Densities and Kinematic Viscosities for the Systems Benzene + Methyl Formate, Benzene + Ethyl Formate, Benzene + Propyl Formate, and Benzene + Butyl Formate

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Densities and kinematic viscosities have been measured for the system benzene + methyl formate at 20 °C and for the systems benzene + ethyl formate, benzene + propyl formate, and benzene + butyl formate from 20 °C to 50 °C. The results for the system benzene + methyl formate have been correlated using a Redlich−Kister type of expression with temperature-independent parameters and the data for the systems benzene + ethyl formate, benzene + propyl formate, and benzene + butyl formate with temperature-dependent parameters. The viscosities have furthermore been compared to values predicted by means of the GC−UNIMOD model.

Introduction

Densities and viscosities are important properties needed for the optimal design of many types of equipment in chemical technology. In this work densities and kinematic viscosities have been measured for mixtures of benzene and formates. The systems are benzene + methyl formate, benzene + ethyl formate, benzene + propyl formate, and benzene + butyl formate. Viscosities for these mixtures have to our knowledge not been published previously.

Experimental Section

Materials. All compounds were obtained from Merck. A check with a gas chromatograph confirms the guaranteed purity lists in Table 1. The refractive index, the density and the viscosity of the pure substances were also measured and compared with literature data as shown in Tables 1 and 2. All measured values show good agreement with data given in the literature. The compounds were used without further purification.

Apparatus and Experimental Procedures. The densities ρ were measured using a Densimeter DA-200 of KYOTO ELECTRONICS with an accuracy of 10^{-4} g cm^{-3}. With the help of a capillary viscometer of the Ubbelohde type, the kinematic viscosities ν were determined. The temperature T was controlled to within ±0.05 °C during the measurements using a thermostat. From the experimental density and the experimental kinematic viscosity, the dynamic viscosity η was calculated. The refractive index n_D was measured with an ABBE refractometer with a resolution ±0.0001. The composition of the liquid phase was fixed gravimetrically with an accuracy of the mole fraction x of ±0.001.

Experimental Results. The experimental results are presented in Tables 3−6. The system benzene + methyl formate was measured only at 20 °C owing to the high volatility of methyl formate.

Data Correlation

Density. The densities ρ_{12}/g cm^{-3} of the binary mixtures benzene (1) + alkyl formate (2) were correlated using eq 1, which gave a good fit of the measured data.

$$\rho_{12} = x_1\rho_{01}(T)/\text{g cm}^{-3} + x_2\rho_{02}(T)/\text{g cm}^{-3} + x_1x_2\,[a^{(0)} + a^{(1)}T/\text{K} + a^{(2)}(T/\text{K})^2 + (b^{(0)} + b^{(1)}T/\text{K} + b^{(2)}(T/\text{K})^2)(x_1 - x_2) + (c^{(0)} + c^{(1)}T/\text{K} + c^{(2)}(T/\text{K})^2)(x_1 - x_2)^2] \quad (1)$$

The temperature dependence of the densities ρ_{0i} (T) of the pure substances i is presented using eq 2.

$$\rho_{0i}/\text{g cm}^{-3} = a_i + b_iT/\text{K} + c_i\,(T/\text{K})^2 \quad (i = 1, 2) \quad (2)$$

$a^{(0)}$, $a^{(1)}$, $a^{(2)}$, $b^{(0)}$, $b^{(1)}$, $b^{(2)}$, $c^{(0)}$, $c^{(1)}$, $c^{(2)}$ and a_i, b_i, c_i are fitted parameters. The estimated values and the deviations between experimental and calculated data for the systems benzene + ethyl formate, benzene + propyl formate, and benzene + butyl formate are given in Table 7. The deviations between experimental (exptl) and calculated (calcd) values are shown as the mean absolute deviation

$$s_{MA} = \sum_{n=1}^{N} \frac{|X_n^{exptl} - X_n^{calcd}|}{N}$$

the mean relative deviation

$$s_{MR} = \frac{100}{N} \sum_{n=1}^{N} \frac{|X_n^{exptl} - X_n^{calcd}|}{X^{exptl}}$$

and the standard deviation

$$s_S = \left(\sum_{n=1}^{N} \frac{(X_n^{exptl} - X_n^{calcd})^2}{N - p} \right)^{1/2}$$

where N is the number of experimental points, p is the number of parameters, and X is the density or the

