the two components of the mixture (*25*, *26*).

Registry No. AN, 75-05-8; 1-PrOH, 71-23-8; 2-PrOH, 67-63-0.

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Mixture Properties of the Water + γ-Butyrolactone + Tetrahydrofuran System. 1. Densities of γ-Butyrolactone + Water at 303.15–343.15 K and of Tetrahydrofuran + γ-Butyrolactone at 278.15–298.15 K; Ultrasonic Velocities at 298.15 K for the Three Binary Systems Inclusive of Tetrahydrofuran + Water and the Ternary System Tetrahydrofuran + Water + γ-Butyrolactone

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Densities of γ-butyrolactone + water and tetrahydrofuran + γ-butyrolactone systems were measured over temperature ranges of 303.15–343.15 and 278.15–298.15 K respectively, and ultrasonic velocities were measured at 298.15 K for the ternary system tetrahydrofuran + water + γ-butyrolactone and all three binary systems inclusive of the tetrahydrofuran + water system. Excess volumes and excess isentropic compressibilities were calculated from measured data and reported for all the systems studied. The data have been fitted to empirical models, and the model constants are also reported.

Introduction

The tetrahydrofuran (THF) + water system has been well studied with regards to its properties (*1*). We have earlier reported the enthalpies of mixing of THF + γ-butyrolactone (GBL) and GBL + water systems (*2*). In the present investigation we report the excess volumes and excess isentropic compressibilities from the measured density and ultrasonic velocity data of the binaries and the ternary system.

Experimental Section

Tetrahydrofuran supplied by Sarabhai Chemicals, India, was passed over activated alumina, refluxed over KOH pellets for 6 h, and finally distilled and collected over sodium wire. The purity of THF was found to be 99.6 mol % by GLC on Carbowax 29M by use of a thermal conductivity detector (TCD). γ-Butyrolactone of Fluka AG, Switzerland, was passed over a 13X molecular sieve column and distilled under reduced pressure in a 0.6-*m* packed column. A suitable fraction was collected and used for the experiment. The purity of GBL was found to be 99 mol % as determined by GLC on a column packed with 12% EGS (ethylene glycol succinate) using TCD. Double-distilled deionized water with 2×10^{-4} S·m conductivity was used for the measurements.

Densities were measured with a Lipkin pycnometer. The volume between marked lines of the pycnometer was determined by measuring the density of distilled mercury at various temperatures (*3*). The diameter of the capillary was determined by using a mercury thread with a traveling microscope. Mixtures of various compositions were prepared by weighing with an accuracy of $\pm 1 \times 10^{-5}$ in mole fraction on a Sartorius, FRG, balance. Care was taken not to allow any air bubble in the pycnometer. The pycnometer was thermostated with an accuracy of ±0.01 K. Low temperatures were achieved by using a Neslab, USA, cryounit. Temperatures were measured with an accuracy of ±0.025 K by use of a platinum resistance thermometer supplied by Minco Products, USA. The liquid levels in the pycnometer were read up to $\pm 1 \times 10^{-4}$ m with a cathetometer. Other precautions were taken, such as flushing the pycnometer with dry air and closing with caps as reported by Bauer (*4*). The overall accuracy of density measurements is estimated to be ± 0.1 kg·m^{-3}. Our pure component density data are compared with the literature values in Table I. Pure component density data of GBL measured by us from 298.15 to 453.15 K (data from 298.15 to 343.15 K

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Table I. Densities of Pure Components at 298.15 K

compd	ρ/kg·m^{-3}	ρ(lit.)/kg·m^{-3}
THF	882.3	882.4[a] 881.2[b]
GBL	1123.7	1124.36[c] 1124.2[d] 1125.3[b]
H_2O	997.0	997.02[e]

[a] Reference 5. [b] Reference 9. [c] Reference 6. [d] Reference 7. [e] Reference 8.

