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Received for review July 17, 1988. Accepted May 30, 1989. We thank Hindustan Lever Research Foundation, Bombay, India, for supporting this work.

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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Table V. Coefficients of Eq 1 for GBL, THF, and H_2O

compd	ln A	α	β	T_0/K	s^a	range/K
GBL	17.9634	-3.1254	27.2464	229.62	0.022	298.15-343.15
THF	89.4202	-14.9602	-664.0694	-164.64	0.020	278.15-298.15
H_2O	22.4872	-4.0267	8.8893	273.15	0.002	298.15-343.15

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

Table VI. Coefficients of Eq 3 for $(x)H_2O + (1 - x)GBL$

T/K	a_0	a_1	a_2	a_3	s^a
303.15	1.5347	-1.5980	1.3707	-1.3737	0.0001
313.15	1.4257	-1.4402	1.2720	-1.3147	0.0001
323.15	1.3305	-1.2893	1.1840	-1.2769	0.0001
333.15	1.2253	-1.1462	1.0987	-1.2062	0.0001
343.15	1.1715	-0.9965	0.9160	-1.1750	0.0001

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

Table VII. Coefficients of Eq 3 for $(x)THF + (1 - x)GBL$

T/K	a_0	a_1	a_2	a_3	s^a
278.15	-0.2746	0.0748	-0.2159	-0.2396	0.52×10^{-7}
283.15	-0.2672	0.0783	-0.1918	-0.2129	0.54×10^{-7}
288.15	-0.2584	0.0739	-0.1473	-0.1749	0.67×10^{-7}
293.15	-0.2446	0.0814	-0.1002	-0.1571	0.46×10^{-7}
298.15	-0.2367	0.0828	-0.0647	-0.1727	0.34×10^{-7}

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

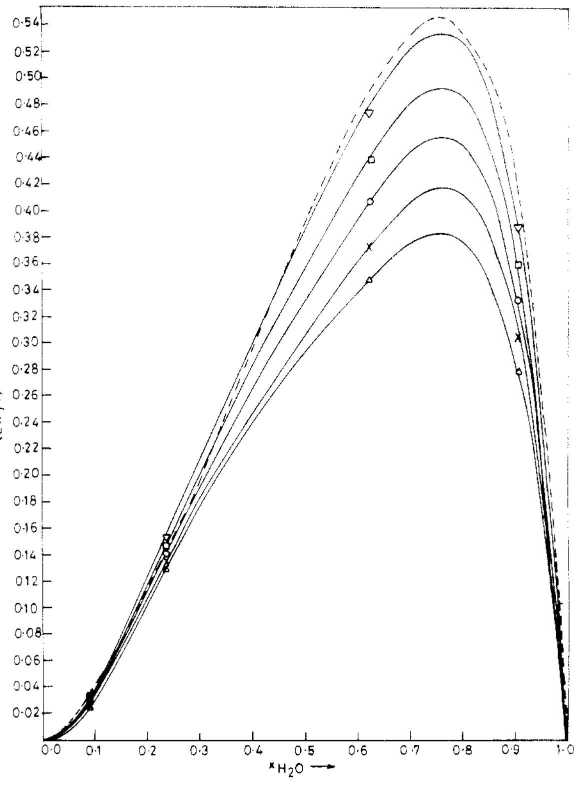

Figure 1. Dependence of excess logarithmic viscosity on mole fraction for $(x)H_2O + (1 - x)GBL$. ---, ref *3* at 298.15 K. —, eq 4. Experimental: ∇, 303.15 K; □, 313.15 K; O, 323.15 K; ×, 333.15 K; Δ, 343.15 K.

Surface tensions were measured according to the capillary rise method. The diameter of the capillary was determined by use of a mercury thread with traveling microscope. Surface tensions of some selected pure components are given in Table II for comparison. Our surface tension data are accurate to $\pm 1.4 \times 10^{-3}$ N·m^{-1}.

Results

Viscosities of (x)water + $(1 - x)$GBL and (x)THF + $(1 - x)$GBL are given in Tables III and IV along with values of pure components. Pure component viscosity data are correlated with temperature by using eq 1:

$$\ln \mu/(\text{N·s·m}^{-2} \times 10^3) = \ln A + \alpha \ln T + \beta/(T - T_0) \quad (1)$$

where T_0 is the freezing point of the pure component. Coefficients of eq 1 are given in Table V. Values of the excess of logarithmic viscosity, which represents the free energy of activation, for flow have been calculated by using eq 2 and are shown in Figures 1 and 2. $(\ln \mu)^E$ readily gives the parameter G in the Grunberg–Nissan equation (*7*, *8*). Figure 1 also shows the literature data (*3*) for H_2O + GBL system. Coefficients of

$$(\ln \mu)^E = \ln \mu - (x \ln \mu_1 + (1 - x) \ln \mu_2) \quad (2)$$

$$(\ln \mu)^E = x(1 - x)\sum a_j(1 - 2x)^j \quad (3)$$

eq 3 are given in Tables VI and VII. The regression coefficients so obtained were correlated with temperature:

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

and the constants of eq 4 are listed in Table VIII for both systems.

