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Densities and Viscosities of Binary Mixtures of *exo*-Tetrahydrodicyclopentadiene with *N*-Undecane or *N*-Tetradecane at $T = (293.15 \text{ to } 313.15)$ K

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Densities and viscosities of *exo*-tetrahydrodicyclopentadiene + *n*-undecane and *exo*-tetrahydrodicyclopentadiene + *n*-tetradecane were measured at different temperatures of (293.15, 298.15, 303.15, and 313.15) K and atmospheric pressure over the entire composition range. The excess molar volumes V_m^E and deviations in viscosity $\delta\eta$ for the mixtures were calculated from the experimental data and fitted to the Redlich−Kister equation, and the regression coefficients and the standard deviations of the fits were given. The viscosity data were correlated with several semiempirical equations, and the double-parameter McAllister equation gives satisfactory results.

Introduction

exo-Tetrahydrodicyclopentadiene ($C_{10}H_{16}$, tricyclo[5.2.1.$0^{2.6}$]decane), also called JP-10, is a special synthetic liquid hydrocarbon fuel with high volumetric energy density for volume limited applications. This single-component fuel takes advantage of the increased energy storage available through strained cyclic geometries.[1] Its heat of combustion (39.4 $MJ\cdot L^{-1}$) is substantially higher than petroleum-based fuels such as RP-3 (34.4 $MJ\cdot L^{-1}$), which makes it the standard missile fuel used by the U.S. Navy and the U.S. Air Force.[2] One problem with JP-10 is that its engineering specifications of ignition and combustion can be too poor in applications such as ramjets.[3]

Presently, many commercial fuels contain *n*-alkanes as major components to improve their performance.[4−6] We are interested in *n*-alkanes as additives for JP-10, designed to enhance combustion rates. In the present work, the densities and viscosities of the binary mixtures of JP-10 + *n*-undecane and JP-10 + *n*-tetradecane are measured at several temperatures and atmospheric pressure as fundamental data for the optimization of chemical processes. Furthermore, the excess molar volumes (V_m^E) of these systems were calculated and fitted to the Redlich−Kister type equation, which can provide important information about intermolecular interactions in binary mixtures.

Experimental Section

Materials and Characterization. JP-10 is obtained from Liming Research Institute of Chemical Industry in China with mass fraction purity better than 98 % analyzed by a 6890/5973 GC/MS, Hewlett-Packard, USA. The components are as follows: tricyclo[5.2.1.$0^{2.6}$]decane (98.06 %), bicyclo[4.4.0]decane (1.13 %), tricyclo[3.3.1.$1^{3.7}$]decane (0.81 %). The *n*-alkanes (*n*-undecane and *n*-tetradecane) with mass fraction purities better than 99 % are supplied by J&K Chemical Reagent Company, Shanghai, China. All reagents were used without further purification. The measured viscosity and density data of JP-10, *n*-undecane, and *n*-tetradecane were compared with the literature data and shown in Table 1.

Table 1. Densities, ρ, and Dynamic Viscosities, η, of Pure Compounds at 298.15 K

	$\rho/g\cdot cm^{-3}$		$\eta/mPa\cdot s$	
compound	exptl	lit.	exptl	lit.
JP-10	0.9314	0.9318, 0.9317[a]	2.784	2.767, 2.782[a]
n-undecane	0.7367	0.7365[b]	1.099	1.081[c]
n-tetradecane	0.7592	0.7594[d]	2.107	2.080[d]

[a] Ref 7. [b] Ref 8. [c] Ref 9. [d] Ref 10.

Apparatus and Procedure. The mixtures were prepared by mass using a Mettler Toledo AL204 balance with a stated precision of $\pm 1\cdot10^{-4}$ g. A special stoppered glass bottle was used to avoid evaporation in mass measurement, after which densities and viscosities were measured immediately. The uncertainty in the mole fractions was $\pm 1\cdot10^{-4}$.

