Density, Viscosity, and Refractive Index for Binary Systems of *n*-C16 and Four Nonlinear Alkanes at 298.15 K

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This work aims at extending the characterization of binary systems containing mixtures of *n*-C16 with branched saturated hydrocarbons of similar, higher, and lower number of carbon atoms. The systems considered are *n*-C16 with isooctane (2,2,4-trimethylpentane), cyprane (2,2,4,4,6,8,8-heptamethylnonane), pristane (2,6,10,14-tetramethylpentadecane), and squalane (2,6,10,15,19,23-tetracosane). Density, viscosity, refractive index, molar refractivity, excess volume, viscosity deviation function, and refractivity deviation function at 298.15 K are reported. Measured data are correlated with semiempirical models and parameters are reported.

Introduction and Objective

The literature reports several papers concerning density, viscosity, and refractive index for pure linear hydrocarbons, and derived properties, such as excess volume, viscosity deviation function, specific and molar refractivity, and refractivity deviation function for their binary mixtures with linear and nonlinear hydrocarbons, at different temperatures and pressures. Among previous investigations, viscosity at different temperatures and pressures for seven hydrocarbons is reported in Krahn and Luft (1994); excess volumes of eight *n*-alkane binary mixtures at 293.15 K and 298.15 K are reported in Jiangning et al. (1994); density, viscosity, and refractive index, excess volume, and viscosity deviation function at 298.15 K for 36 binary systems of *n*-paraffins are given in Aucejo et al. (1995), for nine systems of *n*-paraffins with hexane isomers once more in Aucejo et al. (1995), and for mixtures of cyclohexane and paraffins in Aminabhavi et al. (1996); density and viscosity from 197 K and 348 K and up to 100 MPa are presented in Padua et al. (1996).

The present work reports density and excess volume, kinematic viscosity and viscosity deviation function, refractive index, molar refractivity, and the relevant deviation functions for four *n*-C16 mixtures with alkanes of the same, slightly higher, much higher, and much lower PM. They are: cyprane (2,2,4,4,6,8,8-heptamethylnonane), pristane (2,6,10,15-tetramethylpentadecane), squalane (2,6,10,15,-19,23-hexamethyltetracosane), and isooctane (2,2,4-trimethylpentane, respectively).

Experimental Section

Materials. All compounds used in this work were supplied by Aldrich and were used as received, without any further purification. The stated purity, density, viscosity, and refractive index of the pure components are reported in Table 1 and compared with available literature data.

Mixture preparation. The mixtures for density and refractive index measurements were prepared by mass. A

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Mettler balance, with a precision of 3×10^{-5} g, and airtight bottles were used. The possible error in the mole fraction is estimated to be lower than 5×10^{-5}. The mixtures for the kinematic viscosity were obtained by volume, using a Schott TA 20 − T100 automatic diluting device. Repeating tests on the same sample indicate that the error in mole fraction is less than 5×10^{-5}. Details on mixture preparation are reported in Fermeglia and Lapasin (1988).

Density. A vibrating tube digital densimeter, model DMA 602H − DMA 60 (Anton Paar), was used to measure density. It was equipped with calibrated thermometers with a precision of ±0.01 K, connected with a Heterofrig (Heto Birkerød) constant-temperature bath circulator, with a precision of $\pm$ 0.01 K. For the densimeter calibration, nitrogen and double-distilled water were used. Working procedures are described in more detail in De Lorenzi et al. (1997). The estimated precision is higher than 3×10^{-5} g·cm^{-3}.

Viscosity. Kinematic viscosity was measured with an Ubbelohde suspended-level capillary viscometer, coupled with a Schott electronic timer AVS 300 with a precision of $\pm$ 0.01, and a Schott TA 20−T100 automatic diluting device. The thermostat used was a Haake F3 instrument, with a precision of ±0.02 K. Working procedures are described in more detail in De Lorenzi et al. (1997). The estimated precision in measuring kinematic viscosity is approximately 2×10^{-4} mm^2·s^{-1}. For the calibration of the viscometer the Poulten Selfe & Lee standard oil K5 was used.