Our values of viscosity of the THF + GBL system are compared in Table IV with those reported by Gurevich et al. (*9*) at two concentrations. The agreement at 298.15 K is better than at lower temperatures. Pure component viscosities for GBL and THF reported by Gurevich et al. (*9*) differ considerably from the literature values, especially for GBL as reported in Table I.

Table VIII. Coefficients of Eq 4

	C_0	C_1	C_2	s^a	range/K
			$(x)H_2O + (1 - x)GBL$		
A_0	11.5875	-0.0542	6.96×10^{-5}	0.0078	303.15-343.15
A_1	-7.7268	0.0249	-1.53×10^{-5}	0.0023	303.15-343.15
A_2	-7.3704	0.0638	-1.15×10^{-4}	0.0180	303.15-343.15
A_3	-4.5561	0.0153	-1.58×10^{-5}	0.0081	303.15-343.15
			$(x)THF + (1 - x)GBL$		
A_0	0.1220	-0.0046	1.14×10^{-5}	0.0015	278.15-298.15
A_1	1.0640	-0.0072	1.32×10^{-5}	0.0023	278.15-298.15
A_2	1.6480	-0.0203	4.90×10^{-5}	0.0052	278.15-298.15
A_3	-15.6493	0.1036	-1.73×10^{-4}	0.0093	278.15-298.15

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

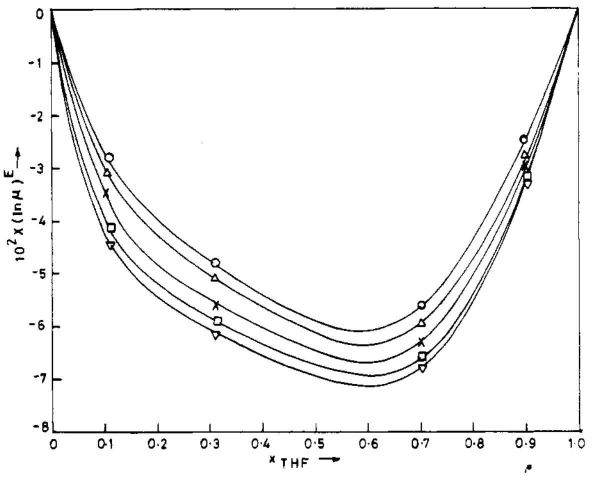

Figure 2. Dependence of excess logarithmic viscosity on mole fraction for (x)THF + $(1 - x)$GBL. —, eq 4. Experimental: ▽, 278.15 K; □, 283.15 K; ×, 288.15 K; △, 293.15 K; ○, 298.15 K.

Table IX. Surface Tension of $(x)H_2O + (1 - x)$GBL

	$\sigma/N{\cdot}m^{-1} \times 10^3$				
x	303.15 K	313.15 K	323.15 K	333.15 K	343.15 K
0.0000	42.69	41.88	40.38	39.65	37.94
0.0898			40.67	40.34	38.66
0.1155	43.18	42.37	41.03	40.41	38.80
0.2337	42.95	42.66	41.45	41.00	39.53
0.6207	44.84	44.06	43.12	42.68	41.58
0.9071	52.04	50.86	49.70	49.10	47.46
1.0000	71.19	69.15	67.19	66.16	64.23

Surface tensions of (x)water + $(1 - x)$GBL and (x)THF + $(1 - x)$GBL are given in Tables IX and X. Pure component surface tensions for GBL, THF, and water given in Tables IX and X are correlated with temperature by using the relation

$$\sigma/(N{\cdot}m^{-1} \times 10^3) = a - bT(K) \qquad (5)$$

Coefficients of eq 5 are given in Table XI.

Table X. Surface Tension of (x)THF + $(1 - x)$GBL

	$\sigma/N{\cdot}m^{-1} \times 10^3$			
x	278.15 K	283.15 K	293.15 K	298.15 K
0.0000[a]	45.79	45.21	44.03	43.44
0.3124	37.28	36.80	35.94	35.19
0.7010	32.08	31.14	30.31	29.25
0.8954	30.25	29.12	28.07	27.25
1.0000	29.50	28.51	27.37	26.59

[a] Extrapolated values from eq 5, coefficients of which are given in Table XI.

Table XI. Coefficients of Eq 5

compd	a	b	s^a	range/K
GBL	78.45	0.1174	0.24	303–343
THF	68.04	0.1392	0.18	278–298
H_2O	122.23	0.1691	0.27	303–343

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

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

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Received for review July 13, 1988. Accepted May 30, 1989. We thank Hindustan Lever Research Foundation, Bombay, India, for supporting this project.