Table II. Coefficients of Eq 1 and Standard Deviation, s, for GBL[a]

A_1	A_2	A_3	A_4	s	range/K
3.0295	−1.6640	−5.4401	5.4162	0.0097	298.15–709 (T_c)

[a] $s = [\sum(\text{expt} - \text{calc})^2/(n - 1)]^{1/2}$; n = number of data points.

Table III. Densities, ρ_m, of $(x)H_2O + (1 - x)GBL$

	ρ_m/kg·m^{-3}				
x	303.15 K	313.15 K	323.15 K	333.15 K	343.15 K
0.0000	1118.9	1108.8	1099.4	1089.7	1080.1
0.0792	1116.4	1106.3	1096.9	1087.2	1077.3
0.0898	1116.0	1106.0	1096.6	1086.8	1077.0
0.2337	1111.9	1102.0	1092.6	1082.4	1072.0
0.6207	1090.0	1081.0	1072.1	1062.9	1051.9
0.9031	1042.7	1035.5	1029.0	1021.6	1014.0
0.9395	1029.2	1023.2	1017.4	1010.6	1003.5
1.0000	995.6	992.2	988.2	983.4	977.8

Table IV. Densities, ρ_m, of (x)THF + $(1 - x)$GBL

	ρ_m/kg·m^{-3}				
x	278.15 K	283.15 K	288.15 K	293.15 K	298.15 K
0.0000	1142.5	1137.7	1133.6	1128.0	1123.7
0.1077	1145.2	1113.1	1108.1	1103.1	1098.4
0.286	1096[a]	1072[a]	1066[a]	1060[a]	1055[a]
	1075[b]	1072[b]	1068[b]	1064[b]	1061[b]
0.3124	1091.8	1066.5	1060.3	1054.5	1049.2
0.483	1044[a]	1025[a]	1019[a]	1013[a]	1008[a]
	1028[b]	1024[b]	1019[b]	1015[b]	1010[b]
0.7010	986.0	972.8	966.7	960.8	955.6
0.8954	932.7	924.8	919.2	913.5	908.4
1.0000	903.6	898.3	892.8	887.2	882.3

[a] Interpolated from our experimental data. [b] Gurevich et al. (*9*).

reported here) are correlated with temperature by using the Guggenheim equation:

$$\rho - \rho_c = \sum A_i(1 - T_r)^{i/3} \tag{1}$$

where $T_r = T/T_c$. A T_c value of GBL equal to 709 K was taken from ref *9*. V_c was estimated according to the methods of Lydersen (*10*) and Vetere (*11*). Lydersen's method yielded ρ_c = 372 kg·m^{-3} while ρ_c = 269 kg·m^{-3} was obtained by use of Vetere's method. Z_c calculated by using both ρ_c values varied from 0.13 to 0.19, which is considerably lower than most of the reported values. Hence the final ρ_c value selected, i.e., ρ_c = 290 kg·m^{-3}, was based on the "best" fit of eq 1 with our experimental density data for GBL. The coefficients of eq 1 are given in Table II.

Ultrasonic velocities were measured with a setup previously used by Venkateswarlu et al. (*12*) using a single-crystal interferometer at 2-MHz frequency, with ±0.15% accuracy.

Results

Densities of $(x)H_2O + (1 - x)$GBL are presented in Table III and of (x)THF + $(1 - x)$GBL in Table IV. Figures 1 and 2 provide the excess molar volumes V_m^E calculated from the densities for each x and T. The errors in the excess volumes are calculated to be $\pm 0.005 \times 10^{-3}$ m^3·kmol^{-1}. Figure 1 also shows the comparison of our data with the literature (*6*, *7*).

