

Figure 2. Experimental solute mole fraction vs. reduced reciprocal temperatures as compared with ideal predictions for the system *n*-butane-benzene and *n*-butane-cyclohexane. The heavy lines are the smoothed data presented in Tables III and IV, while the light lines are the ideal solubility loci for both systems.

smoothed and raw data, as well as the number of cell loadings and data points taken.

The raw data and the smoothed composition lines for the first four systems are presented in Figures 1 and 2. The straight lines on these semilogarithmic plots are "ideal" solubility lines. At high values of $1/T^*$, the data start to become linear in these coordinates, a characteristic observed in our earlier studies and interpreted as the region in which Henry's law for dilute solutions might be expected to be valid. It is felt that linear extrapolation of these data in these semilogarithmic plots would be reasonably accurate. All raw data are available as supplementary material. (See paragraph at end of text regarding supplementary material.)

No plot is presented of the data for the systems *n*-butane-*n*-decane and *n*-butane-*n*-dodecane, as the compositions of these two systems are quite close to those which would be predicted by "ideal" solubility.

All of the S-L-V loci presented herein are continuous from solute triple point to dilute solute region, as the molecular differences between the solute and the solvent are not severe. The nearly "ideal" behavior in the systems *n*-butane with *n*-

decane and *n*-dodecane is not surprising if one recalls the near "ideal" behavior of the systems propane and these same solutes (7). Looking at these solutes and the solubility behavior in ethane (2, 3), propane, and butane, one notes that the solute composition increases with solvent molecule size at a given temperature. The same effect is noted with the solute benzene; however, along a portion of the *n*-butane-cyclohexane locus, the solute composition values are less than their corresponding values on the propane-cyclohexane locus.

From examination of the data herein and earlier data on *n*-paraffin solutes, one can suggest that nonideality is significant with the solvent ethane for *n*-paraffin solutes above *n*-octane, and with the solvent propane for solutes above *n*-decane, while the solvent *n*-butane is ideal for solutes up to *n*-dodecane.

Glossary

L	liquid phase
S	solid phase
T	temperature in K
T_T	triple-point temperature in K: 278.69 K for benzene, 279.83 K for cyclohexane, 243.51 K for <i>n</i> -decane, and 263.61 K for <i>n</i> -dodecane
T^*	T/T_T
x	solute mole fraction in phase L

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Supplementary Material Available: Complete tables of raw data for all six binary systems (8 pages). Ordering information is given on any current masthead page.

Viscosities and Densities of Four Binary Liquid Systems at 25.00 °C

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Densities and viscosities of four binary systems, viz., benzene-toluene, chloroform-toluene, chloroform-benzene, and benzene-*n*-hexane, have been determined at 25.00 °C, over the complete composition ranges. The results have been discussed in the light of ideal solution behavior. A few of the existing predictive equations of mixture viscosities have been tested.

Introduction

The ideal mixture concept, widely used in solution thermodynamics, corresponds to a limiting behavior that has a physical basis in the properties of highly dilute solutions, but may lack such basis in the case where the ideal mixture concept is extended to cover the entire mole fraction range of a liquid system. Presented in this paper are results of density and viscosity determinations on carefully selected binary liquid mixtures, such that could be expected to approximate ideal solution behavior as closely as any liquid system of chemically different components may.

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Table I. Properties of Pure Componentns at 25.00 °C

compd	density, kg/L			kinemtic viscosity $\times 10^6$ m ² /s			absolute viscosity $\times 10^3$ Pa·s		
	exptl value ^a	lit. values		exptl value	lit. values		exptl value	lit. values	
		min	max		min	max		min	max
benzene	0.8735, 0.8733	0.87323 (7)	0.87376 (18)	0.6902	0.6863 (1)	0.6915 (11)	0.6026	0.5996 (11)	0.6032 (7)
toluene	0.8621, 0.8619	0.8610 (16)	0.86232 (19)	0.6413	0.6414 (11)	0.6421 (7)	0.5526	0.5516 (11)	0.5535 (7)
chloroform	1.4727, 1.4723	1.4760 (8)	1.4799 (19)	0.3638	0.3649 (10)		0.5357	0.5400 (10)	
<i>n</i> -hexane	0.6547, 0.6550	0.65481 (18)	0.65502 (18)	0.4494	0.4484 (18)	0.4509 (7)	0.2942	0.2937 (18)	0.2957 (7)

^a First value, new cell; second, old cell.