Refractive Index. Refractive indices were determined with respect to the sodium D line. An Abbe refractometer (ATAGO type 3) was used. It was connected with the same Heterofrig constant-temperature bath used for the density measurements. Calibration of the instrument was performed with double-distilled water. Precision is estimated to be better than 2×10^{-4}.

Results and Discussion

Density. The experimental data measured for the four binary systems investigated are reported in Table 2. Excess

Table 1. Comparison between Measured and Literature Data for Refractive Index n_D, Density δ, and Viscosity v of Pure Components at 298.15 K

solvent	purity level (%)[a]	refractive index		density (g·cm^{-3})		viscosity (Pa s)	
		(literature)	(this work)	(literature)	(this work)	(literature)	(this work)
n-hexadecane (*n*-C16)	99	1.4338[b] 1.43286[c] 1.4334[d] 1.43273[o] 1.43244[f]	1.4329	0.77003[i] 0.77046[j] 0.76999[k] 0.77020[i] 0.77000 0.7707[c] 0.76981[d] 0.77079[e] 0.77079[c] 0.7703[n] 0.76994[o] 0.77030[e] 0.76996[f]	0.77008	3.0782[i] 3.0732[k] 3.005[b] 3.0930[d] 3.052[h] 3.0607[l] 3.0248[p]	3.0810
2,2,4-trimethyl-pentane (*i*-C8)	99.7	1.38898[g] 1.3891[h]	1.3895	0.68781[g] 0.68771[m] 0.68785[a] 0.6876[h] 0.68750[r]	0.68778		0.4842
Cyprane (*i*-C16)	98		1.4381		0.78123		3.3428
Pristane (*i*-C19)	98		1.4370		0.77911		5.4793
Squalane (*i*-C30)	99		1.4503		0.80513		28.254

[a] Values supplied by Aldrich. [b] Aminabhavi et al. (1994). [c] Heric and Coursey (1971). [d] Aucejo A. et al., 1995. [e] Heric and Breuer (1967). [f] Dymond and Young (1980). [g] Riddick et al. (1986). [h] Yoshikawa et al. (1994). [i] Awwad et al. (1986). [j] Wilhelm et al. (1987). [k] Awwad et al. (1986). [l] Beg et al. (1995). [m] Chevalier et al. (1990). [n] Asfour et al. (1990). [o] TRC (1993). [p] De Lorenzi et al. (1994). [q] Malhotra and Woolf (1994). [r] Chandra Rao et al. (1988).

**Table 2. Experimental Density Data at 298.15 K and Calculated Excess Volumes**

x	$\rho/$ (g·cm^{-3})	$V^E/$ (cm^3·mol^{-1})	x	$\rho/$ (g·cm^{-3})	$V^E/$ (cm^3·mole^{-1})
xn-C16 + (1−x) *i*-C16			xn-C16 + (1−x) *i*-C8		
0.0000	0.78123	0.000	0.0000	0.68778	0.000
0.0810	0.78020	0.043	0.0999	0.70226	−0.245
0.2005	0.77875	0.082	0.2018	0.71489	−0.446
0.2928	0.77766	0.102	0.3151	0.72654	−0.516
0.3879	0.77655	0.119	0.4076	0.73477	−0.532
0.4892	0.77540	0.126	0.5081	0.74258	−0.496
0.6130	0.77404	0.120	0.6186	0.75011	−0.421
0.7028	0.77307	0.110	0.7052	0.75535	−0.336
0.8099	0.77195	0.085	0.8039	0.76076	−0.228
0.9145	0.77089	0.050	0.9174	0.76633	−0.086
1.0000	0.77008	0.000	1.0000	0.77008	0.000
xn-C16 + (1-x) *i*-C19			xn-C16 + (1-x) *i*-C30		
0.0000	0.77911	0.000	0.0000	0.80513	0.000
0.0612	0.77860	0.015	0.0869	0.80334	0.011
0.2014	0.77738	0.056	0.1871	0.80109	0.023
0.3002	0.77650	0.081	0.2723	0.79899	0.040
0.4112	0.77550	0.100	0.3912	0.79575	0.058
0.5154	0.77455	0.109	0.4905	0.79273	0.064
0.6033	0.77373	0.114	0.5924	0.78926	0.071
0.7100	0.77272	0.113	0.6885	0.78559	0.072
0.8027	0.77184	0.102	0.7942	0.78102	0.065
0.9142	0.77079	0.072	0.8969	0.77593	0.049
1.0000	0.77009	0.000	1.0000	0.77008	0.000