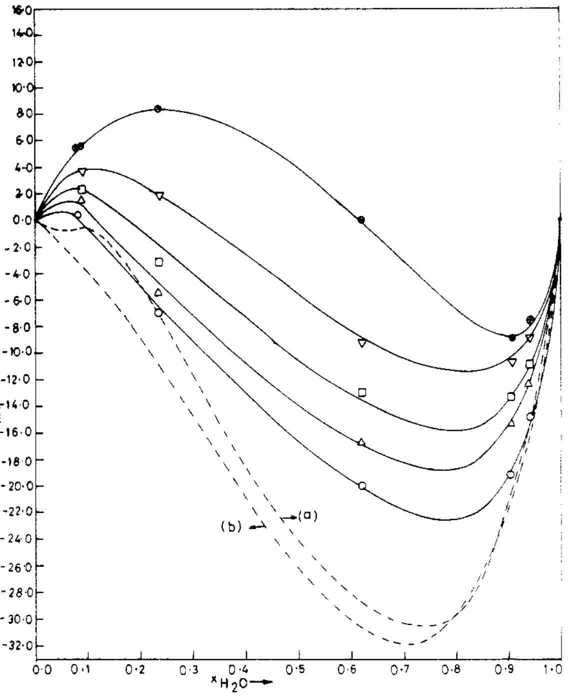

Figure 1. Plot of excess molar volume against mole fraction for $(x)H_2O + (1 - x)$GBL. (a) ---, ref *6* at 298.15 K; (b) ---, ref *7* at 298.15 K; —, eq 2; ○, 303.15 K; △, 313.15 K; □, 323.15 K; ▽, 333.15 K; ⊗, 343.15 K.

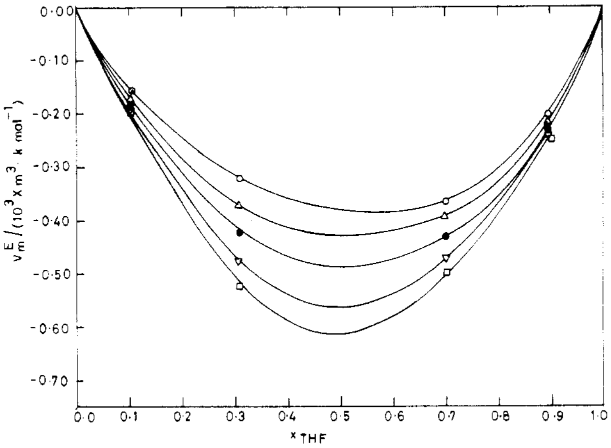

Figure 2. Plot of excess molar volume against mole fraction for (x)THF + $(1 - x)$GBL. —, eq 3; □, 278.15 K; ▽, 283.15 K; ●, 288.15 K; △, 293.15 K; ○, 298.15 K.

Table V. Coefficients of Eq 2 and Standard Deviation, s, for $(x)H_2O + (1 - x)$GBL[a]

T/K	A_1	A_2	B_1	B_2	s
303.15	−0.6601	−0.8092	0.2172	−0.8730	0.0044
313.15	−0.5513	−0.7113	0.2313	−0.8455	0.0046
323.15	−0.4141	−0.6241	0.1425	−0.7970	0.0095
333.15	−0.2221	−0.5844	0.0549	−0.6213	0.0028
343.15	0.1485	−0.6770	−0.3908	−0.4394	0.0051

[a] $s = [\sum(\text{expt} - \text{calc})^2/(n - 1)]^{1/2}$; n = number of data points.

Excess volumes of the H_2O + GBL system at each isotherm were correlated by use of a Pade approximant, eq 2, and those of the THF + GBL system by eq 3. The coefficients of eq 2

$$V_m^E/(\text{m}^3\cdot\text{kmol}^{-1} \times 10^3) = x(1 - x) \times [\{A_1 + A_2(2x - 1)\}/\{1 + B_1(2x - 1) + B_2(2x - 1)^2\}] \tag{2}$$

$$V_m^E/(\text{m}^3\cdot\text{kmol}^{-1} \times 10^3) = x(1 - x)\sum a_i(1 - 2x)^i \tag{3}$$