Experimental Section

Preparation of Solutions. All solutions were prepared by weighing the individual components. A Mettler Gram-atic semi-micro balance having a stated precision of 2×10^{-8} kg was used. Static electricity, which tended to collect on glassware and produce erroneous weight measurements (20), was dissipated by ionizing the air within the balance case with a radioactive cesium pencil.

As suggested by Dullien (5), a standard bottle was calibrated to correlate apparent bottle weight to air density, and air buoyancy corrections were made.

Materials. All the organic chemicals used in this study, either for the density meter calibration or for the density and viscosity measurements, were supplied by J. T. Baker, BDH, Aldrich, and Eastman Organic Chemicals. The stated purity of all the organics was 99+ mol %. The manufacturer's claims were tested by chromatographic analysis. The purity as checked for all organics exceeded the manufacturer's claims.

Density Measurements. A digital precision density meter (Anton Paar K.G., Model DMA 02C) was employed for the determination of densities of liquids (2). This electronic instrument utilizes the principle of variation of the natural frequency of a hollow oscillator when filled with different fluids. The following three-parameter equation was used for the determination of densities

$$\rho = \frac{AT^2}{1 - BT^2} - C \quad (1)$$

where ρ is the density and T is the period of oscillation. During the course of this work, the original (old) density meter cell broke and a new cell was calibrated, using a different set of standard liquids. The densities measured in the two cells differed by a few units in the fourth decimal place. The best values of the parameters A , B , and C were determined at 25.00 °C by using the following substances, the densities of which were known from the literature: old cell: double distilled water ($\rho = 0.99707$) (12), *n*-pentane ($\rho = 0.62135$) (18), *n*-hexane ($\rho = 0.65503$) (18), carbon tetrachloride ($\rho = 1.58441$) (18), *m*-bromotoluene ($\rho = 1.40295$) (7), ethylene dichloride ($\rho = 1.24538$) (18); new cell: double distilled water, *m*-bromotoluene, *n*-octane ($\rho = 1.19859$) (19), carbon tetrachloride ($\rho = 1.58441$) (18). The densities were measured at 25 ± 0.01 °C.

Viscosity Measurements. Equipment. A commercially available Cannon-Ubbelohde viscometer of size 25, supplied by the Cannon Instrument Co., was used in a Model M-1 constant temperature bath, supplied by Cannon Instrument Co. This bath can control the temperature to ± 0.01 °C at 25 °C. Temperature was measured in the bath by a mercury-in-glass thermometer that had been calibrated against a standard thermometer supplied by the National Bureau of Standards. An electric stopwatch was used for time measurements.

Procedure. The method described in "Designation D-445-53T", published by the American Testing of Materials for Newtonian Liquids, for the measurement of viscosity was used in this work.

Viscosity Equation. The following equation has been used to calculate the viscosities (Cannon et al.) (3)

$$\mu/\rho = Ct - E/t^n \quad (2)$$

where C is a calibration constant, and the term E/t^n is known as the kinetic energy correction factor. The parameter E is supposed to be time independent, and the value of n depends on the shape of the capillary ends; for trumpet shaped ends, n has been estimated to be 2, and this value was used in this work.

Three viscosity standards (N.4, N.8, and N1.0) were used to determine the parameters C and E . These standards were supplied by Cannon Instrument Co. At least five readings were taken for each standard liquid. Average values of the calibration constant C , and the kinetic energy correction factor E , were obtained by the method of least squares.