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Table 1. Densities ρ and Refractive Indices n_D of Pure Components at 293.15 K

	ρ_{0i}/g cm^{-3}		n_D^{20}	
component/minimum purity	present work	lit.	present work	lit.
benzene, >99%	0.8789	0.8787 (Steere, 1967) 0.8789 (Reid et al., 1987) 0.8800 (Budavari, 1989)	1.5012	1.499 50 (Budavari, 1989)
methyl formate, ~97%	0.9713	0.9700 (Budavari, 1989) 0.9740 (Reid et al., 1987)	1.3419	1.340 00 (Budavari, 1989)
ethyl formate, >98%	0.9208	0.9170 (Steere, 1967) 0.9190 (Budavari, 1989) 0.9201 (Ohta and Nagata, 1980; Ohta et al., 1983) 0.9208 (Reid et al., 1987)	1.3609	1.357 50 (Budavari, 1989) 1.359 99 (Ohta and Nagata, 1980)
propyl formate, >98%	0.9073	0.9006 (Steere, 1967) 0.9071 (Budavari, 1989) 0.9073 (Reid et al., 1987)	1.377	1.375 30 (Budavari, 1989)
butyl formate, >98%	0.8958	0.9110 (Steere, 1967) 0.8940 (Budavari, 1989)	1.3887	1.386 80 (Budavari, 1989)

Table 2. Kinematic ν and Dynamic η Viscosities of Pure Components at 293.15 K

	$\nu_{0i}/10^{-6}$ m^2/s		η_{0i}/mPa s	
component	present work	lit.	present work	lit.
benzene	0.7408	0.7427 (Dreissbach, 1955)	0.6511	0.6502 (Viswanath and Natrajan, 1989 0.6520 (Landolt-Boernstein, 1969)
methyl formate	0.3611	0.3560 (Reid et al., 1987)	0.3507	0.3467[a] (Landolt-Boernstein, 1969)
ethyl formate	0.4432	0.4475 (Steere, 1967)	0.4087	0.4080 (Landolt-Boernstein, 1969)
propyl formate	0.5978	0.5830 (Steere, 1967)	0.5424	0.5250 (Landolt-Boernstein, 1969)
butyl formate	0.7414	0.7570 (Landolt-Boernstein, 1969)	0.6643	0.6897 (Viswanath and Natrajan, 1989)

[a] At 20.15 °C.

Table 3. Experimental Densities ρ and Kinematic Viscosities ν for the System Benzene + Methyl Formate

T/K	x_1	ρ_{12}/g cm^{-3}	$\nu_{12}/10^{-6}$ m^2/s
293.15	0.0000	0.9713	0.3611
	0.1003	0.9554	0.3884
	0.2012	0.9420	0.4083
	0.3045	0.9296	0.4389
	0.4024	0.9197	0.4665
	0.4996	0.9114	0.4961
	0.5994	0.9032	0.5316
	0.7016	0.8957	0.5744
	0.8039	0.8894	0.6156
	0.9050	0.8840	0.6797
	1.0000	0.8789	0.7408
291.15	0.0000	0.9833	0.3658
298.15	0.0000	0.9634	0.3458

kinematic viscosity. The data for the system benzene + methyl formate were fitted by using eq 1 with temperature-independent parameters. Parameters and deviations are $a_0 = -0.056\,142$, $b_0 = 0.016\,497$, $c_0 = -0.004\,713$ and $s_{MA} = 0.0003$, $s_{MR} = 0.14$; $s_S = 0.0002$. Figure 1 shows experimental and calculated values at 293.15 K for all four systems.