The densities, ρ, of these pure liquids and their binary mixtures were measured by a DMA 5000 M density meter, Anton Paar, at several temperatures, (293.15, 298.15, 303.15, and 313.15) K, and the measurement cell temperature was controlled by a thermostat with a precision of 0.01 K. In a typical run, the densimeter was calibrated with double distilled water and dried air. The uncertainty of the density measurements was $\pm 5\cdot10^{-6}$ $g\cdot cm^{-3}$, corresponding to the uncertainty of $\pm 1\cdot10^{-3}$ $cm^3\cdot mol^{-1}$ in the excess volume, V_m^E.

The kinematic viscosities, ν, were determined at atmospheric pressure and different temperatures using an Ubbelohde viscometer calibrated with double distilled water. The viscometer was kept in a well-stirred water bath, providing temperature stabilization with an accuracy of 0.01 K. Flow times of the selected appropriate viscometer were always > 200 s, using a stopwatch with an accuracy of ± 0.01 s, and each viscosity value of the fluid was reported by averaging over three consecutive runs. The dynamic viscosity η of the liquid was then calculated according to eq 1.

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

where ν is the kinematic viscosity; η is the dynamic viscosity; and ρ is the density. The uncertainty of the viscosity measurements was within $\pm$ 0.008 $mPa\cdot s$.

Results and Discussion

Density and Viscosity Measurement. Experimental values of density and viscosity for the binary mixtures at different

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Table 2. Densities, ρ, Viscosities, η, Excess Molar Volumes, V_m^E, and Viscosity Deviations, $\delta\eta$, of JP-10 + *n*-Alkane Mixtures at (293.15, 298.15, 303.15, and 313.15) K

$x_{n\text{-alkane}}$	ρ / $g \cdot cm^{-3}$	η / $mPa \cdot s$	V_m^E / $cm^3 \cdot mol^{-1}$	$\delta\eta$ / $mPa \cdot s$
		JP-10 + *n*-undecane		
		T/K = 293.15		
0.0000	0.9353	3.076	0.0000	0.000
0.0881	0.9117	2.697	−0.0600	−0.214
0.1777	0.8893	2.390	−0.1012	−0.354
0.2711	0.8676	2.135	−0.1165	−0.432
0.3669	0.8468	1.921	−0.1138	−0.467
0.4654	0.8270	1.747	−0.1013	−0.456
0.5658	0.8082	1.608	−0.0857	−0.405
0.6685	0.7903	1.483	−0.0708	−0.336
0.7780	0.7726	1.373	−0.0554	−0.241
0.8855	0.7565	1.284	−0.0341	−0.128
1.0000	0.7404	1.199	0.0000	0.000
		T/K = 298.15		
0.0000	0.9314	2.784	0.0000	0.000
0.0881	0.9078	2.439	−0.0616	−0.197
0.1777	0.8854	2.162	−0.1041	−0.323
0.2711	0.8637	1.937	−0.1206	−0.390
0.3669	0.8430	1.751	−0.1189	−0.415
0.4654	0.8231	1.594	−0.1071	−0.406
0.5658	0.8044	1.470	−0.0919	−0.360
0.6685	0.7866	1.358	−0.0770	−0.298
0.7780	0.7689	1.259	−0.0605	−0.214
0.8855	0.7527	1.177	−0.0361	−0.113
1.0000	0.7367	1.099	0.0000	0.000
		T/K = 303.15		
0.0000	0.9275	2.525	0.0000	0.000
0.0881	0.9039	2.228	−0.0643	−0.164
0.1777	0.8816	1.982	−0.1089	−0.276
0.2711	0.8599	1.780	−0.1270	−0.338
0.3669	0.8392	1.615	−0.1264	−0.358
0.4654	0.8193	1.470	−0.1154	−0.356
0.5658	0.8006	1.359	−0.1004	−0.315
0.6685	0.7828	1.256	−0.0850	−0.263
0.7780	0.7652	1.167	−0.0673	−0.188
0.8855	0.7490	1.093	−0.0409	−0.099
1.0000	0.7330	1.022	0.0000	0.000
		T/K = 313.15		
0.0000	0.9196	2.112	0.0000	0.000
0.0881	0.8961	1.856	−0.0675	−0.148
0.1777	0.8738	1.662	−0.1163	−0.231
0.2711	0.8521	1.499	−0.1375	−0.279
0.3669	0.8315	1.362	−0.1392	−0.298
0.4654	0.8117	1.246	−0.1295	−0.292
0.5658	0.7930	1.158	−0.1148	−0.256
0.6685	0.7752	1.074	−0.0986	−0.212
0.7780	0.7577	1.000	−0.0785	−0.151
0.8855	0.7416	0.941	−0.0478	−0.077
1.0000	0.7256	0.878	0.0000	0.000
		JP-10 + *n*-tetradecane		
		T/K = 293.15		
0.0000	0.9353	3.077	0.0000	0.000
0.0703	0.9146	2.961	0.0170	−0.064
0.1451	0.8948	2.857	0.0366	−0.114
0.2274	0.8755	2.766	0.0567	−0.144
0.2692	0.8665	2.724	0.0662	−0.156
0.4088	0.8395	2.609	0.0919	−0.169
0.5067	0.8232	2.544	0.1037	−0.162
0.6160	0.8070	2.487	0.1024	−0.140
0.7324	0.7917	2.438	0.0938	−0.104
0.8606	0.7769	2.390	0.0646	−0.058
1.0000	0.7629	2.346	0.0000	0.000
		T/K = 298.15		
0.0000	0.9314	2.784	0.0000	0.000
0.0703	0.9108	2.679	0.0152	−0.057
0.1451	0.8910	2.582	0.0335	−0.103
0.2274	0.8717	2.496	0.0535	−0.133
0.2692	0.8627	2.456	0.0624	−0.145
0.4088	0.8358	2.350	0.0874	−0.157
0.5067	0.8195	2.291	0.0997	−0.150