Results and Discussion

A comparison between the measured densities, kinematic and absolute viscosities of the pure organic liquids used in this study, and the corresponding literature values are given in Table I. The experimental values are in agreement with the literature values, within experimental error, in all cases with the exception of the density and (consequently) the absolute viscosity of chloroform.

The experimental densities of the various mixtures are listed in Table II. The densities of the benzene-toluene system agree, within experimental error, with the data of Sanni et al. (16). The raw density-mole fraction data were fitted to polynomials, using the least-squares method. Table III lists the coefficients of the polynomials and the standard deviations for the systems under investigation.

The partial molar volumes of the components in each system were calculated from the density data. In the chloroform-toluene system, the partial molar volumes were constant within experimental error. In the benzene-toluene, benzene-chloroform, and benzene-*n*-hexane systems, the maximum deviations of the partial molar volumes from the molar volumes of the pure components were as follows: 0.2%, 0.5%, 0.4%, 0.9%, 1.3%, and 1.5%.

The additive-volume behavior of the chloroform-toluene system approaches ideality within experimental error. The rest of the systems deviate from ideal behavior, as measured by the additivity of volumes as a criterion (Asfour) (2).

The experimental viscosities of the various mixtures, and the excess activation energies, $\Delta^* G^E$ are given in Table IV. $\Delta^* G^E$ is related to the viscosities and molar volumes by the following relation (Reed and Taylor) (13)

$$\ln \mu_m V_m = x_A \ln \mu_A V_A^0 + x_B \ln \mu_B V_B^0 + \Delta^* G^E / RT \quad (3)$$

Table II. Densities of Binary Mixtures at 25 °C

weight fraction	mole fraction	density, kg/L	weight fraction	mole fraction	density, kg/L
Benzene-Toluene					
0.0000	0.0000	0.86185	0.6329	0.6074	0.8680
0.0418	0.0490	0.8619	0.6679	0.7034	0.8685
0.0867	0.1007	0.8623	0.7163	0.7486	0.8691
0.14155	0.1628	0.8628	0.7481	0.7779	0.8695
0.2219	0.25175	0.8636	0.7768	0.8041	0.8699
0.2837	0.31845	0.8642	0.8007	0.8257	0.87015
0.4059	0.4462	0.8654	0.8289	0.8511	0.8706
0.4247	0.4655	0.8655	0.8594	0.8782	0.8710
0.4469	0.4880	0.8658	0.9020	0.9157	0.8716
0.4765	0.5177	0.86615	0.9523	0.9593	0.8723
0.5117	0.5530	0.8665	0.9825	0.9851	0.8728
0.5434	0.5840	0.8669	1.0000	1.0000	0.8733
0.6037	0.6425	0.8676			
Chloroform-Toluene					
0.0000	0.0000	0.86185	0.4417	0.3791	1.0547
0.0254	0.0197	0.8709	0.4867	0.4225	1.08005
0.0623	0.04875	0.8845	0.64105	0.5795	1.1743
0.0997	0.0787	0.8989	0.7338	0.6803	1.23845
0.1083	0.0857	0.9021	0.7781	0.7301	1.2724
0.1518	0.1213	0.91965	0.81905	0.7774	1.3051
0.2058	0.1666	0.9421	0.8617	0.8279	1.3411
0.2605	0.2137	0.9660	0.9101	0.8866	1.3848
0.3037	0.2518	0.9859	0.97355	0.9660	1.4464
0.39575	0.3357	1.0309	1.0000	1.0000	1.4723
0.4321	0.3699	1.0495			
Chloroform-Benzene					
0.0000	0.0000	0.8733	0.5111	0.4062	1.1004
0.0442	0.0294	0.8888	0.6072	0.50285	1.1577
0.0657	0.0440	0.8967	0.6501	0.5487	1.1849
0.1065	0.0724	0.91203	0.6891	0.5919	1.2118
0.1448	0.0997	0.9269	0.71355	0.6197	1.2290
0.1814	0.1266	0.9416	0.7412	0.6520	1.2480
0.2533	0.18165	0.9720	0.7518	0.6646	1.2559
0.2983	0.2176	0.99215	0.7544	0.6677	1.2581
0.3409	0.2529	1.0119	0.8022	0.7262	1.2944
0.3685	0.2763	1.02525	0.8422	0.7774	1.3269
0.4054	0.3084	1.0437	0.90595	0.8631	1.3822
0.4410	0.3404	1.0620	0.9641	0.9408	1.4334
0.4559	0.3541	1.0705	1.0000	1.0000	1.4723
0.4780	0.37465	1.0818			
Benzene-n-Hexane					
0.0000	0.0000	0.6547	0.5581	0.5822	0.7583
0.0593	0.0651	0.66405	0.6886	0.7092	0.7885
0.1117	0.1218	0.6737	0.8061	0.8210	0.8182
0.2571	0.2763	0.69785	0.8967	0.9055	0.8433
0.3907	0.4144	0.7238	1.0000	1.0000	0.8735