and 3 (obtained by nonlinear regression) are given in Tables V

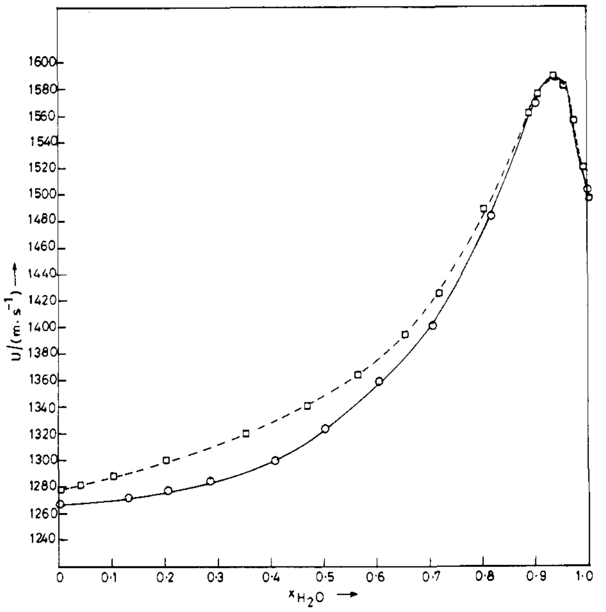

Figure 3. Composition dependence of ultrasonic velocity for (x)THF + $(1 - x)H_2O$ at 298.15 K, 2 MHz. -□-, ref *15*; O, present work.

Table VI. Coefficients of Eq 3 and Standard Deviation, s, for (x)THF + $(1 - x)$GBL[a]

T/K	a_0	a_1	a_2	a_3	s
278.15	-2.4427	-0.1965	0.2861	1.0478	0.32×10^{-6}
283.15	-2.2505	-0.1137	0.1030	0.8350	0.34×10^{-6}
288.15	-1.9501	0.0302	-0.3274	0.5600	0.15×10^{-6}
293.15	-1.7032	0.1317	-0.6155	0.3051	0.26×10^{-6}
298.15	-1.5214	0.2927	-0.6037	0.0594	0.31×10^{-6}

[a] $s = [\sum(\text{expt} - \text{calc})^2/(n - 1)]^{1/2}$; n = number of data points.

and VI, respectively. The regression coefficients so obtained were correlated with temperature

$$A_k = C_0 + C_1(T/\mathrm{K}) + C_2(T/\mathrm{K})^2 \qquad (4)$$

and the constants of eq 4 are listed in Table VII for both the systems. The maximum uncertainity in V_m^E calculated by using eq 4 is $\pm 0.01 \times 10^{-3}$. Our values of mixture densities of THF + GBL system are compared with those of Gurevich et al. in Table IV, and the agreement is satisfactory.

The measured ultrasonic velocities and densities for each system were used to calculate isentropic compressibilities K_s (eq 5) as a function of mole fraction, x, and volume fraction, ϕ, where $\phi = x_iv_i/\sum x_iv_i$.

$$K_s/(\mathrm{TPa}^{-1}) = [(u/\mathrm{m{\cdot}s^{-1}})(\rho/\mathrm{kg{\cdot}m^{-3}})] \qquad (5)$$

These data are reported in Tables VIII and IX for the H_2O + GLB system, in Table X for the THF + GBL system, in Table XI for the THF + H_2O system, and in Table XII for the THF + GBL + H_2O system. For the THF + H_2O system density data were taken from Kiyohara and Benson (*14*). Our ultrasonic velocity data are compared in Figure 3 with those of Kiyohara et al. (*15*). Excess isentropic compressibilities were calculated by using eq 6 and 7:

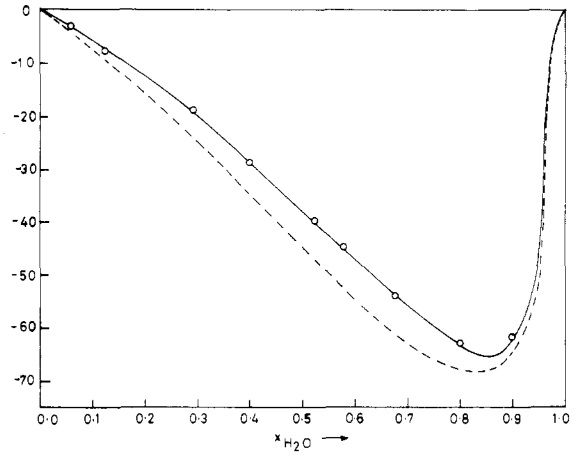

Figure 4. Dependence of excess isentropic compressibility calculated by using eq 6 on mole fraction for $(x)H_2O$ + $(1 - x)$GBL at 298.15 K, 2 MHz. ---, ref *7*; —, fitted; O, this work.

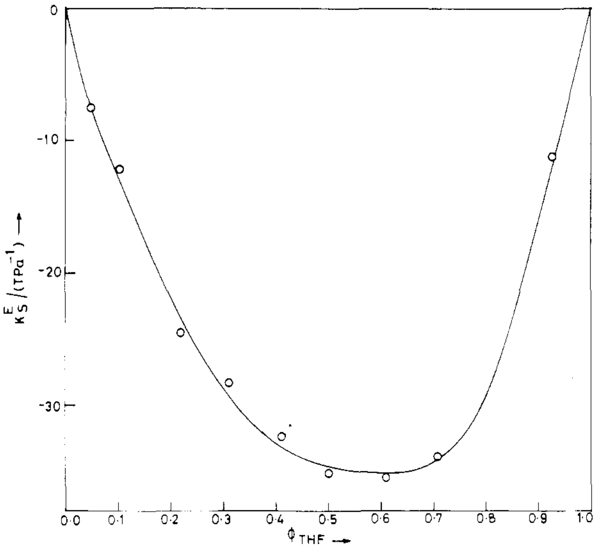

Figure 5. Dependence of excess isentropic compressibility calculated by using eq 7 on volume fraction for (ϕ)THF + $(1 - \phi)$GBL at 298.15 K, 2 MHz. — eq 8; O, experimental.

$$K_s^E/(\mathrm{TPa}^{-1}) = K_s - (xK_{s1} + (1 - x)K_{s2}) \qquad (6)$$

$$K_s^E/(\mathrm{TPa}^{-1}) = K_s - (\phi K_{s1} + (1 - \phi)K_{s2}) \qquad (7)$$

where x and ϕ are mole and volume fractions, respectively.

Our K_s^E values are accurate to ± 2 TPa^{-1}. K_s^E values of $(x)H_2O$ + $(1 - x)$GBL calculated by using eq 6 are compared with the literature data (*7*) in Figure 4. Figures 5 and 6 show

Table VII. Coefficients of Eq 4

	C_0	C_1	C_2	s^a	range/K
			$(x)H_2O + (1 - x)$GBL		
A_1	35.2497	-0.2402	4.02×10^{-4}	0.024	303.15-343.15
A_2	-32.7488	0.1949	-2.95×10^{-4}	0.019	303.15-343.15
B_1	-61.7744	0.3974	-6.36×10^{-4}	0.049	303.15-343.15
B_2	27.2972	-0.1847	3.03×10^{-4}	0.014	303.15-343.15
			(x)THF + $(1 - x)$GBL		
a_0	-16.6694	0.0542	-1.11×10^{-5}	0.030	278.15-298.15
a_1	1.8221	-0.0370	1.07×10^{-4}	0.160	278.15-298.15
a_2	43.7732	-0.2556	3.57×10^{-4}	0.103	278.15-298.15
a_3	8.6147	-0.0057	-7.71×10^{-5}	0.013	278.15-298.15

[a] $s = [\sum(\text{expt} - \text{calc})^2/(n - 1)]^{1/2}$; n = number of data points.

