Physicochemical Properties of Liquid Mixtures. 1.† Viscosity, Density, Surface Tension and Refractive Index of Cyclohexane + 2,2,4-Trimethylpentane Binary Liquid Systems from 25 °C to 50 °C

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The densities, viscosities, refractive indexes, and surface tensions of binary mixtures of cyclohexane + 2,2,4-trimethylpentane from 25 °C to 50 °C were measured. The experimental values were correlated with the temperature and mole fraction of the components. Typical models have been used to fit experimental results.

Introduction

Alkanes are an important series of homologous, nonpolar, organic solvents. They have often been used in studies of solute dynamics because their physicochemical properties as a function of chain length are well-known.[1] They are chemical compounds used in a large range of chemical processes.[2] For this reason, in the literature, a considerable amount of experimental studies of physicochemical properties have been published in the last 10 years.[3−5]

For the design of gas−liquid contactors, it is necessary to know the mass-transfer coefficient. To determine the different equations that modelize the mass-transfer process requires the knowledge of the density, viscosity, and surface tension of the liquid phase.

In the present work the viscosity, density, refractive index, and surface tension of cyclohexane + 2,2,4-trimethylpentane have been measured over the temperature range 25 °C to 50 °C.

Experimental Section

2,2,4-Trimethylpentane and cyclohexane were purchase from Aldrich and Sigma, respectively, with purity >99% and were used without further purification. Solutions were prepared by mass using a balance with an accuracy of $\pm 10^{-7}$ kg.

Densities of pure components and their mixtures were obtained using a pycnometric method (Gay−Lussac's pycnometer). Doubly distilled−deionized water and the pure components were used for calibration. The measured values for pure components were compared with those existing in the literature[6] (Table 1). Pycnometers containing the solutions were placed in a thermostatic bath maintained constant to ±0.05 °C. Then it was weighed with a Mettler AJ150 balance with a precision of $\pm 10^{-7}$ kg. Each density value was the average of at least five measurements. Maximum deviations from the average were always <0.1%. The uncertainly of these measurements was ±0.004 $g\cdot cm^{-3}$.

The kinematic viscosity was determined from the transit time of the liquid meniscus through a capillary viscosimeter supplied by Schott (0.46 mm of diameter), measured with a precision of ±0.001 mPa·s in a Shott-Geräte AVS 350 Ubbelohde viscometer. The capillary viscosimeter was immersed in a bath controlled to ±0.05 °C. The viscometer was calibrated with distilled−deionized water and the pure components, as is recommended by Marsh.[6] The deviations were <±0.1%. Each measurement was repeated at least 10 times. The maximum deviation was <±0.3%. The dynamic viscosity (η) was obtained by the product of kinematic viscosity (ν) and the corresponding density (ρ) of the binary mixture in terms of eq 1 for each temperature and mixture composition.

$$\eta = \rho\nu \quad (1)$$

The surface tensions of the aqueous solutions were measured using a Prolabo tensiometer, which employs the Wilhelmy plate principle.[7] The surface tensions of the pure water and pure components were determined and compared with literature values to calibrate the tesiometer. The detailed experimental procedure has been described elsewhere.[8] The solutions were thermostated with a precision of ±0.05 °C, and the uncertainty of the measurements was ±0.1 $mN\cdot m^{-1}$. In general, each surface tension value reported was an average of 5 to 10 measurements. The maximum deviations from the average value were always <±0.8%.

The refractive indexes from the sodium D line (λ = 589.26 nm) were measured using a thermostated Abbe Refractometer 2WAJ supplied by Shangai Optical Instrument Factory. The refractometer was frequently calibrated by using the glass test piece of known refractive index supplied by the manufactures. Water was circulated into the instrument through a thermostatically controlled bath maintained constant to ±0.05 °C. The mixtures were directly injected into the prism assembly of the instrument using a Hamilton syringe from the stock solution stored

† The present paper forms part of our program on the measurement of transport properties.