Kinematic Viscosities. Equation 3 represents a good approximation for the kinematic viscosity $\nu_{12}/10^{-6}$ m^2/s of a binary mixture benzene (1) + alkyl formate (2) (Schuberth, 1987).

$$\ln \nu_{12}^{id} = x_1 \ln \nu_{01} + x_2 \ln \nu_{02} \qquad (3)$$

For the description of the excess behavior additional terms are added to eq 3 as shown in eq 4.

$$\ln \nu_{12} = x_1 \ln \nu_{01}(T) + x_2 \ln \nu_{02}(T) + x_1 x_2 [A^{(0)} + A^{(1)}/T/\mathrm{K} + A^{(2)}/(T/\mathrm{K})^2 + (B^{(0)} + B^{(1)}/T/\mathrm{K} + B^{(2)}/(T/\mathrm{K})^2)(x_1 - x_2) + (C^{(0)} + C^{(1)}/T/\mathrm{K} + C^{(2)}/(T/\mathrm{K})^2)(x_1 - x_2)^2] \qquad (4)$$

Table 4. Experimental Densities ρ and Kinematic Viscosities ν for the System Benzene + Ethyl Formate

T/K	x_1	ρ_{12}/g cm^{-3}	$\nu_{12}/10^{-6}$ m^2/s	T/K	x_1	ρ_{12}/g cm^{-3}	$\nu_{12}/10^{-6}$ m^2/s
293.15	0.0000	0.9208	0.4432	293.15	0.5934	0.8921	0.5578
	0.1072	0.9146	0.4579		0.7152	0.8876	0.5991
	0.2038	0.9092	0.4708		0.8098	0.8846	0.6380
	0.3027	0.9045	0.4888		0.9085	0.8816	0.6855
	0.4309	0.8987	0.5152		1.0000	0.8789	0.7408
	0.4998	0.8958	0.5310				
303.15	0.0000	0.9076	0.4087	303.15	0.5934	0.8805	0.5016
	0.1072	0.9018	0.4190		0.7152	0.8763	0.5350
	0.2038	0.8970	0.4308		0.8098	0.8733	0.5659
	0.3027	0.8921	0.4448		0.9085	0.8707	0.6046
	0.4309	0.8879	0.4664		1.0000	0.8683	0.6497
	0.4998	0.8839	0.4810				
313.15	0.0000	0.8936	0.3790	313.15	0.5934	0.8692	0.4549
	0.1072	0.8886	0.3886		0.7152	0.8654	0.4841
	0.2038	0.8842	0.3965		0.8098	0.8623	0.5098
	0.3027	0.8798	0.4073		0.9085	0.8599	0.5424
	0.4309	0.8753	0.4248		1.0000	0.8581	0.5774
	0.4998	0.8726	0.4357				
323.15	0.0000	0.8839	0.3523	323.15	0.5934	0.8616	0.4148
	0.1072	0.8792	0.3626		0.7152	0.8574	0.4410
	0.2038	0.8757	0.3704		0.8098	0.8549	0.4642
	0.3027	0.8720	0.3779		0.9085	0.8527	0.4923
	0.4309	0.8676	0.3909		1.0000	0.8501	0.5202
	0.4998	0.8646	0.4010				

The temperature dependence of kinematic viscosities of the pure substances $\nu_{0i}(T)$ is presented using eq 5.

$$\ln \nu_{0i} = A_i + \frac{B_i}{T/\mathrm{K}} + \frac{C_i}{(T/\mathrm{K})^2} \quad (i = 1, 2) \qquad (5)$$

$A^{(0)}$, $A^{(1)}$, $A^{(2)}$, $B^{(0)}$, $B^{(1)}$, $B^{(2)}$, $C^{(0)}$, $C^{(1)}$, $C^{(2)}$ and A_i, B_i, C_i are fitted parameters. The parameters and deviations between experimental and calculated data for the systems benzene + ethyl formate, benzene + propyl formate, and benzene + butyl formate are listed in Table 8. The data for the