Table VIII. Ultrasonic Velocities, u, and Isentropic Compressibilities, K_s, of $(x)H_2O + (1 - x)GBL$ at 2 MHz, 298.15 K

x	$u/m{\cdot}s^{-1}$	K_s/TPa^{-1}	x	$u/m{\cdot}s^{-1}$	K_s/TPa^{-1}
0.0000	1510	390.3	0.5799	1555	376.8
0.0555	1512	389.8	0.6763	1570	372.8
0.1231	1515	389.0	0.7990	1586	371.0
0.2908	1522	387.4	0.9003	1591	377.4
0.4007	1535	382.6	1.0000	1502	444.6
0.5229	1548	378.8			

Table IX. Isentropic Compressibility Data for $(\phi)H_2O + (1 - \phi)GBL$ at 298.15 K, 2 MHz, in Terms of Volume Fractions

ϕ	K_s/TPa^{-1}	ϕ	K_s/TPa^{-1}	ϕ	K_s/TPa^{-1}
0.0000	390.3	0.3301	372.8	0.8499[a]	418.0
0.0137	389.8	0.4839	371.0	0.8841[a]	425.0
0.0320	389.0	0.6805	377.4	0.9204[a]	431.5
0.0882	387.4	0.7174[a]	386.5	0.9589[a]	438.3
0.1362	382.6	0.7581[a]	397.5	0.9791[a]	442.0
0.2054	378.8	0.8175[a]	411.3	1.0000	444.6
0.2456	376.8				

[a] Extrapolated values from the mole fraction data fit.

Table X. Ultrasonic Velocities u, and Isentropic Compressibilities, K_s, of $(\phi)THF + (1 - \phi)GBL$ at 298.15 K, 2 MHz

ϕ	$u/m{\cdot}s^{-1}$	K_s/TPa^{-1}	ϕ	$u/m{\cdot}s^{-1}$	K_s/TPa^{-1}
0.0000	1510	390.3	0.5036	1389	514.8
0.0512	1501	399.0	0.6126	1363	548.9
0.1032	1487	410.8	0.7084	1341	581.0
0.2181	1463	434.8	0.9264	1284	672.6
0.3113	1436	460.6	1.0000	1266	707.2
0.4134	1410	489.0			

Table XI. Ultrasonic Velocities, u, and Isentropic Compressibilities, K_s, of $(\phi)THF + (1 - \phi)H_2O$ at 298.15 K, 2MHz

ϕ	$u/m{\cdot}s^{-1}$	K_s/TPA^{-1}	ϕ	$u/m{\cdot}s^{-1}$	K_s/TPA^{-1}
0.0000	1502	444.6	0.8188	1323	622.6
0.0200	1586	407.3	0.8658	1300	649.2
0.3302	1567	421.7	0.9193	1284	671.4
0.5061	1483	478.9	0.9457	1276	683.5
0.6564	1400	545.5	0.9677	1272	691.6
0.7465	1357	586.4	1.0000	1266	707.1

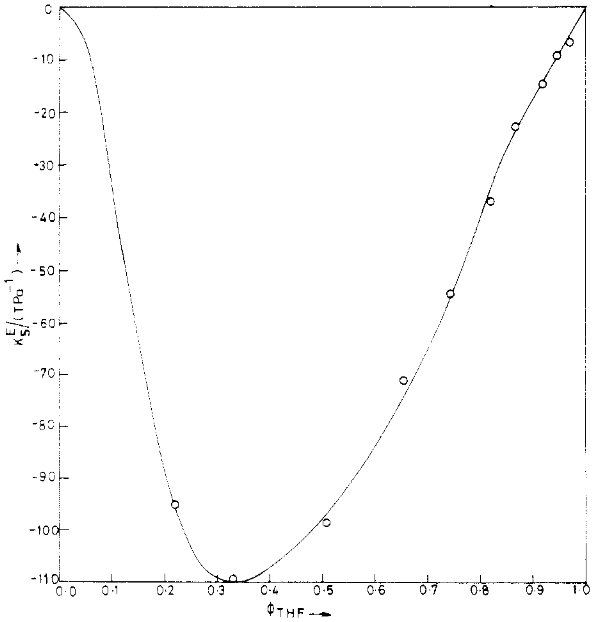

Figure 6. Dependence of excess isentropic compressibility calculated by using eq 7 on volume fraction for $(\phi)THF + (1 - \phi)H_2O$ at 298.15 K, 2 MHz. — eq 8; O, experimental.