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10.1021/je000310x CCC: $20.00
Published on Web 04/21/2001

Table 1. Dynamic Viscosities (η), Densities (ρ), Surface Tensions (σ), and Refractive Indexes (n_D) of Pure Compounds at Different Temperatures

	ρ/g·cm^{-3}		η/mPa·s		n_D		σ/mN·m^{-1}	
T/°C	expt	lit. (ref)	expt	lit. (ref)	expt	lit. (ref)	expt	lit. (ref)
				2,2,4-Trimethylpentane				
25	0.6860	0.6878[a]	0.4784	0.4620[b]	1.3885	1.3890[c]	18.60	18.32[d]
		0.6885[b]				1.3892[b]		
30	0.6808	0.6837[a]	0.4517	0.4360[b]	1.3855	1.3865[c]	18.20	17.88[d]
		0.6844[b]				1.3877[b]		
35	0.6771	0.6801[b]	0.4264	0.4120[b]	1.3835	1.3846[c]	17.70	
40	0.6734	0.6754[a]	0.4056	0.3910[b]	1.3810		17.20	16.99[d]
		0.6760[b]						
45	0.6680		0.3848		1.3795		16.70	
50	0.6647	0.6669[a]	0.3652		1.3765		16.20	16.11[d]
				Cyclohexane				
25	0.7711	0.7712[e]	0.8958	0.8923[g]	1.4225	1.4235[h]	25.10	24.65[i]
		0.7737[f]		0.8770[f]		1.4232[f]		
30	0.7667	0.7644[e]	0.8224	0.8187[g]	1.4210	1.4208[h]	24.10	24.60[i]
		0.7691[f]		0.8040[f]		1.4203[f]		
35	0.7613	0.7597[e]	0.7575	0.7544[g]	1.4185	1.4181[h]	23.5	
		0.7643[f]		0.7370[f]		1.4175[f]		
40	0.7565	0.7562[e]	0.6995	0.6980[g]	1.4160	1.4154[h]	22.90	22.87[i]
		0.7596[f]		0.6820[f]		1.4148[f]		
45	0.7531	0.7526[e]	0.6536	0.6480[g]	1.4135	1.4128[h]	22.20	
50	0.7493	0.7490[e]	0.6045	0.6037[g]	1.4105	1.4101[h]	21.70	21.68[i]

[a] TRC d-1490 1998.[12] [b] Aralaguppi et al., 1999.[13] [c] TRC fa-1490 1998.[12] [d] TRC e-1490 1998.[12] [e] TRC d-2050 1998.[12] [f] Aminabhavi, 1996.[14] [g] TRC c-2100 1998.[12] [h] TRC fa-2050 1998.[12] [i] TRC e-1960 1998.[12]

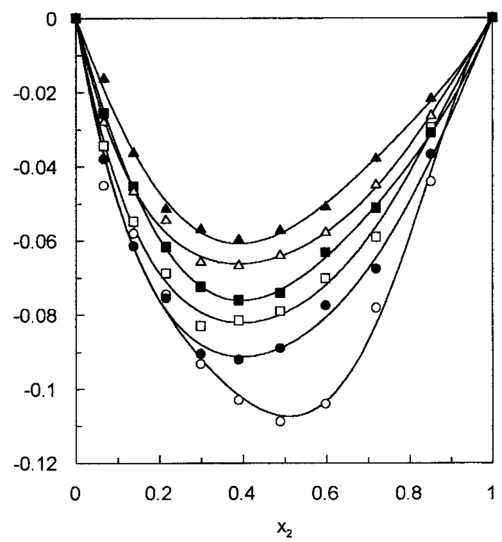

Figure 1. Deviations in viscosity versus temperature for cyclohexane + 2,2,4-trimethylpentane mixtures at 25 °C (○), 30 °C (●), 35 °C (□), 40 °C (■), 45 °C (△), and 50 °C (▲).

