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Density and Viscosity of Propyl Formate + Aromatic Hydrocarbons at T = (303.15, 308.15, and 313.15) K

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Density and viscosity data of propyl formate + benzene, isopropyl benzene, isobutyl benzene, or butylbenzene have been determined at T = (303.15, 308.15, and 313.15) K. From this data, the excess volume, V^E, deviation in viscosity, $\Delta\eta$, and excess Gibbs energy of activation, ΔG^{*E}, have been estimated. The results of V^E have been correlated using the semiempirical equation proposed by Hwang et al. The viscosity data have been correlated with the equation of Tamura and Kurata, Heric, and Auslander.

Introduction

The viscosity of liquid mixtures has attracted much attention in the literature from both practical and theoretical viewpoints to gain a clearer insight into the behavior of liquid mixtures. In the chemical and petroleum industries, the viscosity of pure components and mixtures is an important property in hydraulics calculations for surface facilities and pipeline systems.

Propyl formate is an isomer of propan-2-ol. It is used as a solvent in the pharmaceutical industry and for resins and cellulose esters. Other uses include the manufacturing of perfumes, insecticides, and fungicide fumigation. Therefore, its presence and interactions with the aromatic hydrocarbons are of interest to understand mechanisms of solvation. Emmerling et al.[1] have measured the kinematic viscosities of benzene + propyl formate, while other authors[2,3] have measured the density, refractive index, and speed of sound of isopropyl acetate and propyl acetate with aromatic hydrocarbons. As an extension of our earlier work[4−6] along these lines, we present in this paper densities and viscosities of binary mixtures of propyl formate with benzene, isopropyl benzene, isobutyl benzene, and butylbenzene at (303.15, 308.15, and 313.15) K. This study will provide information about the molecular interactions on the formation of constituent components and their influence on the excess properties of the mixtures. From the experimental data, the excess molar volume, V^E, deviation in viscosity, $\Delta\eta$, and excess Gibbs energy of activation, ΔG^{*E}, have been calculated. These excess or deviation properties have been fitted to the Redlich−Kister[7] polynomial equation. Further, V^E results were also fitted to the semiempirical equation proposed by Hwang et al.[8] In addition, the viscosity data were correlated with the Tamura and Kurata,[9] Heric,[10] and Auslander[11] models.

Experimental Section

Propyl formate, benzene, isopropyl benzene, isobutyl benzene, and butylbenzene all provided by Fluka, analytical grade of mass purity > 99.5 %, were used without purification, owing to their high purity grade. The purity of these liquids was verified by measuring their density and viscosity at 303.15 K, which agreed satisfactorily with the corresponding literature values (Table 1). Binary mixtures were prepared by mass in airtight ground stoppered bottles. The mass measurements accurate to ± 0.01 mg were made on a digital electronic balance (Mettler AE 240, Switzerland). To prevent the sample from preferential evaporation, the mixtures were prepared by transferring aliquots via syringe into stoppered bottles. A set of nine compositions of the mole fraction varying from 0.1 to 0.9 in steps of 0.1 was prepared. The mixtures were completely miscible over the whole composition range. The possible uncertainty in the mole fraction was estimated to be less than $\pm 1 \cdot 10^{-4}$. Densities of pure liquids and their mixtures were determined by using an Anton Paar digital density meter (model: DMA 35) with an uncertainty of ± 0.08 %.

Table 1. Comparison of Experimental Densities (ρ) and Viscosities (η) of Pure Liquids with Literature Values at 303.15 K

		$\rho \cdot 10^{-3}$/kg·m^{-3}		η/mPa·s	
liquid	T/K	exptl	lit.	exptl	lit.
propyl formate	303.15	0.8935	0.8942[5]	0.477	0.482[1]
	308.15	0.8886	0.8885[5]	0.456	
	313.15	0.8838	0.8828[5]	0.431	0.432[1]
benzene	303.15	0.8676	0.8683[12]	0.569	0.562[12]
	308.15	0.8632		0.534	0.533[14]
	313.15	0.8589	0.8581[1]	0.497	
isopropyl benzene	303.15	0.8527	0.8532[a]	0.690	0.687[a]
	308.15	0.8490		0.651	
	313.15	0.8454		0.611	
isobutyl benzene	303.15	0.8444	0.8450[a]	0.905	
	308.15	0.8411		0.848	
	313.15	0.8376		0.784	
butylbenzene	303.15	0.8513	0.8522[13]	0.901	0.893[13]
	308.15	0.8480		0.844	
	313.15	0.8446		0.787	

[a] Extrapolated value from ref 12.