^a Concentration of first-named component.

where V_A^0 and V_B^0 are the molar volumes of pure components A and B, and V_m is the molar volume of the liquid mixture. The raw viscosity-mole fraction data were fitted to polynomials by using the method of least squares. Table V gives the values of the least-squares constants. Figure 1 shows the change of $\Delta^* G^E$ with composition for all four systems under investigation.

It is evident that, as measured by this criterion of ideal solution behavior, the benzene-toluene and chloroform-benzene systems come very near to being ideal solutions, whereas the chloroform-toluene and benzene-n-hexane systems are far from behaving ideally. It is noted that the kinematic viscosity,

Table III. Least-Squares Constants for the Equation

$$\rho = \sum_{i=0}^{i=n} A_i x_A^i \text{ kg/L}$$

system	A_0	A_1	A_2	A_3	std dev, kg/L
benzene (A)-toluene (B)	0.86170	6.4747×10^{-3}	3.0602×10^{-3}	1.895×10^{-3}	7.6×10^{-5}
chloroform (A)-toluene (B)	0.86164	0.46523	0.10011	4.572×10^{-2}	3.3×10^{-4}
chloroform (A)-benzene (B)	0.87311	0.53366	6.1643×10^{-2}	3.9696×10^{-3}	2.4×10^{-4}
benzene (A)-n-hexane (B)	0.65463	0.14801	2.4784×10^{-2}	4.6073×10^{-2}	3.8×10^{-4}