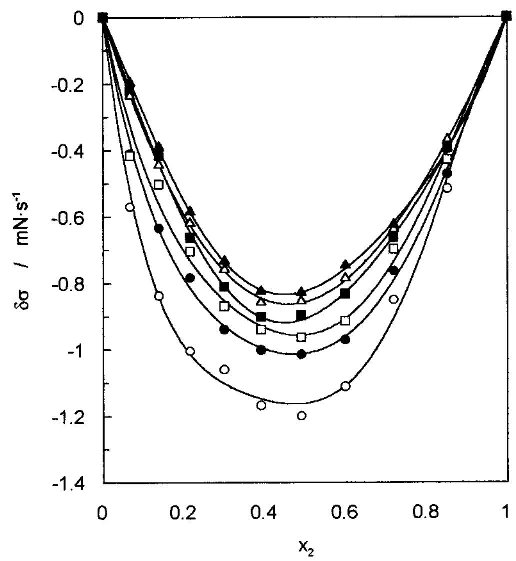

Figure 2. Deviations in surface tension versus temperature for cyclohexane + 2,2,4-trimethylpentane mixtures at 25 °C (○), 30 °C (●), 35 °C (□), 40 °C (■), 45 °C (△), and 50 °C (▲).

at the working temperature to avoid evaporation. The refractive index measurements were done after the liquid mixtures attained the constant temperature of the refractometer. This procedure was repeated at least three times. The average of these readings was taken for the refractive index values. The maximum deviations from the average value were always $<\pm 0.5\%$.

Results and Discussion

A comparison of bibliographic and our experimental values of the pure compounds is shown in Table 1. Our experimental results and the bibliographic data have a discrepancy < 0.6% for 90% of the values compared.

The densities (ρ), refractive indexes (n_D), dynamic viscosities (η), and surface tensions (σ) of cyclohexane (1) + 2,2,4-trimethylpentane (2) binary mixtures (25, 30, 35, 40, 45, and 50 °C) are listed in Table 2. All these physicochemical properties decrease upon increasing the temperature. For each temperature, density, refractive index, and surface tension decrease with increasing x_2.

The influence of the temperature on density (ρ), refractive index (n_D), and surface tension (σ) was a linear dependence in the temperature range studied,[9] whereas several authors in the literature have been found nonlinear dependences over a large ranges of temperature. In contrast with the observed behavior of density, surface tension, and refractive index, the influence of the temperature on the dynamic viscosity was not a linear dependence. For this reason, Andrade's equation has been used to fit our experimental results.

$$\eta = A e^{-BT} \tag{2}$$

Surface tension equations, which had been developed by our research group and published in several papers,[8] were fitted to the experimental results.

Table 2. Dynamic Viscosities (η), Densities (ρ), Surface Tensions (σ), and Refractive Indexes (n_D) of Liquid Binary Mixtures from 25 to 50 ×bcC

