.74451	0.74071	0.73686
0.38654	0.77041	0.76672	0.76298	0.75551	0.74793	0.74409	0.74020
0.44097	0.77269	0.76897	0.76523	0.75773	0.75012	0.74627	0.74236
0.48350	0.77461	0.77089	0.76714	0.75962	0.75200	0.74813	0.74423
0.58849	0.78011	0.77638	0.77265	0.76509	0.75740	0.75351	0.74954
0.61515	0.78174	0.77804	0.77427	0.76672	0.75903	0.75513	0.75118
0.69605	0.78713	0.78343	0.77963	0.77204	0.76436	0.76044	0.75646
0.79594	0.79527	0.79153	0.78776	0.78020	0.77250	0.76857	0.76459
0.90295	0.80660	0.80288	0.79912	0.79157	0.78388	0.77997	0.77598
0.95231	0.81320	0.80945	0.80570	0.79815	0.79048	0.78657	0.78260
			x(2-Butanol) + (1 − x)(Dodecane)				
0.04602	0.76012	0.75643	0.75279	0.74546	0.73806	0.73435	0.73061
0.05070	0.76017	0.75650	0.75285	0.74551	0.73811	0.73439	0.73064
0.09287	0.76095	0.75729	0.75361	0.74619	0.73872	0.73495	0.73115
0.18128	0.76289	0.75918	0.75547	0.74797	0.74036	0.73652	0.73263
0.24985	0.76465	0.76089	0.75715	0.74959	0.74190	0.73802	0.73407
0.28364	0.76559	0.76185	0.75809	0.75050	0.74277	0.73886	0.73490
0.30044	0.76610	0.76233	0.75857	0.75094	0.74322	0.73930	0.73532
0.33974	0.76736	0.76357	0.75979	0.75215	0.74436	0.74042	0.73640
0.38271	0.76880	0.76502	0.76121	0.75351	0.74569	0.74171	0.73768
0.48939	0.77314	0.76930	0.76543	0.75762	0.74962	0.74557	0.74145
0.59429	0.77850	0.77461	0.77068	0.76272	0.75456	0.75042	0.74623
0.69831	0.78513	0.78124	0.77727	0.76920	0.76088	0.75665	0.75233
0.80469	0.79385	0.78992	0.78596	0.77780	0.76939	0.76506	0.76064
0.90245	0.80421	0.80031	0.79636	0.78821	0.77973	0.77536	0.77088
0.95177	0.81069	0.80680	0.80284	0.79474	0.78627	0.78189	0.77738

Molar volumes V_m were derived from density and molar mass data. Isentropic compressibilities κ_s were calculated from density and speed of sound values using the Laplace equation $\kappa_\mathrm{s} = 1/(\rho u^2)$. Excess properties were calculated by using $Z^\mathrm{E} = Z - Z^\mathrm{id}$ where Z^E is the excess quantity of the property Z (in our case, V_m, $C_{p,\mathrm{m}}$, or κ_s) and Z^id is its ideal value. Z^id values were calculated using the criterion of Benson and Kiyohara,[21] which is defined as follows:

$$V_\mathrm{m}^\mathrm{id} = xV_{\mathrm{m},1}^{*} + (1 - x)V_{\mathrm{m},2}^{*} \tag{3}$$

$$C_{p,\mathrm{m}}^\mathrm{id} = xC_{p,\mathrm{m},1}^{*}{*}+ (1 - x)C_{p,\mathrm{m},2}^{*} \tag{4}$$

$$\kappa_\mathrm{s}^\mathrm{id} = \kappa_T^\mathrm{id} - \frac{TV_\mathrm{m}^\mathrm{id}(\alpha_p^\mathrm{id})^2}{C_{p,\mathrm{m}}^\mathrm{id}} \tag{5}$$

Superscript * denotes properties of the pure liquids. To obtain $\kappa_\mathrm{s}{}^\mathrm{id}$, it was necessary to calculate the ideal magnitude of the isobaric thermal expansivity α_p and isothermal compressibility κ_T from the next set of equations:

$$\alpha_p^\mathrm{id} = \phi\alpha_{p,1}^{*} + (1 - \phi)\alpha_{p,2}^{*} \tag{6}$$

$$\kappa_T^\mathrm{id} = \phi\kappa_{T,1}^{*} + (1 - \phi)\kappa_{T,2}^{*} \tag{7}$$

$$\phi = \frac{xV_{\mathrm{m},1}^{*}}{V_\mathrm{m}^\mathrm{id}} \tag{8}$$

where ϕ is the volume fraction of component 1. $\alpha_{p,i}^{*}$ were obtained from density data using a numerical procedure