Kinematic viscosities were obtained with a modified suspended level Ubbelohde viscometer. The viscometer has been calibrated so as to determine the two constants A and B in the equation $\eta = \rho(At - B/t)$, obtained by measuring the flow time t with pure water and benzene. The flow time of a definite volume of liquid through the capillary was measured with the help of a precalibrated RACER stopwatch corrected to ± 0.1 s. The viscometer was always kept in a vertical position in a water thermostat at the required temperature. Four to five sets of

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readings for the flow times were taken for each pure liquid or liquid mixture, and the arithmetic mean was taken for the calculations. The dynamic viscosities were calculated as ($\eta = \upsilon\rho$).

The uncertainty in viscosities thus estimated was found to be ± 0.7 %. In all property measurements, the temperature was controlled within ± 0.01 K using a constant temperature bath (model: IRI O16C, INSREF India) by circulating water from the thermostat.

Results and Discussion

The densities (ρ) and viscosities (η) of mixtures at (303.15, 308.15, and 313.15) K are listed as a function of mole fraction in Table 2. The density values have been used to calculate excess molar volumes V^E using the following equation.

$$V^E\cdot 10^6/(\mathrm{m^3\cdot mol^{-1}}) = (x_1M_1 + x_2M_2)/\rho_{12} - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \qquad (1)$$

where ρ_{12} is the density of the mixture and x_1, M_1, and ρ_1 and x_2, M_2, and ρ_2 are the mole fraction, the molecular weight, and the density of pure components 1 and 2, respectively. The calculated excess molar volumes are included in Table 2. The viscosity deviations $\Delta\eta$ were calculated using

$$\Delta\eta\ (\mathrm{mPa\cdot s}) = \eta_{12} - (x_1\eta_1 + x_2\eta_2) \qquad (2)$$

where η_{12} is the viscosity of the mixture and x_1 and x_2 and η_1 and η_2 are the mole fraction and the viscosity of pure components 1 and 2, respectively.

Table 2. Density (ρ), Excess Volume (V^E), and Viscosity (η) for the Binary Liquid Mixtures