x_2	ρ/g·cm^{-3}	η/mPa·s	n_D	σ/mN·m^{-1}	x_2	ρ/g·cm^{-3}	η/mPa·s	n_D	σ/mN·m^{-1}
				t = 25 °C					
1.0000	0.6860	0.4784	1.3885	18.60	0.3895	0.7264	0.6484	1.4062	21.40
0.9238	0.6901	0.4888	1.3908	18.85	0.3430	0.7306	0.6717	1.4077	21.70
0.8517	0.6942	0.5006	1.3930	19.10	0.2984	0.7349	0.6954	1.4092	22.10
0.7834	0.6982	0.5135	1.3945	19.40	0.2557	0.7392	0.7147	1.4111	22.40
0.7185	0.7023	0.5267	1.3960	19.70	0.2147	0.7436	0.7344	1.4130	22.70
0.6569	0.7062	0.5406	1.3975	19.90	0.1754	0.7482	0.7572	1.4148	22.90
0.5982	0.7101	0.5549	1.3990	20.10	0.1376	0.7528	0.7806	1.4165	23.30
0.5423	0.7142	0.5752	1.4008	20.40	0.1012	0.7574	0.8041	1.4180	23.70
0.4891	0.7182	0.5959	1.4025	20.70	0.0662	0.7620	0.8283	1.4195	24.10
0.4382	0.7223	0.6219	1.4044	21.00	0.0350	0.7665	0.8617	1.4210	24.60
					0.0000	0.7711	0.8958	1.4225	25.10
				t = 30 °C					
1.0000	0.6808	0.4517	1.3855	18.20	0.3895	0.7226	0.5933	1.4036	20.80
0.9238	0.6851	0.4625	1.3878	18.40	0.3430	0.7269	0.6129	1.4053	21.10
0.8517	0.6893	0.4737	1.3900	18.60	0.2984	0.7312	0.6330	1.4070	21.40
0.7834	0.6935	0.4848	1.3918	18.90	0.2557	0.7357	0.6528	1.4088	21.70
0.7185	0.6977	0.4963	1.3935	19.20	0.2147	0.7401	0.6732	1.4105	22.05
0.6569	0.7019	0.5132	1.3951	19.40	0.1754	0.7446	0.6953	1.4123	22.40
0.5982	0.7060	0.5306	1.3967	19.60	0.1376	0.7491	0.7180	1.4140	22.70
0.5423	0.7101	0.5454	1.3984	19.90	0.1012	0.7535	0.7410	1.4158	23.00
0.4891	0.7142	0.5607	1.4000	20.20	0.0662	0.7579	0.7645	1.4175	23.30
0.4382	0.7184	0.5768	1.4018	20.50	0.0350	0.7623	0.7932	1.4193	23.70
					0.0000	0.7667	0.8224	1.4210	24.10
				t = 35 °C					
1.0000	0.6771	0.4264	1.3835	17.70	0.3895	0.7187	0.5545	1.4010	20.30
0.9238	0.6813	0.4368	1.3858	17.95	0.3430	0.7229	0.5717	1.4027	20.60
0.8517	0.6854	0.4475	1.3880	18.20	0.2984	0.7271	0.5895	1.4043	20.90
0.7834	0.6896	0.4570	1.3898	18.45	0.2557	0.7314	0.6066	1.4062	21.20
0.7185	0.6938	0.4668	1.3915	18.70	0.2147	0.7357	0.6242	1.4080	21.55
0.6569	0.6979	0.4812	1.3928	18.85	0.1754	0.7400	0.6438	1.4096	21.80
0.5982	0.7020	0.4960	1.3940	19.00	0.1376	0.7442	0.6639	1.4112	22.20
0.5423	0.7062	0.5098	1.3959	19.35	0.1012	0.7485	0.6844	1.4131	22.45
0.4891	0.7103	0.5240	1.3977	19.70	0.0662	0.7528	0.7054	1.4150	22.70
0.4382	0.7145	0.5390	1.3994	20.00	0.0350	0.7571	0.7312	1.4168	23.00