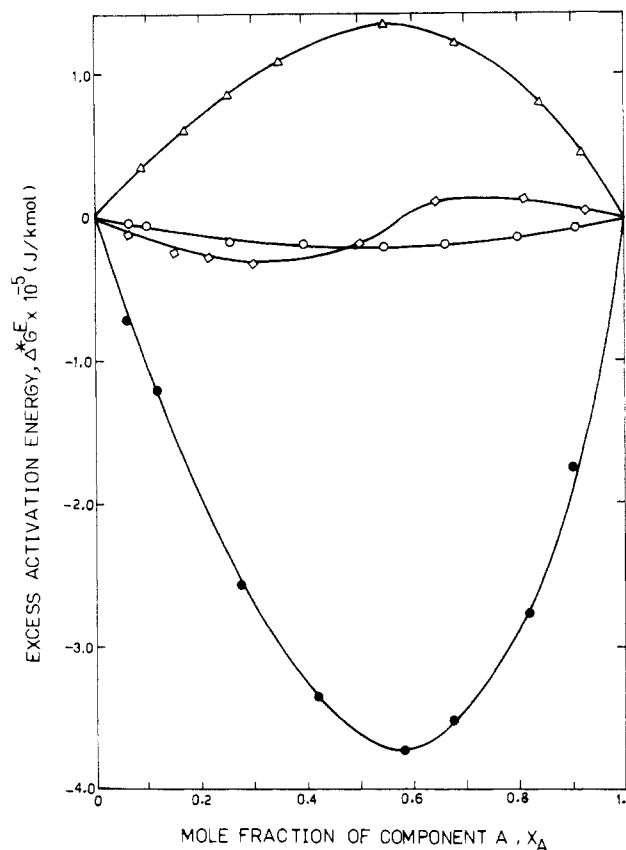


Figure 1. Variation of $\Delta^* G^E$ with composition at 25.00 °C: (O) benzene (A)-toluene (B) system; (Δ) chloroform (A)-toluene (B) system; (\diamond) chloroform (A)-benzene (B) system; (\bullet) benzene (A)-n-hexane (B) system.

ν , of the chloroform-toluene system plots as a perfect straight line vs. the mole fraction. The fact that ν decreases, whereas ρ increases as the composition changes from pure toluene to pure chloroform, gave rise to a pronounced maximum in the absolute viscosity of this system.

The solubility parameters of the four substances used in this study, listed in Table VI, would suggest that the three systems, benzene-toluene, chloroform-benzene, and chloroform-toluene, are all very nearly ideal, whereas the benzene-n-hexane system is much less ideal. The later expectation was confirmed by both the density and viscosity data.

The following predictive equations of solution viscosities have been tested with the experimental data. The semitheoretical McAllister equation (11):

$$\ln \nu_m = x_A^3 \ln \nu_A + 3x_A^2 x_B \ln \nu_{AB} + 3x_A x_B^2 \ln \nu_{BA} + x_B^3 \ln \nu_B - \ln \left\{ x_A + x_B \frac{M_B}{M_A} \right\} + 3x_A^2 x_B \ln \left\{ \left[2 + \frac{M_B}{M_A} \right] / 3 \right\} + 3x_A x_B^2 \ln \left\{ \left[1 + 2 \frac{M_B}{M_A} \right] / 3 \right\} + x_B^3 \ln \left[\frac{M_B}{M_A} \right] \quad (4)$$

This equation involves two adjustable parameters ν_{AB} and ν_{BA}

Table IV. Viscosities of the Binary Liquid Mixtures

x_A	kinematic viscosity \times $10^6 \text{ m}^2/\text{s}$	absolute viscosity \times $10^3 \text{ Pa}\cdot\text{s}$	$\Delta^*G^E \times$ 10^{-5} J/kmol
Benzene (A)-Toluene (B)			
0.0000	0.6413	0.5526	0.00
0.0655	0.6430	0.5543	-0.86
0.0991	0.6440	0.5554	-1.09
0.2566	0.6474	0.5591	-4.04
0.3969	0.6535	0.5652	-4.18
0.5481	0.6602	0.5720	-4.68
0.6618	0.6664	0.5783	-4.25
0.8029	0.6753	0.5874	-3.01
0.9087	0.6828	0.5951	-1.66
1.0000	0.6902	0.6026	0.00
Chloroform (A)-Toluene (B)			
0.0000	0.6413	0.5526	0.00
0.0912	0.6158	0.5573	8.38
0.1727	0.5933	0.5608	14.91
0.2562	0.5703	0.5636	20.52
0.3481	0.5455	0.5661	25.75
0.5471	0.4929	0.5687	32.81
0.6862	0.4536	0.5638	29.56
0.8428	0.4091	0.5531	19.18
0.9237	0.3858	0.5450	10.46
1.0000	0.3638	0.5357	0.00
Chloroform (A)-Benzene (B)			
0.0000	0.6902	0.6026	0.00
0.0672	0.6530	0.5938	-3.46
0.1508	0.6123	0.5847	-6.24
0.2151	0.5849	0.5795	-6.74
0.3057	0.5491	0.5722	-7.63
0.5108	0.4829	0.5613	-4.57
0.6489	0.4485	0.5590	2.39
0.8164	0.4063	0.5493	3.11
0.9301	0.3789	0.5403	0.69
1.0000	0.3638	0.5356	0.00
Benzene (A)- <i>n</i> -Hexane			
0.0000	0.4494	0.2942	0.00
0.0651	0.4490	0.2983	-16.93
0.1218	0.4508	0.3034	-28.92
0.2763	0.4558	0.3184	-61.18
0.4144	0.4686	0.3390	-79.92
0.5822	0.4958	0.3759	-89.20
0.7092	0.5282	0.4165	-83.97
0.8210	0.5710	0.4673	-66.37
0.9055	0.6177	0.5208	-41.51
1.0000	0.6902	0.6029	0.00