Table 5. Experimental Densities ρ and Kinematic Viscosities ν for the System Benzene + Propyl Formate

T/K	x_1	ρ_{12}/g cm^{-3}	$\nu_{12}/10^{-6}$ m^2/s	T/K	x_1	ρ_{12}/g cm^{-3}	$\nu_{12}/10^{-6}$ m^2/s
293.15	0.0000	0.9073	0.5978	293.15	0.5731	0.8895	0.6250
	0.0943	0.9044	0.5992		0.7017	0.8862	0.6428
	0.2009	0.9008	0.6016		0.8013	0.8836	0.6660
	0.2748	0.8987	0.6039		0.8981	0.8812	0.6993
	0.3877	0.8951	0.6065		1.0000	0.8789	0.7408
	0.4930	0.8918	0.6142				
303.15	0.0000	0.8958	0.5385	303.15	0.5731	0.8785	0.5615
	0.0943	0.8930	0.5385		0.7017	0.8751	0.5844
	0.2009	0.8894	0.5392		0.8013	0.8727	0.6056
	0.2748	0.8874	0.5413		0.8981	0.8703	0.6282
	0.3877	0.8839	0.5440		1.0000	0.8683	0.6497
	0.4930	0.8808	0.5515				
313.15	0.0000	0.8847	0.4887	313.15	0.5731	0.8678	0.5065
	0.0943	0.8814	0.4893		0.7017	0.8643	0.5286
	0.2009	0.8781	0.4898		0.8013	0.8619	0.5450
	0.2748	0.8762	0.4909		0.8981	0.8602	0.5626
	0.3877	0.8727	0.4948		1.0000	0.8581	0.5774
	0.4930	0.8699	0.4986				
323.15	0.0000	0.8755	0.4436	323.15	0.5731	0.8596	0.4619
	0.0943	0.8727	0.4445		0.7017	0.8565	0.4765
	0.2009	0.8698	0.4454		0.8013	0.8543	0.4888
	0.2748	0.8677	0.4467		0.8981	0.8520	0.5024
	0.3877	0.8645	0.4505		1.0000	0.8501	0.5202
	0.4930	0.8618	0.4548				

Table 6. Experimental Densities ρ and Kinematic Viscosities ν for the System Benzene + Butyl Formate

T/K	x_1	ρ_{12}/g cm^{-3}	$\nu_{12}/10^{-6}$ m^2/s	T/K	x_1	ρ_{12}/g cm^{-3}	$\nu_{12}/10^{-6}$ m^2/s
293.15	0.0000	0.8958	0.7414	293.15	0.5013	0.8861	0.6838
	0.1070	0.8936	0.7250		0.6521	0.8835	0.6814
	0.1857	0.8920	0.7140		0.7040	0.8827	0.6836
	0.3054	0.8897	0.6986		0.8079	0.8812	0.6911
	0.4084	0.8878	0.6900		0.9049	0.8799	0.7096
	0.4942	0.8864	0.6840		1.0000	0.8789	0.7408
303.15	0.0000	0.8848	0.6653	303.15	0.5013	0.8758	0.6114
	0.1070	0.8827	0.6506		0.6521	0.8730	0.6083
	0.1857	0.8814	0.6418		0.7040	0.8721	0.6090
	0.3054	0.8793	0.6279		0.8079	0.8704	0.6146
	0.4084	0.8776	0.6173		0.9049	0.8692	0.6279
	0.4942	0.8756	0.6120		1.0000	0.8683	0.6497
313.15	0.0000	0.8744	0.6007	313.15	0.5013	0.8656	0.5508
	0.1070	0.8728	0.5871		0.6521	0.8629	0.5469
	0.1857	0.8714	0.5774		0.7040	0.8619	0.5469
	0.3054	0.8691	0.5665		0.8079	0.8605	0.5501
	0.4084	0.8671	0.5568		0.9049	0.8591	0.5598
	0.4942	0.8659	0.5510		1.0000	0.8581	0.5774
323.15	0.0000	0.8661	0.5456	323.15	0.5013	0.8572	0.5018
	0.1070	0.8642	0.5306		0.6521	0.8548	0.4964
	0.1857	0.8625	0.5216		0.7040	0.8543	0.4961
	0.3054	0.8607	0.5110		0.8079	0.8526	0.4990
	0.4084	0.8588	0.5052		0.9049	0.8512	0.5052
	0.4942	0.8572	0.5010		1.0000	0.8501	0.5202