volumes V^E were obtained from density data from:

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \quad (1)$$

where x_1 and x_2 are the mole fractions of components 1 and 2, ρ_1 and ρ_2 are the densities of pure components 1 and 2, ρ is the density of the mixture, and M_1 and M_2 are the molecular weights of components 1 and 2.

The excess volume is appreciable and negative for the
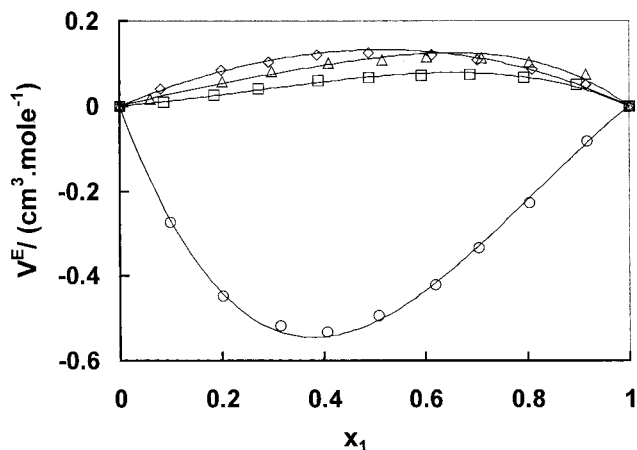
Figure 1. Excess volume, V^E, for *n*-hexadecane + isooctane (○), + cyprane (◇), + pristane (△), + squalane (□) system at 298.15 K.

n-hexadecane + 2,2,4-trimethylpentane system; for the other systems V^E is positive and much smaller; it decreases from cyprane to squalane and the minima are shifted to higher *n*-C16 mole fractions, as reported in Figure 1. Excess volumes were fitted to the Redlich and Kister-type expression (Redlich and Kister, 1984):

$$V^E = x_1 x_2 \sum_{j=0}^{n-1} a_j (x_1 - x_2)^j \quad (2)$$

where the a_j are adjustable parameters, n is the order of the polynomial, and x_1 and x_2 are the mole fractions of components 1 and 2.

The most appropriate order of the exponent in the fitting to the Redlich and Kister-type expression was determined

Table 3. Values of Coefficients and Relevant Standard Deviations for Eq 2

	n-C16 + *i*-C8	*n*-C16 + *i*-C16	*n*-C16 + *i*-C19	*n*-C16 + *i*-C30
a_0	−2.0177	0.49672	0.43563	0.26235
a_1	1.0298	0.03557	0.25626	0.19565
a_2	−0.0836	0.11521	0.24036	0.08271
σ (cm^3·mol^{-1})	0.0102	0.0021	0.0066	0.0037