to be determined from experimental data. McAllister's correlation has been discussed in detail in the literature (6, 11, 14).

The equation of Tamura and Kurata (17):

$$\mu_m = x_A \phi_A \mu_A + x_B \phi_B \mu_B + 2[x_A x_B \phi_A \phi_B]^{1/2} \mu_{AB} \quad (5)$$

where x_A , x_B , μ_A , μ_B , ϕ_A , and ϕ_B are the mole fractions,

Table V. Least-Squares Constants for the Equations

system	A_0	A_1	A_2	A_3	A_4	A_5	std dev, m^2/s
benzene (A)-toluene (B)	0.64155	1.7994×10^{-2}	2.7307×10^{-2}	3.2638×10^{-3}			3.4×10^{-10}
chloroform (A)-toluene (B)	0.64155	-0.29292	9.0273×10^{-2}	-0.1241	4.8901×10^{-2}		4.1×10^{-10}
chloroform (A)-benzene (B)	0.69023	-0.58301	0.48042	-0.2843	5.9944×10^{-2}		9.19×10^{-10}
benzene (A)- <i>n</i> -hexane (B)	0.44927	5.3107×10^{-3}	-4.2092×10^{-2}	0.58158	-0.76935	0.46551	4.8×10^{-10}

system	A_0	A_1	A_2	A_3	A_4	A_5	std dev, Pa·s
benzene (A)-toluene (B)	0.55282	1.9735×10^{-2}	2.5262×10^{-2}	4.7394×10^{-3}			2.9×10^{-7}
chloroform (A)-toluene (B)	0.55284	4.286×10^{-2}	2.8577×10^{-2}	-0.11968	3.1019×10^{-2}		4.7×10^{-7}
chloroform (A)-benzene (B)	0.6028	-0.14706	0.19153	-0.11228			1.07×10^{-7}
benzene (A)- <i>n</i> -hexane (B)	0.29408	7.3487×10^{-2}	-6.1428×10^{-2}	0.59175	-0.7752	0.4809	3.24×10^{-7}

absolute viscosities, and volume fractions of the components A and B, respectively, and μ_{AB} is an empirical constant at each temperature level.

Cullinan's (4) equation:

$$\mu_m = \mu_A^{x_A} \mu_B^{x_B} \frac{V_A}{V_G} \quad (6)$$

where

$$V_A = x_A \bar{V}_A + x_B \bar{V}_B \quad (7)$$

$$V_G = \bar{V}_A^{x_A} \bar{V}_B^{x_B} \quad (8)$$

where V_A and V_B are the partial molal volumes of components A and B, respectively. Equation 6 is supposed to be applicable for regular solutions (4).

Equations 5 and 4 involve one and two constants, respectively, which were evaluated from the experimental data by the method of least squares, and these constants are listed in Table VII. The viscosities, as predicted by the various equations, together with the average errors were calculated for each system at different compositions

$$\% \text{ av error} = \frac{1}{n} \sum_{i=1}^n \frac{|\text{calcd} - \text{exptl}|}{\text{exptl}} \times 100 \quad (9)$$

where n is the number of experimental measurements at different compositions. The results are given in Table VIII.