Table 3. Speeds of Sound of Mixtures at Several Temperatures

	u/(m·s^{-1})						
x	278.15 K	283.15 K	288.15 K	298.15 K	308.15 K	313.15 K	318.15 K
x(1-Butanol) + (1 − x)(Dodecane)							
0.02749	1354.43	1334.52	1314.74	1275.72	1237.32	1218.43	1199.74
0.08048	1351.65	1331.73	1311.88	1272.60	1234.00	1214.93	1196.12
0.19017	1346.36	1326.51	1306.67	1267.38	1228.68	1209.61	1190.76
0.28812	1341.97	1322.13	1302.34	1263.17	1224.60	1205.57	1186.70
0.38654	1337.56	1317.80	1298.07	1259.03	1220.59	1201.58	1182.73
0.44097	1335.22	1315.38	1295.76	1256.85	1218.52	1199.59	1180.76
0.48350	1333.14	1313.58	1293.98	1255.23	1217.02	1198.11	1179.44
0.58849	1328.39	1309.07	1289.78	1251.40	1213.58	1194.84	1176.25
0.61515	1327.00	1307.93	1288.69	1250.47	1212.77	1194.12	1175.56
0.69605	1323.29	1304.43	1285.21	1247.52	1210.28	1191.80	1173.49
0.79594	1318.02	1299.42	1280.87	1244.06	1207.63	1189.59	1171.64
0.90295	1312.42	1294.40	1276.41	1240.79	1205.58	1188.12	1170.77
0.95231	1310.21	1292.36	1274.76	1239.77	1205.26	1188.14	1171.10
x(2-Butanol) + (1 − x)(Dodecane)							
0.04602	1352.71	1332.55	1312.74	1273.46	1234.92	1215.90	1197.08
0.05070	1352.23	1332.16	1312.32	1273.10	1234.48	1215.47	1196.62
0.09287	1349.11	1329.10	1309.26	1269.82	1231.07	1211.97	1193.03
0.18128	1342.70	1322.72	1302.82	1263.38	1224.51	1205.33	1186.25
0.24985	1337.80	1317.72	1297.76	1258.36	1219.48	1200.26	1181.20
0.30044	1333.96	1313.93	1294.05	1254.67	1215.85	1196.60	1177.53
0.33974	1331.15	1311.08	1291.23	1251.84	1213.01	1193.81	1174.71
0.38271	1327.93	1307.94	1288.14	1248.76	1209.91	1190.64	1171.59
0.48939	1320.58	1300.59	1280.62	1241.13	1202.20	1182.95	1163.80
0.59429	1313.68	1293.66	1273.68	1234.11	1195.02	1175.58	1156.43
0.69831	1306.65	1286.88	1267.06	1227.66	1188.52	1169.12	1149.84
0.80469	1298.97	1279.62	1260.19	1221.36	1182.58	1163.24	1143.97
0.90245	1291.01	1272.34	1253.50	1215.73	1177.79	1158.73	1139.65
0.95177	1286.96	1268.68	1250.22	1213.24	1175.92	1157.14	1138.24