system benzene + methyl formate were fitted by using eq 4 with temperature-independent parameters. Parameters and deviations are $A_0 = -0.162\ 889\ 7$, $B_0 = -0.117\ 047\ 2$, $C_0 = 0.0$ and $s_{MA} = 0.019$, $s_{MR} = 0.36$, $s_S = 0.0027$. Figure 2 shows experimental values at 293.15 K of measured kinematic viscosities and their fitting curves of all four systems.

Prediction of Kinematic Viscosities. The kinematic mixture viscosity was predicted using the GC−UNIMOD method (Cao et al., 1993a,b). UNIFAC−VLE parameters were used for the prediction of kinematic and dynamic viscosities without any mixture viscosity information. The mean absolute deviations between experimental and predicted values are between 0.84% and 2.47% (system benzene + methyl formate, 2.14%; system benzene + ethyl formate at 20 °C, 2.47%; system benzene + propyl formate at 20 °C, 2.19%; system benzene + butyl formate at 20 °C, 2.29%). The calculations confirm that VLE and viscosity prediction can be done with one parameter set with reasonable accuracy.

Table 7. Parameters and Deviations for Densities for Pure Substances I and for the Binary Systems

Benzene		
$a_i = 1.777\ 93$	$b_i = -0.497\ 195\text{E}{-}06$	$c_i = 0.65\text{E}{-}05$
$s_{MA} = 0.0002$	$s_{MR} = 0.021$	$s_S = 0.004$
Ethyl Formate		
$a_i = 2.115\ 513$	$b_i = -6.639\ 625\text{E}{-}03$	$c_i = 8.75\text{E}{-}06$
$s_{MA} = 0.0005$	$s_{MR} = 0.057$	$s_S = 0.001$
Propyl Formate		
$a_i = 1.764\ 285\ 4$	$b_i = -4.608\ 725\text{E}{-}03$	$c_i = 5.75\text{E}{-}06$
$s_{MA} = 0.0002$	$s_{MR} = 0.017$	$s_S = 0.003$
Butyl Formate		
$a_i = 1.803\ 310$	$b_i = -0.500\ 960\text{E}{-}02$	$c_i = 0.65\text{E}{-}02$
$s_{MA} = 0.0003$	$s_{MR} = 0.017$	$s_S = 0.003$
Benzene + Ethyl Formate		
$a^{(0)} = 5.4517\text{E}{-}01$[a]	$b^{(0)} = 5.7983\text{E}{-}01$	$c^{(0)} = 1.4138$
$a^{(1)} = -3.8709\text{E}{-}03$	$b^{(1)} = -3.6183\text{E}{-}03$	$c^{(1)} = -9.1769\text{E}{-}03$
$a^{(2)} = 6.6720\text{E}{-}06$	$b^{(2)} = 5.6255\text{E}{-}06$	$c^{(2)} = 1.4848\text{E}{-}05$
$s_{MA} = 0.0004$	$s_{MR} = 0.046$	$s_s = 0.0006$
Benzene + Propyl Formate		
$a^{(0)} = 4.5570\text{E}{-}01$	$b^{(0)} = 2.3520\text{E}{-}02$	$c^{(0)} = 1.1110$
$a^{(1)} = -3.0119\text{E}{-}03$	$b^{(1)} = -1.8031\text{E}{-}04$	$c^{(1)} = -7.1272\text{E}{-}03$
$a^{(2)} = 4.9071\text{E}{-}06$	$b^{(2)} = 3.1975\text{E}{-}07$	$c^{(2)} = 1.1411\text{E}{-}05$
$s_{MA} = 0.0002$	$s_{MR} = 0.029$	$s_s = 0.0003$
Benzene + Butyl Formate		
$a^{(0)} = -5.2498\text{E}{-}01$	$b^{(0)} = 1.3442$	$c^{(0)} = 3.3400\text{E}{-}01$
$a^{(1)} = 3.3427\text{E}{-}03$	$b^{(1)} = -8.7856\text{E}{-}03$	$c^{(1)} = -2.2564\text{E}{-}03$
$a^{(2)} = -5.3488\text{E}{-}06$	$b^{(2)} = 1.4313\text{E}{-}05$	$c^{(2)} = 3.7805\text{E}{-}06$
$s_{MA} = 0.0002$	$s_{MR} = 0.0179$	$s_s = 0.0002$