Table 2. Continued

$x_{n\text{-alkane}}$	ρ / $\mathrm{g \cdot cm^{-3}}$	η / $\mathrm{mPa \cdot s}$	V_m^E / $\mathrm{cm^3 \cdot mol^{-1}}$	$\delta\eta$ / $\mathrm{mPa \cdot s}$
0.6160	0.8034	2.243	0.0974	−0.123
0.7324	0.7882	2.197	0.0841	−0.092
0.8606	0.7734	2.151	0.0532	−0.051
1.0000	0.7592	2.107	0.0000	0.000
		T/K = 303.15		
0.0000	0.9275	2.525	0.0000	0.000
0.0703	0.9069	2.429	0.0140	−0.052
0.1451	0.8871	2.343	0.0296	−0.090
0.2274	0.8680	2.259	0.0461	−0.123
0.2692	0.8590	2.223	0.0535	−0.133
0.4088	0.8321	2.125	0.0780	−0.143
0.5067	0.8159	2.072	0.0903	−0.135
0.6160	0.7998	2.028	0.0872	−0.110
0.7324	0.7846	1.983	0.0754	−0.082
0.8606	0.7698	1.940	0.0501	−0.044
1.0000	0.7557	1.897	0.0000	0.000
		T/K = 313.15		
0.0000	0.9196	2.112	0.0000	0.000
0.0703	0.8992	2.034	0.0124	−0.042
0.1451	0.8795	1.958	0.0253	−0.079
0.2274	0.8604	1.888	0.0394	−0.107
0.2692	0.8514	1.857	0.0468	−0.116
0.4088	0.8247	1.777	0.0696	−0.123
0.5067	0.8085	1.734	0.0846	−0.115
0.6160	0.7925	1.698	0.0811	−0.095
0.7324	0.7774	1.663	0.0685	−0.070
0.8606	0.7627	1.629	0.0475	−0.037
1.0000	0.7486	1.594	0.0000	0.000

temperatures and atmospheric pressure are given in Table 2. The excess molar volumes V_m^E can be calculated from the experimental results using eq 2.