Table 4. Isobaric Molar Heat Capacities of Mixtures at Several Temperatures

	$C_{p,\mathrm{m}}$/(J·mol^{-1}K^{-1})					
x	283.15 K	288.15 K	298.15 K	308.15 K	313.15 K	318.15 K
x(1-Butanol) + (1 − x)(Dodecane)						
0.08048	361.08	364.22	370.79	376.59	379.38	382.63
0.19017	340.76	344.32	351.89	359.80	363.77	367.77
0.28812	321.63	325.39	333.33	341.79	346.13	350.48
0.38654	301.93	305.76	313.86	322.51	327.00	331.46
0.44097	290.92	294.85	303.08	311.79	316.36	320.93
0.48350	282.09	286.04	294.28	303.09	307.67	312.27
0.54061	269.83	273.71	281.93	290.73	295.36	300.02
0.58849	259.76	263.80	272.13	281.00	285.53	290.20
0.61515	253.75	257.68	265.84	274.56	279.13	283.62
0.69605	236.27	240.13	248.35	257.05	261.55	266.23
0.79594	213.74	217.56	225.46	233.92	238.39	242.97
0.90295	189.46	192.97	200.34	208.32	212.48	216.75
0.95231	178.05	181.36	188.36	195.95	200.02	204.16
x(2-Butanol) + (1 − x)(Dodecane)						
0.04602	368.81	371.20	375.87	380.02	381.77	383.08
0.05070	368.27	370.87	375.91	380.01	381.76	383.28
0.09287	362.22	365.51	371.99	377.88	380.52	382.79
0.18128	347.53	351.44	359.44	367.12	370.85	374.26
0.24985	335.43	339.54	348.04	356.53	360.66	364.70
0.30044	326.19	330.40	339.21	348.12	352.50	356.68
0.33974	319.07	323.35	332.29	341.43	345.94	350.35
0.38271	310.89	315.22	324.42	333.69	338.16	342.58
0.48939	291.24	295.51	304.59	313.86	318.50	323.17
0.59429	271.63	276.16	285.33	294.46	299.14	303.75
0.69831	250.80	255.61	265.16	274.50	279.14	284.00
0.80469	228.10	233.26	243.52	253.41	258.32	263.01
0.90245	205.58	210.78	221.25	231.68	236.78	241.63
0.95177	193.81	198.92	209.36	219.79	225.00	230.03

described previously,[15] and $\kappa_{T,i}$* were calculated through the next equation:

$$\kappa^{*}_{T,i} = \kappa^{*}_{\mathrm{s},i} + \frac{TV^{*}_{\mathrm{m},i}\alpha^{*}_{p,i}}{C^{*}_{p,\mathrm{m},i}} \qquad (9)$$

Figure 1 shows V_m^E for the studied systems. They are positive over the whole composition range and increase with temperature as is commonly observed in short alkyl chain alkanol + long alkyl chain alkane systems.[22–24] The V_m^E values are higher for the 2-butanol system, according to previous work devoted to the study of mixtures that

Table 5. Smoothing Coefficients A_{ij} and Standard Deviations s of the Fit to Equations 1 and 2

	A_{ij}							
	i							
j	0	1	2	3	4	5	6	7
				x(1-Butanol) + (1 − x)(Dodecane)				
$\rho/(g\cdot cm^{-3})$				$s = 0.00003$				
0	0.76313	0.01499	0.06977	−0.22076	0.49445	−0.49133	0.19452	
1	−0.00715	−0.00101	0.00248	0.00010	−0.01608	0.02493	−0.01157	
2	−0.00002	−0.00022	0.00090	−0.00235	0.00286	−0.00147	0.00023	
$u/(m\cdot s^{-1})$	$s = 0.10$							
0	1377.00	−81.21	251.71	−830.74	1402.39	−1183.38	390.01	
1	−40.40	−1.96	16.15	−53.80	112.89	−103.46	35.46	
2	0.33	0.10	−3.35	16.18	−34.51	32.77	−11.31	
$C_{p,m}/(J\cdot mol^{-1}\cdot K^{-1})$				$s = 0.16$				
0	365.32	−80.63	−968.01	4239.23	−10334.69	13832.46	−9527.11	2635.40
1	3.74	71.25	−547.64	1976.35	−3689.53	3633.04	−1766.14	324.28
2	0.20	−11.00	120.38	−506.79	1067.54	−1196.65	682.29	−155.68
				x(2-Butanol) + (1 − x)(Dodecane)				
$\rho/(g\cdot cm^{-3})$				$s = 0.00003$				
0	0.76310	0.01153	0.05785	−0.14175	0.29994	−0.27629	0.10735	
1	−0.00715	−0.00258	0.01807	−0.06422	0.10349	−0.07689	0.02193	
2	−0.00001	−0.00023	−0.00034	0.00374	−0.00843	0.00730	−0.00221	
$u/(m\cdot s^{-1})$				$s = 0.09$				
0	1377.10	−96.99	202.08	−799.44	1570.63	−1436.24	483.41	
1	−40.42	−5.72	47.67	−137.33	152.22	−45.59	−6.18	
2	0.33	0.41	−7.52	29.81	−48.29	32.54	−7.48	
$C_{p,m}/(J\cdot mol^{-1}\cdot K^{-1})$				$s = 0.3$				
0	365.6	−7.4	−1565.4	6619.7	−14870.2	18255.9	−11642.3	3016.3
1	3.7	70.1	−442.1	1641.9	−3765.7	5072.9	−3578.3	1007.4
2	0.2	−17.7	191.1	−871.3	2081.3	−2716.7	1826.0	−492.9