x_1	$\rho\cdot10^{-3}$ / $\mathrm{kg\cdot m^{-3}}$	$V^E\cdot10^6$ / $\mathrm{m^3\cdot mol^{-1}}$	η / $\mathrm{mPa\cdot s}$	$\rho\cdot10^{-3}$ / $\mathrm{kg\cdot m^{-3}}$	$V^E\cdot10^6$ / $\mathrm{m^3\cdot mol^{-1}}$	η / $\mathrm{mPa\cdot s}$	$\rho\cdot10^{-3}$ / $\mathrm{kg\cdot m^{-3}}$	$V^E\cdot10^6$ / $\mathrm{m^3\cdot mol^{-1}}$	η / $\mathrm{mPa\cdot s}$
Propyl Formate (1) + Benzene (2)									
		T/K = 303.15			T/K = 308.15			T/K = 313.15	
0.0000	0.8676		0.569	0.8632		0.534	0.8589		0.497
0.0918	0.8696	0.061	0.540	0.8651	0.067	0.512	0.8606	0.085	0.479
0.1874	0.8714	0.150	0.518	0.8668	0.162	0.490	0.8623	0.178	0.462
0.2822	0.8736	0.185	0.504	0.8689	0.208	0.476	0.8643	0.227	0.448
0.3785	0.8758	0.230	0.490	0.8711	0.244	0.466	0.8665	0.258	0.437
0.4767	0.8782	0.247	0.480	0.8734	0.270	0.456	0.8687	0.289	0.431
0.5772	0.8806	0.274	0.474	0.8758	0.286	0.449	0.8711	0.300	0.425
0.6802	0.8834	0.257	0.471	0.8785	0.274	0.451	0.8737	0.286	0.426
0.7854	0.8862	0.232	0.469	0.8813	0.246	0.448	0.8765	0.265	0.423
0.8924	0.8894	0.168	0.467	0.8844	0.187	0.446	0.8795	0.206	0.421
1.0000	0.8935		0.477	0.8886		0.456	0.8838		0.431
Propyl Formate (1) + Isopropyl Benzene (2)									
		T/K = 303.15			T/K = 308.15			T/K = 313.15	
0.0000	0.8527		0.690	0.8490		0.651	0.8454		0.611
0.1349	0.8560	0.109	0.652	0.8521	0.128	0.617	0.8482	0.159	0.580
0.2617	0.8595	0.197	0.617	0.8554	0.225	0.585	0.8514	0.253	0.551
0.3812	0.8633	0.244	0.585	0.8591	0.268	0.557	0.8550	0.292	0.526
0.4870	0.8671	0.261	0.559	0.8627	0.296	0.533	0.8585	0.318	0.504
0.5882	0.8710	0.275	0.537	0.8665	0.310	0.513	0.8622	0.328	0.485
0.6817	0.8751	0.265	0.517	0.8704	0.306	0.495	0.8660	0.321	0.468
0.7675	0.8792	0.243	0.506	0.8744	0.280	0.485	0.8698	0.304	0.459
0.8499	0.8835	0.211	0.494	0.8786	0.243	0.474	0.8739	0.264	0.448
0.9277	0.8881	0.150	0.484	0.8830	0.189	0.464	0.8782	0.206	0.439
1.0000	0.8935		0.477	0.8886		0.456	0.8838		0.431
Propyl Formate (1) + Isobutyl Benzene (2)									
		T/K = 303.15			T/K = 308.15			T/K = 313.15	
0.0000	0.8444		0.905	0.8411		0.848	0.8376		0.784
0.1508	0.8488	0.092	0.813	0.8452	0.111	0.771	0.8414	0.143	0.718
0.2870	0.8533	0.159	0.743	0.8495	0.182	0.706	0.8456	0.211	0.661
0.4072	0.8579	0.178	0.687	0.8540	0.210	0.654	0.8500	0.229	0.614
0.5190	0.8629	0.182	0.640	0.8587	0.217	0.610	0.8545	0.250	0.575
0.6161	0.8677	0.168	0.602	0.8634	0.201	0.575	0.8591	0.227	0.543
0.7080	0.8729	0.132	0.569	0.8685	0.154	0.544	0.8640	0.188	0.515
0.7895	0.8780	0.094	0.540	0.8734	0.120	0.519	0.8688	0.147	0.492
0.8656	0.8832	0.057	0.516	0.8784	0.086	0.497	0.8737	0.107	0.471
0.9341	0.8883	0.021	0.495	0.8834	0.041	0.476	0.8786	0.058	0.451
1.0000	0.8935		0.477	0.8886		0.456	0.8838		0.431
Propyl Formate (1) + Butylbenzene (2)									
		T/K = 303.15			T/K = 308.15			T/K = 313.15	
0.0000	0.8513		0.901	0.8480		0.844	0.8446		0.787
0.1483	0.8547	0.134	0.823	0.8510	0.168	0.772	0.8473	0.208	0.723
0.2855	0.8584	0.213	0.753	0.8546	0.252	0.710	0.8507	0.289	0.667
0.4066	0.8622	0.280	0.696	0.8582	0.305	0.658	0.8543	0.333	0.619
0.5164	0.8662	0.292	0.649	0.8621	0.323	0.616	0.8580	0.351	0.580
0.6148	0.8703	0.296	0.609	0.8660	0.329	0.579	0.8618	0.348	0.546
0.7042	0.8745	0.268	0.575	0.8701	0.298	0.547	0.8658	0.315	0.517
0.7884	0.8791	0.223	0.545	0.8745	0.251	0.520	0.8699	0.279	0.491
0.8642	0.8838	0.144	0.519	0.8790	0.180	0.497	0.8742	0.214	0.470
0.9339	0.8886	0.074	0.498	0.8837	0.096	0.476	0.8787	0.138	0.451
1.0000	0.8935		0.477	0.8886		0.456	0.8838		0.431