Table 4. Experimental Kinematic Viscosity Data at 298.15 K and Calculated Dynamic Viscosity and Viscosity Deviation Function

x	$\nu/(10^{-6}$ m^2·s$^{-1})$	Δf	$\eta/$ (Pa·s)	x	$\nu/(10^{-6}$ m^2·s$^{-1})$	Δf	$\eta/$ (Pa·s)
	x*n*-C16 + (1−*x*) *i*-C16				x*n*-C16 + (1−x) *i*-C8		
0.0000	4.2791	0.0000	3.3428	0.0000	0.7038	0.0000	0.4842
0.0974	4.1697	−0.0194	3.2522	0.1010	0.9101	0.1070	0.6392
0.2011	4.0856	−0.0328	3.1815	0.1990	1.1399	0.1788	0.8143
0.3015	4.0224	−0.0416	3.1275	0.2987	1.3993	0.2211	1.0144
0.3995	3.9797	−0.0457	3.0897	0.4006	1.6885	0.2367	1.2398
0.4995	3.9526	−0.0458	3.0642	0.4999	2.0067	0.2365	1.4891
0.6002	3.9394	−0.0424	3.0496	0.6004	2.3567	0.2180	1.7650
0.7004	3.9387	−0.0358	3.0449	0.7002	2.7315	0.1837	2.0620
0.8000	3.9495	−0.0264	3.0492	0.8003	3.1452	0.1389	2.3912
1.0000	4.0010	0.0000	3.0811	0.9001	3.5544	0.0729	2.7200
				1.0000	4.0007	0.0000	3.0808
	x*n*-C16 + (1−x) *i*-C19				x*n*-C16 + (1−x) *i*-C30		
0.0000	7.0328	0.0000	5.4793	0.0000	35.0899	0.0000	28.254
0.0966	6.5905	−0.0093	5.1292	0.0450	32.3590	0.0248	26.054
0.1996	6.2306	−0.0063	4.8434	0.0990	29.3697	0.0518	23.586
0.2995	5.8788	−0.0073	4.5648	0.1473	26.8569	0.0736	21.539
0.3995	5.5486	−0.0082	4.3034	0.2008	24.2926	0.0958	19.452
0.4994	5.2441	−0.0081	4.0626	0.2977	20.2466	0.1336	16.164
0.5995	4.9608	−0.0073	3.8386	0.4495	14.9164	0.1671	11.844
0.6995	4.6979	−0.0058	3.6309	0.6011	10.8611	0.1805	8.5690
0.8001	4.4530	−0.0033	3.4375	0.8001	6.7976	0.1310	5.3067
0.8500	4.3187	−0.0062	3.3318	1.0000	4.0008	0.0000	3.0809
1.0000	4.0008	0.0000	3.0809				

on the basis of the *F* test at the 95% confidence level. The standard deviation (σ) obtained in the fitting was between 0.0021 and 0.0102 and is defined as follows:

$$\sigma = \sqrt{\frac{\sum (X_{exp} - x_{calc})^2}{N - n}} \quad (3)$$

where N is the number of experimental data, n is the number of parameters of the fitting equation, and X is the property considered (excess volume in this case). Redlich and Kister adjustable parameters a_j and σ are reported in Table 3.

A comment on the effects of mixing on molecular scale is out of the scope of this study; nevertheless it can be remarked that appreciable volume-reducing effects are due to mixing materials of different molar volumes, for example, for the *n*-C16 + isooctane system, where the small, almost egg-like molecules of 2,2,4-trimethylpentane enter the bulk of the planar, linear all-trans *n*-C16 molecules. On the other hand, adding the nonlinear, nonplanar molecules of the same or similar or even much larger molecular weight to the *n*-C16 molecules has a small and positive volume-increasing effect, which diminishes from cyprane to squalane (see Figure 1).