$$V_m^E = \frac{M_1x_1 + M_2x_2}{\rho_m} - \left(\frac{M_1x_1}{\rho_1} + \frac{M_2x_2}{\rho_2}\right) \tag{2}$$

In the equation, x_1 and x_2 are the mole fractions; M_1 and M_2 are the molar masses; and ρ_m, ρ_1, and ρ_2 are the densities of the mixture, where component 1 is n-undecane or n-tetradecane and component 2 is JP-10. The viscosity deviations $\delta\eta$ can be obtained from the following relation

$$\delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \tag{3}$$

where η_i ($i = 1, 2$) represents the dynamic viscosity of the ith component, and η is the corresponding viscosity of the mixture. The calculated values of V_m^E and $\delta\eta$ are reported in Table 2.

The values of excess molar volumes V_m^E and the viscosity deviations $\delta\eta$ were fitted to the Redlich−Kister type polynomial equation

$$Y = x_1x_2\sum_{i=1}^{K} A_i(x_1 - x_2)^{i-1} \tag{4}$$

where Y is either V_m^E or $\delta\eta$; x_1 and x_2 are the mole fractions of n-alkane and JP-10, respectively; and A_i are the polynomial coefficients obtained by the least-squares method. The standard deviation, σ, was defined as

$$\sigma = \left[\frac{\sum (Y_{\text{exptl}} - Y_{\text{calcd}})^2}{n - k}\right]^{1/2} \tag{5}$$

where n is the total number of measurements and k is the number of estimated parameters. The correlation parameters calculated from eq 4 are listed in Table 3, together with the standard deviations (σ).

The variations of the excess molar volumes V_m^E with the mole fraction of the n-alkanes are shown in Figure 1. It can be seen that values of V_m^E are negative for the binary mixture of JP-10 + n-undecane over the entire composition range, and the excess molar volume decreases with the increase in temperature. It is positive for the other binary mixture of JP-10 + n-tetradecane on the contrary, and the values decrease with temperature from (293.15 to 313.15) K.

The reasons of variations can be discussed in terms of chemical, physical, and molecular structural factors. The chemical factors are the specific forces between molecules, such as hydrogen bonds and charge transfer, forming hydrogen bonds bringing negative V_m^E values, breaking hydrogen bonds, and complexes giving positive V_m^E values. The physical interactions include electrostatic forces, induction forces, and dispersion forces, making a weak contribution to V_m^E, and the sign of V_m^E may be positive or negative. The structural characteristics arise from the geometrical fitting of one component into another structure, due to the different sizes of the component molecules, leading to negative contributions to the excess molar volumes.[11,12]

For the systems of JP-10 + n-alkanes, JP-10 is a cyclic alkane, and n-undecane and n-tetradecane are chain alkanes. There is an opportunity for the small chain molecule n-undecane to enter into the interstitial space of JP-10, which can lead to contraction in volume. Nevertheless, the larger size of n-tetradecane hindered the molecular packing between two components, and the compounds are nonpolar alkane molecules. The weak dispersive interactions between molecules can also play a role in this aspect resulting in positive V_m^E value. For the system of JP-10 + n-undecane, the contribution of the dispersive interactions is weaker than the one of structural characteristics to induce the negative excess molar volume.

Table 3. Parameters of the Redlich−Kister Equation and Standard Deviations σ of JP-10 + n-Alkane Mixtures