Table 3. Standard Deviation (σ) and Values of the Constant from the Redlich−Kister Equation 4

system	functions	T/K	A_0	A_1	A_2	σ (V^E)
propyl formate (1) + benzene (2)	V^E	303.15	1.0369	0.5137	0.3530	0.014
		308.15	1.1033	0.5514	0.4477	0.018
		313.15	1.1511	0.5528	0.6764	0.018
	$\Delta\eta$	303.15	−0.1696	0.0282	−0.0795	0.002
		308.15	−0.1578	0.0111	−0.0391	0.003
		313.15	−0.1365	−0.0031	−0.0287	0.003
	ΔG^{*E}	303.15	−776.80	65.25	−346.80	10.00
		308.15	−777.35	0.56	−170.69	14.78
		313.15	−724.92	−65.17	−132.17	16.67
propyl formate (1) + isopropyl benzene (2)	V^E	303.15	1.0060	0.5237	0.7963	0.021
		308.15	1.1026	0.6488	1.1591	0.030
		313.15	1.1720	0.5973	1.4488	0.031
	$\Delta\eta$	303.15	−0.1045	−0.0360	−0.0119	0.001
		308.15	−0.0921	−0.0098	0.0269	0.001
		313.15	−0.0674	−0.0059	0.0136	0.001
	ΔG^{*E}	303.15	−127.03	−149.55	65.10	1.73
		308.15	−86.76	−93.11	134.74	1.80
		313.15	−55.66	−76.87	140.73	2.62
propyl formate (1) + isobutyl benzene (2)	V^E	303.15	0.7502	−0.1635	−0.3080	0.004
		308.15	0.8572	−0.1102	−0.1253	0.005
		313.15	0.9585	−0.1355	0.1188	0.007
	$\Delta\eta$	303.15	−0.1792	0.0359	−0.0401	0.002
		308.15	−0.1476	0.0300	0.0476	0.001
		313.15	−0.1154	0.0265	0.0602	0.001
	ΔG^{*E}	303.15	162.12	71.03	−108.67	1.91
		308.15	218.46	95.60	192.04	2.76
		313.15	295.32	131.27	272.74	4.24
propyl formate (1) + butylbenzene (2)	V^E	303.15	1.2062	0.1729	−0.1504	0.010
		308.15	1.3120	0.1601	0.1933	0.007
		313.15	1.3521	0.1549	0.8454	0.010
	$\Delta\eta$	303.15	−0.1344	−0.0018	0.0152	0.001
		308.15	−0.1164	0.0086	0.0246	0.001
		313.15	−0.0976	0.0054	0.0343	0.001
	ΔG^{*E}	303.15	295.47	−2.50	55.18	1.95
		308.15	308.57	54.25	134.72	4.47
		313.15	340.53	61.81	209.94	6.31

The excess Gibbs energy of activation ΔG^{*E} of viscous flow was obtained using the following relation.

$$\Delta G^{*E}\,(\mathrm{J\cdot mol^{-1}}) = RT[\ln(\eta_{12}\nu_{12}) - (x_1 \ln(\eta_1\nu_1) + x_2 \ln(\eta_2\nu_2))] \qquad (3)$$

where ν_{12} is the molar volume of the mixture and ν_1 and ν_2 are the molar volume of the pure components 1 and 2, respectively. The $\Delta\eta$ and ΔG^{*E} data are graphically represented in Figures 2 and 3. The excess molar volumes, deviation in viscosities, and excess Gibbs energy of activation were fitted to the Redlich−Kister[7] type equation.

$$\Delta Y = x_1 x_2 \sum_{i=0}^{k} A_i (x_1 - x_2)^{i-1} \qquad (4)$$

where k is the number of estimated parameter and A_i, the polynomial coefficient, was obtained by the least-squares regression method. ΔY is V^E, $\Delta\eta$, or ΔG^{*E}, and i is the degree of the polynomial. The standard deviation σ between the fit quantities of eq 4 and the computed quantities of eqs 1 to 3 were calculated as the following.

$$\sigma(Y^E) = \left[\sum (Y^E_{\mathrm{expt}} - Y^E_{\mathrm{calc}})^2/(N - n)\right]^{1/2} \qquad (5)$$

where N represents the number of data points and n is the number of coefficients. The calculated values of the coefficients A_i along with the standard deviation σ are given in Table 3.