Viscosity. Experimental kinematic viscosities as a function of composition are reported in Table 4, together with the dynamic viscosity calculated by using the density reported in Table 2, interpolated with eq 2. The system *n*-C16 + cyprane displays a minimum around 0.65 mole fraction of *n*-C 16 (see Table 4).
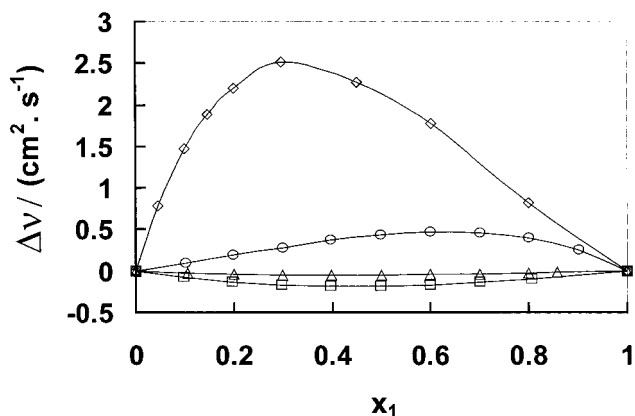
Figure 2. Kinematic viscosity deviation function for *n*-hexadecane + isooctane (○), + cyprane (◇), + pristane (△), + squalane (□) system at 298.15 K.

Table 5. Values of Coefficients and Relevant Standard Deviations for McAllister Equation (eq 4)

	n-C16 + *i*-C8	*n*-C16 + *i*-C16	*n*-C16 + *i*-C19	*n*-C16 + *i*-C30
$V_{1,2}$	2.7640	3.8929	4.7881	10.538
$V_{2,1}$	1.7398	3.8901	5.7064	19.553
σ (10^{-6} m^2·s^{-1})	0.0077	0.0014	0.0143	0.0304

Kinematic viscosity data were fitted to the McAllister (1960) expression:

$$\ln \nu = x_1^3 \ln\nu_1 + 3x_1^2 x_2 \ln\nu_{1,2} + 3x_1 x_2^2 \ln\nu_{2,1} + x_2^3 \ln\nu_2 +$$
$$-\ln(x_1 + x_2 M_2/M_1) + 3x_1^2 x_2 \ln[(2 + M_2/M_1)/3] +$$
$$3x_1 x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \quad (4)$$

where ν_1 and ν_2 are the kinematic viscosities of the pure components, M_1 and M_2 are the molecular weights of components 1 and 2, and $\nu_{1,2}$ and $\nu_{2,1}$ are adjustable parameters. The adjustable parameters obtained in the fitting and the relevant σ values (eq 3) are reported in Table 5. The following expression has been used for the evaluation of the viscosity deviation function:

$$\Delta f(\eta V) = \ln(\eta V) - \sum x_i \ln(\eta V)_i \quad (5)$$

Figure 2 shows the trends of the viscosity deviation function for the systems investigated.

Refractive Index and Refractivity. The refractive indices n_D of pure components and their mixtures investigated in this work are reported in Table 6. Molar refractivity, also reported in Table 6, was obtained from n_D experimental data according to the following expression:

$$R = \left[\frac{n_D^2 - 1}{n_D^2 + 2}\right] \frac{M}{\rho} \quad (6)$$

where M is the mean molecular weight of the mixture and ρ is the mixture density, interpolated by means of eq 2.

The Lorentz and Lorenz mixing rule in the form reported in Tasic et al. (1992) has been verified for the system under investigation. The equation used is the following:

$$n_D = \left[\frac{(2A + 1)}{(1 - A)}\right]^{0.5} \quad (7)$$

where A is given by:

Table 6. Experimental Refractive Index Data at 298.15 K and Calculated Molar Refractivity and Refractive Index by Lorentz−Lorenz Equation (Eq 7) and Refractivity Deviation Function (Eq 9)[a]