					0.0000	0.7613	0.7575	1.4185	23.50
				t = 40 °C					
1.0000	0.6734	0.4056	1.3810	17.20	0.3895	0.7148	0.5166	1.3984	19.70
0.9238	0.6775	0.4131	1.3835	17.45	0.3430	0.7190	0.5322	1.4002	20.00
0.8517	0.6816	0.4210	1.3860	17.70	0.2984	0.7232	0.5483	1.4020	20.40
0.7834	0.6857	0.4315	1.3875	17.95	0.2557	0.7274	0.5646	1.4038	20.70
0.7185	0.6899	0.4424	1.3890	18.20	0.2147	0.7315	0.5813	1.4055	21.10
0.6569	0.6940	0.4546	1.3903	18.35	0.1754	0.7357	0.6000	1.4073	21.40
0.5982	0.6982	0.4671	1.3915	18.50	0.1376	0.7398	0.6192	1.4090	21.80
0.5423	0.7024	0.4781	1.3933	18.90	0.1012	0.7440	0.6377	1.4106	22.00
0.4891	0.7065	0.4895	1.3950	19.30	0.0662	0.7483	0.6567	1.4122	22.20
0.4382	0.7107	0.5029	1.3967	19.40	0.0350	0.7524	0.6779	1.4141	22.50
					0.0000	0.7565	0.6995	1.4160	22.90
				t = 45 °C					
1.0000	0.6680	0.3848	1.3795	16.70	0.3895	0.7106	0.4880	1.3962	19.20
0.9238	0.6723	0.3929	1.3813	17.00	0.3430	0.7149	0.5024	1.3979	19.50
0.8517	0.6766	0.4013	1.3830	17.30	0.2984	0.7191	0.5172	1.3996	19.80
0.7834	0.6808	0.4106	1.3845	17.50	0.2557	0.7234	0.5318	1.4013	20.00
0.7185	0.6850	0.4203	1.3860	17.70	0.2147	0.7277	0.5467	1.4030	20.40
0.6569	0.6893	0.4305	1.3875	17.85	0.1754	0.7319	0.5628	1.4048	20.60
0.5982	0.6935	0.4411	1.3890	18.00	0.1376	0.7362	0.5793	1.4065	21.00
0.5423	0.6978	0.4527	1.3905	18.35	0.1012	0.7404	0.5952	1.4083	21.30
0.4891	0.7021	0.4647	1.3920	18.70	0.0662	0.7447	0.6115	1.4100	21.60
0.4382	0.7063	0.4762	1.3941	18.90	0.0350	0.7489	0.6323	1.4118	21.90
					0.0000	0.7531	0.6536	1.4135	22.20
				t = 50 °C					
1.0000	0.6647	0.3652	1.3765	16.20	0.3895	0.7071	0.4559	1.3940	18.60
0.9238	0.6689	0.3727	1.3783	16.50	0.3430	0.7113	0.4687	1.3954	18.90
0.8517	0.6732	0.3804	1.3800	16.80	0.2984	0.7156	0.4820	1.3968	19.20
0.7834	0.6774	0.3888	1.3818	16.95	0.2557	0.7198	0.4944	1.3986	19.50
0.7185	0.6816	0.3975	1.3835	17.10	0.2147	0.7241	0.5072	1.4003	19.85
0.6569	0.6859	0.4059	1.3853	17.30	0.1754	0.7283	0.5235	1.4023	20.20
0.5982	0.6901	0.4147	1.3870	17.50	0.1376	0.7325	0.5402	1.4042	20.50
0.5423	0.6944	0.4251	1.3880	17.80	0.1012	0.7368	0.5565	1.4058	20.70
0.4891	0.6986	0.4358	1.3890	18.10	0.0662	0.7410	0.5733	1.4073	21.00
0.4382	0.7029	0.4457	1.3915	18.40	0.0350	0.7451	0.5887	1.4089	21.40
					0.0000	0.7493	0.6045	1.4105	21.70