contains these butanol isomers.[14,25,26] To our knowledge, only Gonzalez et al.[18] report data for 2-butanol + dodecane. They measured V_m^E for 2-butanol + alkane systems obtaining at equimolecular composition an unusual tendency of V_m^E against carbon number n of the hydrocarbon, with a maximum located at $n = 10$. Our data do not confirm this behavior because the values obtained for the 2-butanol system are 10% higher than those of González et al. and the maximum would not occur.

Excess isentropic compressibilities κ_s^E are shown in Figure 2, presenting behavior similar to that of the excess molar volumes. The observed temperature and composition dependencies of κ_s^E are also usually found in similar systems.[22−24,27] The differences between the two systems for this quantity and also for V_m^E can be easily explained by the different capability of self-association of the two alkanols. The weaker association of 2-butanol due to its higher steric hindrance produces an enhancement of the destruction of the alcohol structure by the alkane molecules, which implies higher V_m^E and κ_s^E.

Figure 3 shows excess isobaric molar heat capacities $C_{p,m}^E$ for both systems. Results follow the behavior usually

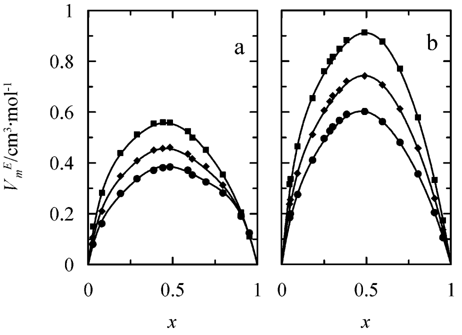

Figure 1. Excess molar volumes V_m^E for (a) x(1-butanol) + (1 − x)(dodecane) and (b) x(2-butanol) + (1 − x)(dodecane) at ●, T = 283.15 K; ◆, T = 298.15 K; and ■, T = 313.15 K. (−) Calculated values from density fits (eqs 1 and 2).

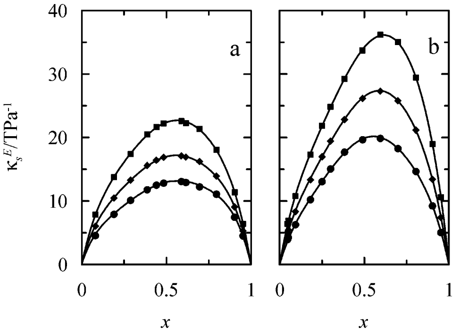

Figure 2. Excess isentropic compressibilities κ_s^E for (a) x(1-butanol) + (1 − x)(dodecane) and (b) x(2-butanol) + (1 − x)-(dodecane) at ●, T = 283.15 K; ◆, T = 298.15 K; and ■, T = 313.15 K. (−) Calculated values from density and speed of sound fits (eqs 1 and 2).

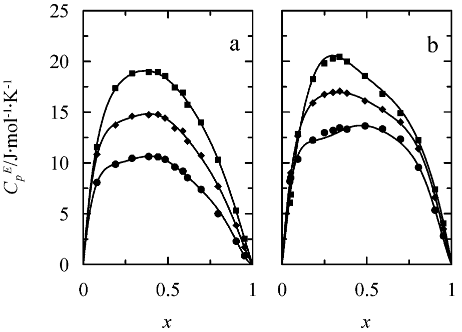

Figure 3. Excess isobaric molar heat capacities $C_{p,m}^E$ for (a) x(1-butanol) + (1 − x)(dodecane) and (b) x(2-butanol) + (1 − x)-(dodecane) at ●, T = 283.15 K; ◆, T = 298.15 K; and ■, T = 313.15 K. (−) Calculated values from isobaric heat capacity fits (eqs 1 and 2).

found for these kinds of systems: highly positive values increasing strongly with temperature.[22–24] Between the two systems, there are not strong differences, which also was found in other comparative studies of systems containing these butanol isomers.[25] Taking into account that $C_{p,\mathrm{m}}^{\mathrm{E}}$ is mainly affected by the variation of the order with temperature of the real mixture with reference to that of the ideal one,[6] these results show that it is similar between both systems.

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