| X	ϕ	n_D exp	n_D calc eq 7	R	$\Delta R\phi$	ΔRx
			xn-C16 + (1−x) *i*-C8			
0.0000	0.0000	1.3895	1.3895	39.33	0.0000	0.0000
0.0883	0.1464	1.3970	1.3964	42.66	−2.0932	0.0558
0.2009	0.3080	1.4045	1.4037	46.85	−3.8952	0.0706
0.2989	0.4301	1.4110	1.4092	50.61	−4.6685	0.1968
0.5217	0.6588	1.4202	1.4190	58.82	−4.9325	0.1454
0.5905	0.7186	1.4224	1.4214	61.35	−4.6256	0.1243
0.7245	0.8232	1.4264	1.4257	66.28	−3.5675	0.0855
0.8146	0.8861	1.4288	1.4283	69.61	−2.5765	0.0746
0.8690	0.9215	1.4305	1.4297	71.67	−1.8333	0.1175
1.0000	1.0000	1.4329	1.4329	76.41	0.0000	0.0000
			xn-C16 + (1−x) *i*-C16			
0.0000	0.0000	1.4381	1.4381	76.11	0.0000	0.0000
0.1074	0.1088	1.4375	1.4374	76.15	0.0081	0.0078
0.2008	0.2031	1.4370	1.4369	76.18	0.0146	0.0098
0.3079	0.3110	1.4364	1.4363	76.21	0.0147	0.0076
0.4074	0.4109	1.4359	1.4358	76.25	0.0208	0.0178
0.4557	0.4593	1.4356	1.4355	76.26	0.0143	0.0133
0.4839	0.4875	1.4355	1.4354	76.28	0.0217	0.0248
0.6038	0.6072	1.4349	1.4347	76.31	0.0246	0.0189
0.6112	0.6146	1.4348	1.4347	76.31	0.0151	0.0166
0.7336	0.7364	1.4342	1.4341	76.34	0.0160	0.0099
0.7952	0.7975	1.4340	1.4338	76.38	0.0304	0.0314
1.0000	1.0000	1.4329	1.4329	76.41	0.0000	0.0000
			xn-C16 + (1−x) *i*-C19			
0.0000	0.0000	1.4370	1.4370	90.30	0.0000	0.0000
0.0924	0.0799	1.4367	1.4366	89.03	−0.1599	0.0134
0.1750	0.1532	1.4363	1.4363	87.87	−0.3006	0.0008
0.2639	0.2342	1.4360	1.4359	86.64	−0.4001	0.0056
0.4541	0.4151	1.4352	1.4351	84.00	−0.5311	0.0074
0.5628	0.5234	1.4348	1.4347	82.50	−0.5254	0.0173
0.7037	0.6696	1.4342	1.4341	80.55	−0.4513	0.0244
0.7808	0.7524	1.4340	1.4337	79.50	−0.3513	0.0453
0.8398	0.8173	1.4337	1.4335	78.67	−0.2795	0.0348
1.0000	1.0000	1.4329	1.4329	76.41	0.0000	0.0000
			xn-C16 + (1−x) *i*-C30			
0.0000	0.0000	1.4503	1.4503	141.21	0.0000	0.0000
0.1077	0.0633	1.4493	1.4492	134.27	−2.8449	0.0390
0.1892	0.1156	1.4484	1.4482	128.99	−4.7347	0.0402
0.2797	0.1786	1.4473	1.4471	123.13	−6.5122	0.0446
0.4057	0.2765	1.4456	1.4454	114.97	−8.3259	0.0494
0.4961	0.3554	1.444	1.4440	109.06	−9.1225	−0.0027
0.5618	0.4179	1.4431	1.4429	104.85	−9.2877	0.0446
0.6065	0.4632	1.4422	1.4421	101.93	−9.2672	0.0212
0.6963	0.5621	1.4405	1.4404	96.11	−8.6739	0.0202
0.8181	0.7158	1.4377	1.4377	88.19	−6.6365	−0.0071
0.8941	0.8254	1.4358	1.4359	83.26	−4.4608	−0.0123
1.0000	1.0000	1.4329	1.4329	76.41	0.0000	0.0000

[a] X, mole fraction; ϕ, volume fraction.