[a] Read as 5.4517×10^{-1}.

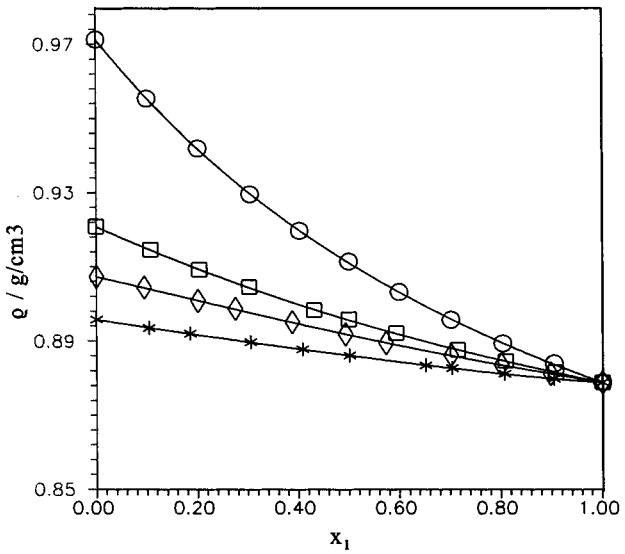

Figure 1. Values for experimental densities ρ and fitting lines at 20 °C for the systems ○, benzene + methyl formate; □, benzene + ethyl formate; ◇, benzene + propyl formate; *, benzene + butyl formate.

Conclusion

New experimental values for mixture viscosities and densities for the systems benzene + methyl formate, benzene + ethyl formate, benzene + propyl formate, and benzene + butyl formate were measured and the results were correlated. Kinematic mixture viscosities were predicted by the GC−UNIMOD method, and the results were presented and compared with experimental data. The

Table 8. Parameters and Deviations for Kinematic Viscosities for Pure Substances *i* and for the Binary Systems