Further, V^E results were also fitted to the semiempirical equation proposed by Hwang et al.[8]

$$V^E\cdot 10^6/(\mathrm{m^3\cdot mol^{-1}}) = x_1 x_2[b_0 + b_1 x_1^{\,3} + b_2 x_2^{\,3}] \qquad (6)$$

On the analysis of standard deviation values, it is observed that the equation of Hwang et al.[8] also represents precisely the V^E data.

The plots of V^E versus x_1 for the studied binary mixtures at T = 303.15 K are displayed in Figure 1, while at T = (308.15 and 313.15) K the V^E values more or less follow the same trend as displayed in Figure 1. For all of the mixtures, the V^E values are positive. The positive values of V^E increase with the increase in temperature. The magnitude of V^E increases in the following order: butyl benzene > isopropyl benzene > benzene > isobutyl benzene. The positive V^E values indicate that the interactions between the molecules of two mixed components are weaker than in the pure component. This is due to the fact that a nonpolar aromatic hydrocarbon such as benzene, isopropyl benzene, isobutyl benzene, or butylbenzene is added to a polar solvent of propyl formate and the molecules of aromatic hydrocarbon scatter among the ester molecules, thereby reducing the interaction among the ester molecules. The results of $\Delta\eta$ versus x_1 are displayed in Figure 2, wherein we observe that the $\Delta\eta$ values are negative in all of the cases. The magnitude of negative deviation decrease with the rise of temperature from (303.15 to 313.15) K. An inspection of our observed results reveals that the viscosity values of propyl formate + benzene at (303.15 and 313.15) K agreed well with Emmerling et al.; it is found that there is a close agreement between the two sets of

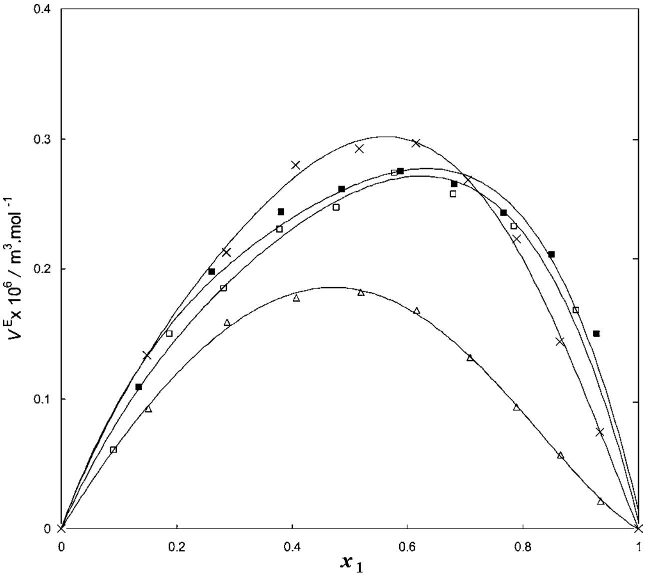

Figure 1. Excess volume V^E vs mole fraction at 303.15 K for propyl formate with □, benzene; ■, isopropyl benzene; △, isobutyl benzene; and ×, butylbenzene.