$$A = \left\{ \left[\left[\left(\frac{n_1^2 - 1}{n_1^2 + 2} \right) \frac{1}{\rho_1} \right] - \left[\left(\frac{n_1^2 - 1}{n_1^2 + 2} \right) \frac{w_2}{\rho_1} \right] + \left[\left(\frac{n_2^2 - 1}{n_2^2 + 2} \right) \frac{w_2}{\rho_2} \right] \rho \right\}$$ (8)

where n_1 and n_2 are the pure component refractive indices, w_j the weight fraction, ρ the mixture density, and ρ_1 and ρ_2 the pure component densities. The molar refractivity deviation function shown in Figure 3 and reported in Table 7 has been calculated by the following expression:

$$\Delta R = R - \phi_1 R_1 - \phi_2 R_2$$ (9)

where ϕ_1 and ϕ_2 are volume fractions and R, R_1, and R_2 the molar refractivity of the mixture and of the pure components, respectively. There is no general rule that states how to calculate a refractivity deviation function:
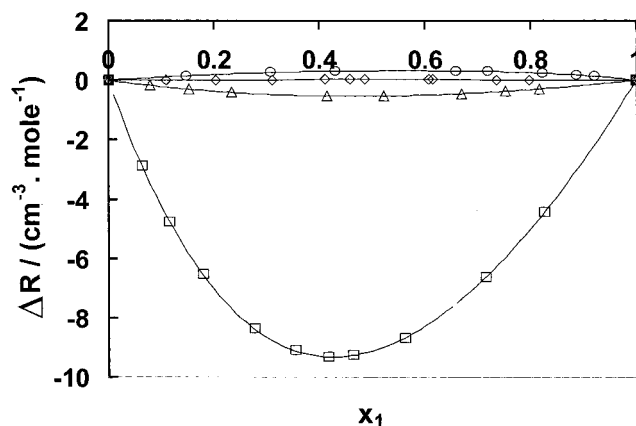
Figure 3. Molar refractivity deviation function for *n*-hexadecane + isooctane (○), + cyprane (◇), + pristane (△), + squalane (□) system at 298.15 K.

Table 7. Values of Coefficients and Relevant Standard Deviations for Eq 9 in Terms of Volume Fractions

	n-C16 + *i*-C8	*n*-C16 + *i*-C16	*n*-C16 + *i*-C19	*n*-C16 + *i*-C30
a_0	−19.9896	0.0711	−2.1497	−36.4684
a_1	−5.7662	0.0499	0.3458	10.3481
a_2	−1.8724		0.2342	−3.0455
a_3	0.3332			
σ (cm³·mol⁻¹)	0.0422	0.0072	0.0065	0.0179

Table 8. Values of Coefficients and Relevant Standard Deviations for Eq 9 in Terms of Mole Fractions

	n-C16 − *i*-C8	*n*-C16 − *i*-C16	*n*-C16 − *i*-C19	*n*-C16 − *i*-C30
a_0	0.6178	0.0754	0.0379	0.1256
a_1	−0.7734	0.0498	0.1648	−0.2279
a_2	0.0940		0.2805	0.0237
a_3	1.8303			
σ (cm³·mol⁻¹)	0.0339	0.0072	0.0063	0.0169

Konti et al. (1997) report deviations in molar refractivity with volume fraction referring to the Lorentz−Lorenz mixing rules. However, the molar refractivity is isomorphic to a volume for which the ideal behavior may be expressed in terms of mole fraction: in this case smaller deviations occur but data are more scattered because of the higher sensitivity of the expression to rounding errors in the mole fraction. For the sake of completeness, Table 6 reports both calculations of refractivity deviation function.

Molar refractivity deviation was fitted to a Redlich and Kister-type expression and the adjustable parameters and the relevant standard deviation σ are reported in Tables 7 and 8 for the expression in terms of volume fractions and in terms of mole fractions, respectively. The degree of the polynomial was determined on the basis of the F test at 95% confidence.

Finally, it can be concluded that the expressions used for interpolating the experimental data measured in this work gave good results, as can be seen by inspecting the σ values obtained.

The experimental data measured in this work are interesting inasmuch as they are able to give an insight into the effects on density, flow, and refractivity that derive from mixing saturated alkane molecules of different form and size in the the absence of polar or hydrogen bond contributions.

Acknowledgment

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