	T/K	A_0	A_1	A_2	A_3	σ
			JP-10 + n-undecane			
V_m^E/cm^3·mol^{-1}	293.15	−0.39324	0.33286	−0.24583	−0.13545	0.00199
	298.15	−0.41803	0.32162	−0.24324	−0.12304	0.00210
	303.15	−0.45169	0.31474	−0.24940	−0.12016	0.00211
	313.15	−0.50989	0.30880	−0.24609	−0.13799	0.00233
$\delta\eta$/mPa·s	293.15	−1.76551	0.80111	−0.28218	0.09447	0.00224
	298.15	−1.56592	0.73838	−0.31330	0.12128	0.00164
	303.15	−1.37362	0.62659	−0.20792	0.04375	0.00218
	313.15	−1.11664	0.49998	−0.24261	0.24007	0.00310
			JP-10 + n-tetradecane			
V_m^E/cm^3·mol^{-1}	293.15	0.40484	0.13816	0.01792	0.06300	0.00096
	298.15	0.38969	0.11984	−0.06810	0.00504	0.00208
	303.15	0.34538	0.12613	−0.04075	−0.00641	0.00168
	313.15	0.31430	0.14114	−0.03585	−0.02662	0.00268
$\delta\eta$/mPa·s	293.15	−0.64688	0.26834	−0.0922	0.05939	0.00116
	298.15	−0.59279	0.29825	−0.05756	−0.03715	0.00127
	303.15	−0.53774	0.28538	−0.03378	−0.3869	0.00136
	313.15	−0.4658	0.28254	−0.01323	−0.11878	0.00084

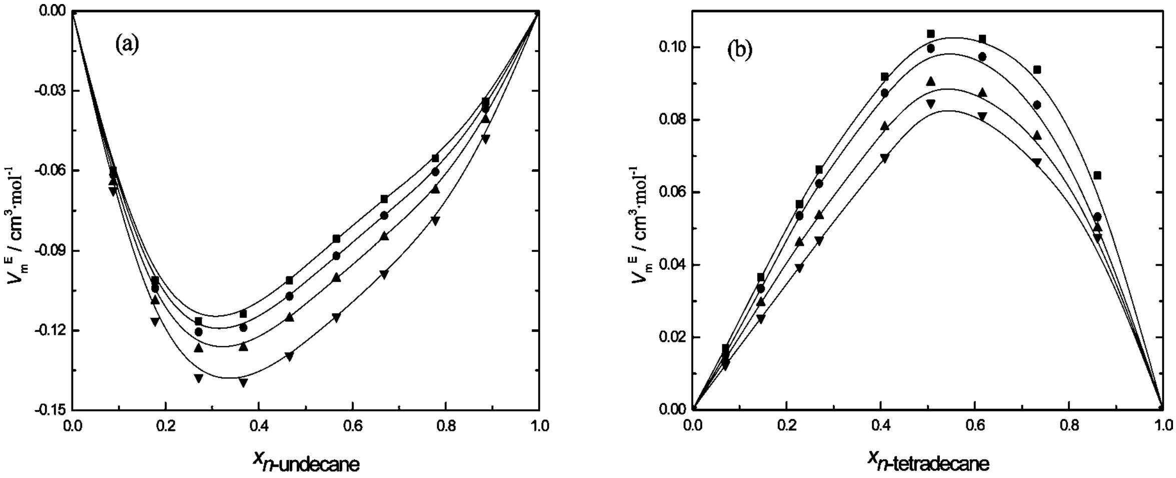

Figure 1. Excess molar volume variation with mole fraction for the systems (a) JP-10 + n-undecane and (b) JP-10 + n-tetradecane at different temperatures (T): ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K.