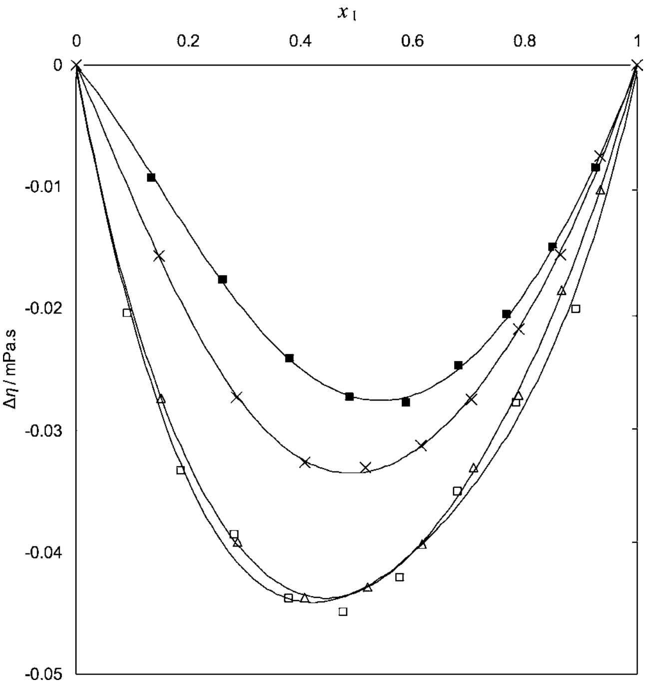

Figure 2. Deviation in viscosity $\Delta\eta$ vs mole fraction at 303.15 K for propyl formate with □, benzene; ■, isopropyl benzene; △, isobutyl benzene; and ×, butylbenzene.

observations. These negative $\Delta\eta$ values suggest that the interactions between unlike molecules are weaker. Figure 3 depicts the variation of ΔG^{*E} versus x_1. It is observed that the ΔG^{*E} values for propyl formate + benzene are negative, while for propyl formate + isobutyl benzene and + butyl benzene they are positive over the entire range of composition at all of the studied temperatures. However, for propyl formate + isopropyl benzene, ΔG^{*E} values show positive deviations at lower compositions ($x_1 < 0.35$) of ester, and as the composition of ester increases ($x_1 > 0.35$) these ΔG^{*E} values become negative. The effect of temperature on ΔG^{*E} is also significant, as the values of ΔG^{*E} increase with the increase in temperature for all of the systems. The maximum and minimum in ΔG^{*E} curves give an indication of the extent of interaction between the component molecules in the mixtures. The positive trend in ΔG^{*E} indicates that the interaction strength between two component molecules is weaker.

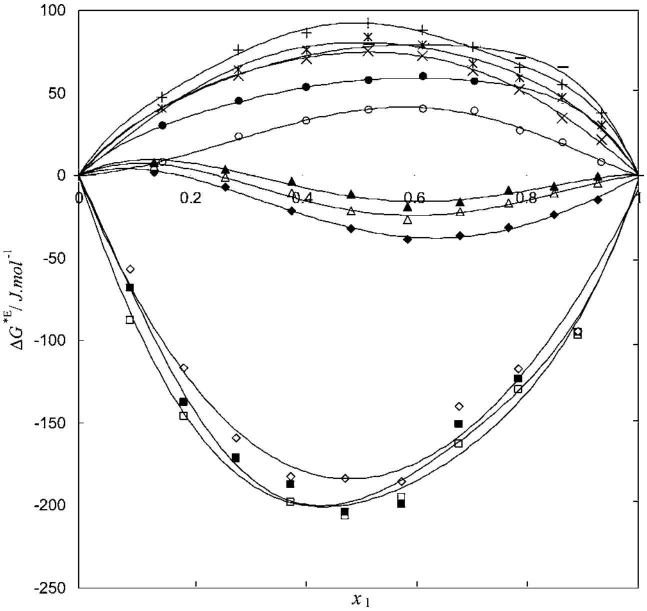

Figure 3. Deviation in excess Gibbs free energy ΔG^{*E} vs mole fraction for the binary mixtures of propyl formate + benzene at (□, 303.15; ■, 308.15; ◇, 313.15) K, propyl formate + isopropylbenzene at (♦, 303.15; △, 308.15; ▲, 313.15) K, propyl formate + isobutylbenzene at (○, 303.15; ●, 308.15; −, 313.15) K, and propyl formate + butylbenzene at (×, 303.15; ∗, 308.15; +, 313.15) K.

Viscosity Models and Interaction Parameters

The viscosities of binary mixtures have been correlated with some of the empirical equations available in literature. Tamura and Kurata[9] proposed the following equation for the viscosity of binary liquid mixtures.

$$\eta = x_1\Phi_1\eta_1 + x_2\Phi_2\eta_2 + 2(x_1x_2\Phi_1\Phi_2)^{0.5}C \qquad (7)$$

where C is the interaction parameter and Φ_1 and Φ_2 are the volume fractions of pure components 1 and 2, respectively.