	Benzene	
$A_i = -1.409\ 655$	$B_i = -547.468\ 26$	$C_i = 255\ 854.43$
$s_{MA} = 0.0001$	$s_{MR} = 0.023$	$s_S = 0.0003$
	Ethyl Formate	
$A_i = -3.697\ 604$	$B_i = 979.3143$	$C_i = -39\ 263.87$
$s_{MA} = 0.0001$	$s_{MR} = 0.029$	$s_S = 0.0003$
	Propyl Formate	
$A_i = -4.831\ 343$	$B_i = 1624.454$	$C_i = -105\ 252.28$
$s_{MA} = 0.0004$	$s_{MR} = 0.071$	$s_S = 0.0008$
	Butyl Formate	
$A_i = -3.888\ 557$	$B_i = 1144.5855$	$C_i = -27\ 077.378$
$s_{MA} = 0.0000$	$s_{MR} = 0.003$	$s_S = 0.0000$
	Benzene + Ethyl Formate	
$A^{(0)} = -2.854\ 173\ 7$	$B^{(0)} = -7.482\ 773\ 5$	$C^{(0)} = 3.718\ 059\ 7E{+}01$
$A^{(1)} = 1.670\ 228\ 5E{+}03$[a]	$B^{(1)} = 4.610\ 087\ 9E{+}03$	$C^{(1)} = -2.238\ 464\ 5E{+}04$
$A^{(2)} = -2.699\ 694\ 8E{+}05$	$B^{(2)} = -7.139\ 749\ 9E{+}05$	$C^{(2)} = 3.366\ 297\ 9E{+}06$
$s_{MA} = 0.0004$	$s_{MR} = 0.08$	$s_s = 0.0006$
	Benzene + Propyl Formate	
$A^{(0)} = 7.848\ 582\ 4E{-}03$	$B^{(0)} = -4.314\ 363\ 8E{+}01$	$C^{(0)} = -6.240\ 965\ 6E{+}01$
$A^{(1)} = 1.711\ 298\ 3E{+}02$	$B^{(1)} = 2.688\ 954\ 8E{+}04$	$C^{(1)} = 3.866\ 355\ 6E{+}04$
$A^{(2)} = -7.770\ 418\ 4E{+}04$	$B^{(2)} = -4.186\ 298\ 3E{+}06$	$C^{(2)} = -5.970\ 384\ 5E{+}06$
$s_{MA} = 0.0007$	$s_{MR} = 0.124$	$s_s = 0.001$
	Benzene + Butyl Formate	
$A^{(0)} = -2.172\ 319\ 9$	$B^{(0)} = 6.809\ 825$	$C^{(0)} = -2.378\ 975\ 1E{+}01$
$A^{(1)} = 1.400\ 420\ 7E{+}03$	$B^{(1)} = -4.036\ 261\ 4E{+}03$	$C^{(1)} = 1.448\ 840\ 7E{+}04$
$A^{(2)} = -2.514\ 315\ 5E{+}05$	$B^{(2)} = 5.850\ 159\ 5E{+}05$	$C^{(2)} = -2.208\ 724\ 35E{+}06$
$s_{MA} = 0.0004$	$s_{MR} = 0.073$	$s_s = 0.0006$

[a] Read as $1.670\ 228\ 5 \times 10^3$.

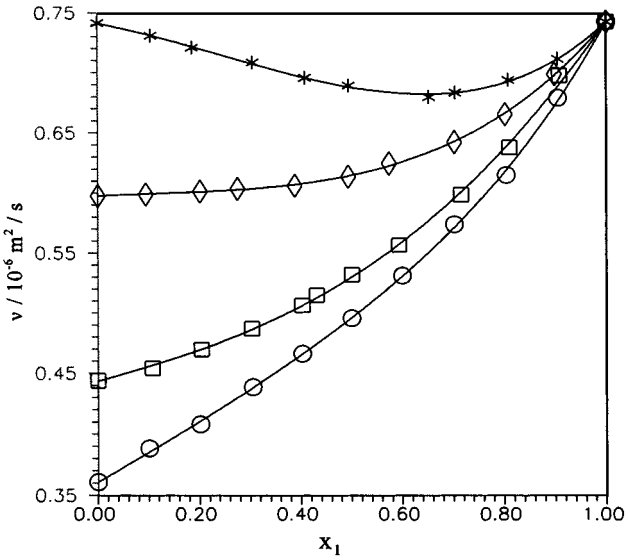

Figure 2. Experimental kinematic viscosity values ν and fitting lines at 20 °C for the systems ○, benzene + methyl formate; □, benzene + ethyl formate; ◇, benzene + propyl formate; *, benzene + butyl formate.

mean absolute deviation between experimental and predicted data are within 0.84 to 2.47%. These results confirm the good prediction capability of GC−UNIMOD for mixture viscosities.

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