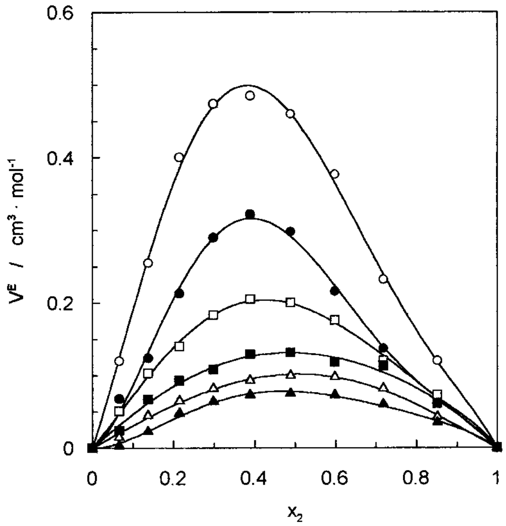

Figure 3. Excess volumes of cyclohexane + 2,2,4-trimethylpentane mixtures at 25 °C (○), 30 °C (●), 35 °C (□), 40 °C (■), 45 °C (△), and 50 °C (▲).

Table 3. Values of McAllister Three-Body Model Parameters

	McAllister parameters		
t/°C	$10^6\nu_{12}$/m²·s⁻¹	$10^6\nu_{21}$/m²·s⁻¹	AAD %
25	0.7351	0.8789	0.37
30	0.7232	0.7988	0.31
35	0.6880	0.7456	0.27
40	0.6452	0.7076	0.14
45	0.6223	0.6657	0.24
50	0.5892	0.6308	0.13

Table 4. Coefficents of the Redlich−Kister Type Equation for Viscosity Deviation ($\delta\eta$), Excess Volume(V^E), and Surface Tension Deviation ($\delta\sigma$)

	Redlich−Kister parameters for viscosity deviation/mPa·s				
t/°C	q_1	q_2	q_3	q_4	AAD($\delta\eta$)/%
25	−0.1227	−1.8679	3.7185	−2.4185	1.35
30	−0.2660	−0.2994	0.5870	−0.6749	0.12
35	−0.1818	−0.5481	0.9794	−0.8518	0.09
40	−0.2699	0.2822	−0.8698	0.4486	0.22
45	−0.1685	−0.3232	0.6025	−0.6015	0.39
50	−0.1875	0.1807	−0.7565	0.4567	1.46

	Redlich−Kister parameters for excess volume/(cm³·mol⁻¹)				
T/°C	q_1	q_2	q_3	q_4	AAD(V^E)/%
25	1.7180	5.4334	−14.8029	8.6215	0.33
30	0.3754	7.5380	−16.8182	9.6410	2.21
35	0.6867	1.5670	−3.7004	1.9569	0.82
40	0.5018	0.2624	−0.6400	0.4161	3.11
45	0.3237	0.2462	−0.0357	−0.2438	3.82
50	0.0092	1.8016	−3.3490	1.9106	3.41

	Redlich−Kister parameters for surface tension/mN·s⁻¹				
T/°C	q_1	q_2	q_3	q_4	AAD($\delta\sigma$)/%
25	−1.0053	−23.1501	49.5651	−35.8770	0.74
30	−2.0247	−12.0542	24.8951	−17.9128	0.57
35	−1.9898	−10.3589	20.3063	−13.8965	1.26
40	−3.3729	2.8391	−10.7647	8.1669	0.03
45	−2.7374	−1.1821	−0.9368	1.1204	0.12
50	−3.6994	5.7224	−14.8849	10.1484	0.14

In the case of the viscosity, and according with the literature, the kinematic viscosity/composition values were fitted to different models: the McAllister three-body model, the Grunberg−Nissan model, the Hind model, and the Heric model. In all of the cases the fits of the experimental data to the corresponding equations were satisfactory. In the present work we show the fits of experimental kinematic viscosity using the McAllister three-body model.[10] The McAllister three-body model is given by the equation

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2x_2 \ln \nu_{12} + 3x_1x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln\left[x_1 + x_2\frac{M_2}{M_1}\right] + 3x_1^2x_2 \ln\left[2 + \frac{M_2/M_1}{3}\right] + 3x_1x_2^2 \ln\left[\frac{1 + 2(M_2/M_1)}{3}\right] + x_2^3 \ln[M_2/M_1] \quad (3)$$

where x_1 and x_2 are the mole fractions of components 1 and 2 in a binary mixture, respectively, M_1 and M_2 are their molecular weights, and ν_1, ν_2, and ν are the kinematic viscosities of the pure components and the liquid mixture, respectively. ν_{12} and ν_{21} are adjustable parameters which are determined by fitting kinematic viscosity−composition data.

The excess molar volumes (V^E) and viscosity ($\delta\eta$) and surface tension deviations ($\delta\sigma$) were defined by eq 4 (Figures 1−3)

$$\delta Y = Y_m - (x_1Y_1 + x_2Y_2) \quad (4)$$

The deviation values of viscosity and excess molar volumes were fitted using a Redlich−Kister type equation (eq 5).[11]

$$\delta Y = x_1x_2\sum_{j=1}^{4} q_j x_2^{(j-1)/2} \quad (5)$$

The results are shown in Tables 3 and 4.

Acknowledgment

The authors thank the reviewers for their useful comments.

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Received for review September 19, 2000. Accepted February 13, 2001. The authors thank the Dirección General de Enseñanza Superior of Spain for financial support of their work (PB98-0622 and PB98-1089). D.G.-D. thanks Xunta de Galicia for a research grant.

JE000310X