Molecular interactions may also be interpreted by the following two-parameter model of Heric[10]

$$\ln(\eta) = x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + x_1 \ln(M_1) + x_2 \ln(M_2) - \ln(x_1M_1 + x_2M_2) + x_1x_2[\gamma_{12} + \gamma_{21}(x_1 - x_2)] \qquad (8)$$

where γ_{12} and γ_{21} are the interaction parameters.

Auslander[11] has proposed the three-parameter equation for the viscosity of binary liquid mixtures as the following.

$$\eta = \frac{\eta_1x_1(x_1 + B_{12}x_2) + \eta_2[A_{21}x_2(B_{21}x_1 + x_2)]}{x_1(x_1 + B_{12}x_2) + A_{21}x_2(B_{21}x_1 + x_2)} \qquad (9)$$

where η, η_1, and η_2 are the dynamic viscosities of mixture and pure components, respectively. B_{12}, A_{21}, and B_{21} are the interaction parameters representing binary 1, 2 interactions. The correlating ability of eqs 7 to 9 was tested by calculating the

Table 4. Adjustable Parameters of Auslander Equation 9 and Percentage Standard Deviation in Correlating Viscosities of Binary Mixtures

		Auslander			
system	T/K	B_{12}	A_{21}	B_{21}	σ %
propyl formate (1) + benzene (2)	303.15	0.0032	0.3524	1.5941	0.17
	308.15	0.0024	0.0010	1.6320	0.21
	313.15	0.0048	0.3253	1.6324	0.22
propyl formate (1) + isopropyl benzene (2)	303.15	−0.0039	0.4175	1.4832	0.03
	308.15	−0.0004	0.0010	1.5342	0.05
	313.15	−0.0020	0.3842	1.5489	0.06
propyl formate (1) + isobutyl benzene (2)	303.15	0.0033	0.4645	1.3841	0.03
	308.15	0.0086	0.4445	1.4409	0.11
	313.15	0.0105	0.4240	1.4934	0.14
propyl formate (1) + butylbenzene (2)	303.15	0.0025	0.4710	1.3930	0.03
	308.15	0.0022	0.4539	1.4237	0.07
	313.15	0.0023	0.4774	1.4430	0.09

percentage standard deviations (σ %) between the experimental and the calculated viscosity as the following.

$$\sigma\,(\%) = \left[1/(N-n)\sum\{(100(\eta_{\mathrm{exptl}}-\eta_{\mathrm{calc}})/(\eta_{\mathrm{exptl}})^2\}^{1/2}\right] \tag{10}$$

where N represents the number of data points and n is the number of adjustable parameters. The values of interaction parameters along with their percentage standard deviation of eq 9 are listed in Table 4.

Inspection of observed results reveals that the values of percentage standard deviation with a single parameter of Tamura−Kurata are in the range from (0.06 to 0.31) % and from (0.04 to 0.20) % for the two-parameter model of Heric and from (0.03 to 0.22) % for the three-parameter model of Auslander. The average values of percentage standard deviation are (0.20, 0.11, and 0.11) % respectively for eqs 7 to 9. Further, it was observed that the correlating values with the two-parameter equation of Heric reduce the percentage standard deviation values below that of single-parameter equation of Tamura−Kurata. On the basis of our present analysis it may be concluded that the correlating ability satisfactorily improves for the studied mixtures as the number of adjustable parameters are increased. In all of the studied systems, the three-parameter equation proposed by Auslander gives a very good correlation at 303.15 K. However, at higher temperatures in some of the studied systems the two-parameter equation of Heric gave an almost similar correlation as that of Auslander.

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Supporting Information Available:

Values of constants obtained from Hwang et al. of eq 6 and the parameters for correlating viscosity models of Tamura-Kurata and Heric of eqs 7 and 8 for all the studied binary mixtures at T = (303.15, 308.15, and 313.15) K. This material is available free of charge via the Internet at http://pubs.acs.org.

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