Conclusions

The densities and viscosities of four regular solutions of varying degrees of nonideality have been determined at 25 °C. The standard deviations of the density and viscosity data were 1×10^{-4} - $4 \times 10^{-4} \text{ kg/L}$ and 1×10^{-7} - $5 \times 10^{-7} \text{ Pa}\cdot\text{s}$, respectively.

If additivity of volumes on mixing is used as a criterion of ideal solution behavior, then the chloroform-toluene system is ideal. The same system also exhibits ideal behavior in the dependence on its kinematic viscosity on the mole fraction, whereas the absolute viscosity exhibits a maximum as a function of composition (Asfour) (2).

All three viscosity models tested were at their poorest in the case of the benzene-*n*-hexane system. The McAllister model with two adjustable parameters was best, giving an overall mean deviation of 0.1% for the four systems, and a mean deviation of 0.18% for benzene-*n*-hexane, which is about as good as the experimental data and the five-constant polynomial. The Tamura-Kurata model with one adjustable parameter gave an overall mean deviation of 0.7% and a mean deviation of 2% in the case of benzene-*n*-hexane.

Cullinan's model with no adjustable parameters gave an overall mean deviation of 3.5% and a mean deviation of 10.8%

Table VI. Solubility Parameter (δ) of the Compounds Used in This Study at 25 °C^a

compd	δ , (J/m ³) ^{1/2}
benzene	1.875×10^4
toluene	1.823×10^4
chloroform	1.882×10^4
<i>n</i> -hexane	1.490×10^4

^a From ref 9.

Table VII. Numerical Values of the Constants Involved in Eq 4 and 5 Used for Predicting Viscosity of Binary Mixtures at 25 °C

system	ν_{AB}^a (eq 4)	ν_{BA}^a (eq 4)	μ_{AB}^b (eq 5)
benzene (A)-toluene (B)	0.24404	0.64777	0.5812
chloroform (A)-toluene (B)	0.17518	0.5548	0.5962
chloroform (A)-benzene (B)	0.16422	0.52708	0.5515
benzene (A)- <i>n</i> -hexane (B)	0.16751	0.45445	0.2800

^a These constants, if substituted in the McAllister model, will yield the kinematic viscosities of the liquid mixtures in centistokes units. The resulting viscosities may be converted to m²/s (SI units) by multiplication with 10⁻⁶. ^b This constant, if substituted in the Tamura and Kurata model, will yield absolute viscosities in centipoise units. To convert to Pa·s (SI units) multiply the resulting viscosities by 10⁻³.

Table VIII. Average Errors of Various Equations

system	% error		
	eq 4	eq 5	eq 6
benzene (A)-toluene (B)	0.03	0.04	0.87
chloroform (A)-toluene (B)	0.06	0.16	1.71
chloroform (A)-benzene (B)	0.11	0.53	0.55
benzene (A)- <i>n</i> -hexane (B)	0.18	2.02	10.80

in the case of benzene-*n*-hexane.

The 0.03% average error found for the benzene-toluene system with the help of McAllister's model is also representative of the average percent difference between our data and those measured by McAllister (11), who also determined viscosities of this binary system. Our data, however, show much less scatter in the 0.1-0.35 benzene mole fraction range and, therefore, they are to be preferred to McAllister's data in this range of compositions.

Ridgway and Butler (15) reported absolute viscosities of benzene-*n*-hexane. A comparison with literature data on pure benzene and *n*-hexane, and with our data for the whole composition range, suggests that their measurements might have had systematic error of about $+0.005 \times 10^{-3}$ Pa·s, on the average.

Glossary

A	calibration constant
B	calibration constant

C	calibration constant
E	kinetic energy correction factor
$\Delta^* G^E$	excess activation energy for viscous flow
R	gas constant
T	absolute temperature
V	molal volume
V^E	excess molal volume
t	efflux time
x	mole fraction
ν	kinematic viscosity
μ	absolute viscosity
ρ	density
ϕ	volume fraction

Subscript

A	first-named component in a binary system
B	second-named component in a binary system
m	mixture

Superscript

E	excess
0	pure component

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