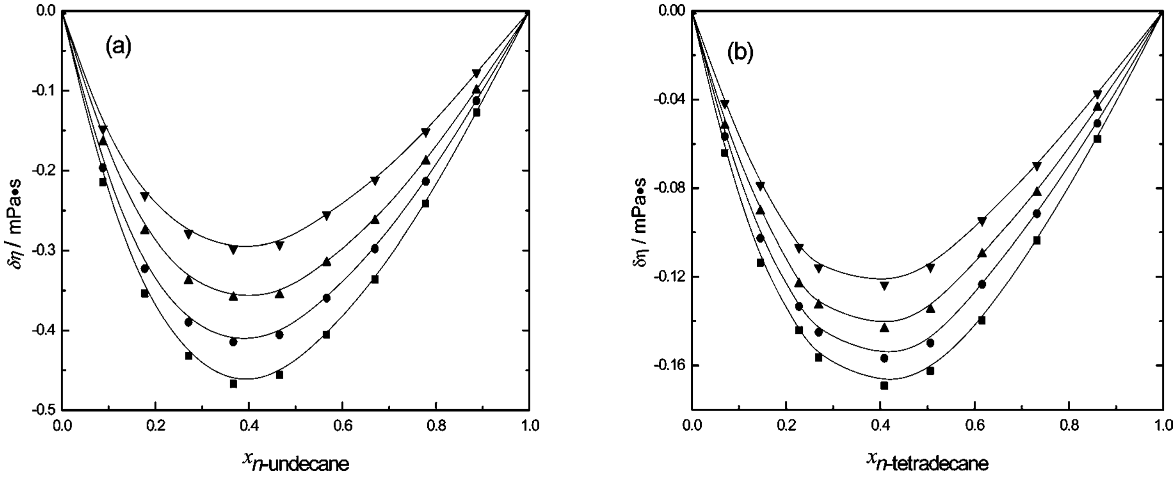

Figure 2. Viscosity deviation variation with mole fraction for the systems (a) JP-10 + n-undecane and (b) JP-10 + n-tetradecane at different temperatures (T): ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K.

The effect of temperature on V_m^E is noteworthy. With rise in temperature, the molecular motion becomes more fierce. There is a systematic decrease in V_m^E values for the system of JP-10 + n-tetradecane and an increase in absolute V_m^E values of JP-10 + n-undecane with increasing temperature.

The viscosity deviation $\delta\eta$ values for both of the binary systems are negative over the complete composition range, shown in Figure 2. The molecular size and shape of the component molecules and average degree of association of the mixture are important factors that influence the viscosity deviation in liquid mixtures. The absolute viscosities decrease as temperature increases due to decreased interaction between component molecules.[13]

Viscosity Data Correlation. The study of viscosity for binary liquid systems represents a tool for investigating the formation of intermolecular complexes and provides a valuable aid for

Table 4. Correlation Deviations and Parameters of Semi-Empirical Viscosity Equations for JP-10 + *n*-Alkane Systems at (293.15, 298.15, 303.15, and 313.15) K

T/K		293.15	298.15	303.15	313.15	293.15	298.15	303.15	313.15
		JP-10+ *n*-undecane				JP-10 + *n*-tetradecane			
Kendall−Monroe	Dev/%	10.33	10.16	9.60	9.42	3.47	3.48	3.45	3.57
	σ/mPa·s	0.2222	0.1997	0.1733	0.1453	0.1060	0.0964	0.0868	0.0753
	A_{12}	−0.4962	−0.4834	−0.4638	−0.4550	−0.2075	−0.2096	−0.2080	−0.2176
Grunberg−Nissan	Dev/%	0.82	0.93	0.79	1.06	0.47	0.52	0.55	0.57
	σ/mPa·s	0.0189	0.0208	0.0154	0.0179	0.0160	0.0155	0.0148	0.0130
Grunberg−Nissan ($A_{12} = 0$)	Dev/%	7.87	7.76	7.34	7.30	3.28	3.28	3.24	3.37
	σ/mPa·s	0.1713	0.1547	0.1342	0.1143	0.1005	0.0911	0.0817	0.0712
	A_{12}	1.2582	1.1618	1.0897	0.9392	2.3918	2.1521	1.9449	1.6230
Hind	Dev/%	2.58	2.62	2.36	2.53	0.64	0.70	0.73	0.75
	σ/mPa·s	0.0596	0.0563	0.0458	0.0422	0.0219	0.0210	0.0198	0.0173
Hind ($A_{12} = 0$)	Dev/%	21.66	21.81	22.27	22.45	26.62	26.60	26.61	26.55
	σ/mPa · s	0.4295	0.3953	0.3725	0.3196	0.8083	0.7279	0.6581	0.5495
	W_{vis}/RT	−0.4294	−0.4293	−0.4291	−0.4288	−0.0395	−0.0416	−0.0340	−0.0492
Katti−Chaudry	Dev/%	0.77	0.76	0.77	0.77	0.96	0.33	0.36	0.38
	σ/mPa·s	0.0178	0.0175	0.0178	0.0177	0.0096	0.0098	0.0097	0.0088
Katti−-Chaudry ($x_1x_2W_{vis} = 0$)	Dev/%	6.77	6.78	6.77	6.76	0.69	0.69	0.66	0.77
	σ/mPa·s	0.1480	0.1479	0.1479	0.1477	0.0225	0.0212	0.0188	0.0183
	A_{12}	0.3766	0.2998	0.2097	0.0902	0.9108	0.7996	0.6992	0.5095
	A_{21}	0.6001	0.4951	0.4292	0.2290	0.9417	0.8424	0.7429	0.5688
McAllister	Dev/%	0.08	0.09	0.12	0.30	0.09	0.05	0.08	0.24
	σ/mPa·s	0.0022	0.0020	0.0024	0.0052	0.0032	0.0020	0.0024	0.0062