Table XII. Ultrasonic Velocity Data of $(\phi_1)THF + (\phi_2)GBL + (1 - \phi_1 - \phi_2)H_2O$ at 298.15 K, 2 MHz

$(\phi_1)THF$	$(\phi_2)GBL$	$(1 - \phi_1 - \phi_2)H_2O$	$u/m{\cdot}s^{-1}$
0.8688	0.1056	0.0256	1307
0.8195	0.1215	0.0590	1325
0.7974	0.1160	0.0865	1334
0.7343	0.1382	0.1275	1360
0.6540	0.1539	0.1921	1392
0.5680	0.1843	0.2477	1425
0.4388	0.2122	0.3490	1482
0.2763	0.2487	0.4751	1549
0.7584	0.2066	0.0350	1328
0.7428	0.2100	0.0472	1335
0.5143	0.1871	0.2986	1451
0.5859	0.2895	0.1246	1392
0.6363	0.2276	0.1361	1381
0.2553	0.4419	0.3028	1519
0.4632	0.2801	0.2567	1335
0.6373	0.3201	0.0426	1352
0.6273	0.3270	0.0457	1359
0.5493	0.3657	0.0850	1582
0.4605	0.4148	0.1246	1425
0.2884	0.5157	0.1959	1479
0.2424	0.5170	0.2406	1514
0.5364	0.3226	0.1410	1380
0.4504	0.4989	0.0507	1460
0.4411	0.4697	0.0892	1419
0.4177	0.4697	0.1126	1438
0.1786	0.6388	0.1827	1504
0.4453	0.4687	0.0860	1420
0.3521	0.3746	0.2733	1408
0.2680	0.6563	0.0757	1461
0.3405	0.6074	0.0521	1435
0.2330	0.7168	0.0502	1457
0.1531	0.7594	0.0875	1488
0.2204	0.7409	0.0387	1462
0.1227	0.8271	0.0502	1486
0.1171	0.8229	0.0600	1488

Table XIII. Coefficients of Eq 8 for $(\phi)H_2O + (1 - \phi)GBL$, $(\phi)THF + (1 - \phi)GBL$, and $(\phi)THF + (1 - \phi)H_2O$ at 298.15 K

const	$(\phi)H_2O + (1 - \phi)GBL$	$(\phi)THF + (1 - \phi)GBL$	$(\phi)THF + (1 - \phi)H_2O$
a_0	-15.43	-13.80	-38.93
a_1	18.88	1.83	-21.85
a_2	-7.34	-9.02	-27.13
a_3	8.16	12.25	-43.38
a_4	-43.52	9.15	69.00
a_5	34.95	-18.07	94.60
s^a	0.28	0.49	1.07

[a] $s = [\sum(\text{expt} - \text{calc})^2/(n - 1)]^{1/2}$; n = number of data points.

the K_s^E values calculated by using eq 7 for THF + GBL and THF + H_2O systems, respectively. These have been fitted to the equation:

$$K_s^E/(TPa^{-1}) = \phi(1 - \phi)\sum a_j(1 - 2\phi)^j \qquad (8)$$

The coefficients of eq 8 are given in Table XIII.

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Registry No. GBL, 96-48-0; THF, 109-99-9; H_2O, 7732-18-5.

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Mixture Properties of the Water + γ-Butyrolactone + Tetrahydrofuran System. 2. Viscosities and Surface Tensions of γ-Butyrolactone + Water at 303.15–343.15 K and γ-Butyrolactone + Tetrahydrofuran at 278.15–298.15 K

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Viscosity and surface tension measurements of (x)water + (1 − x)γ-butyrolactone (GBL) and (x)tetrahydrofuran + (1 − x)GBL were measured over temperature ranges of 303.15–343.15 and 278.15–298.15 K, respectively. Data are correlated to models for interpolation and limited extrapolation.

Introduction

In part 1 of this series (*1*) density and sound velocity measurements have been reported for water + γ-butyrolactone (GBL) and tetrahydrofuran (THF) + GBL systems. Viscosities of THF + water are available in the literature (*2*). In the present investigation viscosity and surface tension measurements are presented for the above systems.

Experimental Section

The method of purification and the purity of samples are reported in part I (*1*). Viscosities were measured with a Ubbelohde viscometer. Efflux times were measured up to ±0.1 s. Triple-distilled water and carefully purified toluene were used as calibrating fluids to determine the viscometer constant. Viscosities of pure THF and GBL at 298.15 K are reported in Table I. The uncertainty in our viscosity measurement is estimated to be $\pm 0.001 \times 10^{-3}$ N·s·m^{-2}.

Table I. Viscosity of Pure Components GBL, THF, and H_2O at 298.15 K

compd	μ/N·s·m^{-2} × 10^3	μ(lit.)/N·s·m^{-2} × 10^3
GBL	1.738	1.7315;^{b} 1.575^c
THF	0.454	0.455;^{d} 0.470^c
H_2O^a	0.7967	0.798^e

^{a} At 303.15 K. ^{b} Reference *3*. ^{c} Reference *9*. ^{d} Reference *4*. ^{e} Reference *5*.

Table II. Surface Tension Data of Some Pure Components

compd	T/K	σ/N·m^{-1} × 10^3	σ(lit.)^a/N·m^{-1} × 10^3
ethanol	303.15	21.55	21.48
n-octane	318.15	19.12	19.24
	328.15	18.23	18.29
	338.15	17.26	17.33
H_2O	303.15	71.19	71.40
	343.15	64.23	65.49
THF	298.15	26.59	26.40^b

^{a} Reference *6*. ^{b} Reference *4*.

Table III. Viscosity of (x)H_2O + (1 − x)GBL

	μ/N·s·m^{-2} × 10^3				
x	303.15 K	313.15 K	323.15 K	333.15 K	343.15 K
0.0000	1.6097	1.3901	1.2131	1.0766	0.9513
0.0898	1.5598	1.3391	1.1635	1.0389	0.9055
0.2337	1.5946	1.3492	1.1594	1.0129	0.8893
0.6207	1.6736	1.3485	1.1116	0.9330	0.7994
0.9031	1.2572	1.0049	0.8246	0.6901	0.5875
1.0000	0.7967	0.6522	0.5470	0.4689	0.4097

Table IV. Viscosity of (x)THF + (1 − x)GBL

	μ/N·s·m^{-2} × 10^3				
x	278.15 K	283.15 K	288.15 K	293.15 K	298.15 K
0.0000	2.546^a	2.285^a	2.072^a	1.892^a	1.738
0.1077	2.070	1.877	1.717	1.583	1.462
0.286	1.545^b	1.418^b	1.298^b	1.210^b	1.124^b
	1.462^c	1.365^c	1.278^c	1.198^c	1.127^c
0.3124	1.494	1.367	1.261	1.172	1.089
0.483	1.182^b	1.105^b	1.020^b	0.955^b	0.890^b
	1.030^c	0.967^c	0.910^c	0.858^c	0.810^c
0.7010	0.827	0.772	0.724	0.682	0.641
0.8954	0.639	0.602	0.569	0.539	0.510
1.0000	0.564	0.534	0.506	0.480	0.454

^{a} Extrapolated values using eq 1, coefficients of which are given in Table V. ^{b} Interpolated from our experimental data. ^{c} Reference *9*.

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