determining their stoichiometry and their relative thermostability. Many semiempirical and empirical equations can be used to correlate the experimental data.[14] In this work, five typical semiempirical relations are selected. The Kendall−Monroe equation,[15] which has no adjustable parameter, is expressed as eq 6.

$$\eta = (x_1\eta_1^{1/3} + x_2\eta_2^{1/3})^3 \tag{6}$$

The single-parameter correlations of Grunberg−Nissan,[16] Hind,[17] and Katti−Chaudry[18] are eqs 7, 8, and 9, respectively.

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1x_2A_{12} \tag{7}$$

$$\eta = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2A_{12} \tag{8}$$

$$\ln \eta V = x_1 \ln \eta_1V_1 + x_2 \ln \eta_2V_2 + x_1x_2W_{vis}/RT \tag{9}$$

where V_i is molar volume of pure component i and W_{vis} is an adjustable parameter.

The double-parameter McAllister equation[19] was defined as

$$\begin{aligned}\ln \eta = x_1^3 \ln \eta_1 + x_2^3 \ln \eta_2 + 3x_1^2x_2 \ln A_{12} + 3x_1x_2^2 \ln A_{21} - \\ \ln[x_1 + x_2M_2/M_1] + 3x_1^2x_2 \ln[2 + (M_2/M_1)/3] + \\ 3x_1x_2^2 \ln[1 + (2M_2/M_1)/3] + x_2^3\ln[M_2/M_1]\end{aligned} \tag{10}$$

where A_{12} and A_{21} are interaction parameters.

The correlation parameters and the standard deviations for the two mentioned binary mixtures are listed in Table 4, together with the average percentage deviations given by

$$\mathrm{Dev} = \frac{1}{N}\sum_{i=1}^{N} \frac{|\eta_{calcd} - \eta_{exptl}|}{\eta_{exptl}} \cdot 100\ \% \tag{11}$$

where N is the number of experimental points.

Grunberg−Nissan, Hind, and Katti−Chaudry equations give comparatively similar deviations. The Kendall−Monroe equation gives large prediction deviations, which is not appropriate for the investigated binary mixtures. The double-parameter McAllister equation gives satisfactory results with deviations from (0.05 to 0.3) %.

Conclusions

Densities and viscosities of JP-10 + *n*-undecane and JP-10 + *n*-tetradecane were experimentally determined over the whole composition range at atmospheric pressure from (293.15 to 313.15) K. The excess molar volumes were calculated and fitted with the Redlich−Kister type polynomial equation. The V_m^E values are negative for the binary mixture of JP-10 with *n*-undecane but positive for the system of JP-10 with *n*-tetradecane, and it is interesting to study further at what chain length of *n*-alkane the numerical values of the excess molar volume change from negative to positive. These deviation results can be explained in terms of the changes of molecular shapes and interactions. The deviations of viscosity, $\delta\eta$, are negative for all the mixtures over the entire composition range and become less negative with increasing temperature. The viscosity data were correlated with five semiempirical equations, and the McAllister equation gives satisfactory results.

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