Density, Viscosity, and Refractive Index of the Binary Mixtures of Cyclohexane with Hexane, Heptane, Octane, Nonane, and Decane at (298.15, 303.15, and 308.15) K

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Experimental values of density, viscosity, and refractive index are presented for the binary mixtures of cyclohexane with hexane, heptane, octane, nonane, and decane at (298.15, 303.15, and 308.15) K over the entire mole fraction range of the mixture components at atmospheric pressure. From these data, excess molar volume, deviations in viscosity, and molar refractivity of the mixtures have been calculated. These quantities are further fitted to the Redlich−Kister relation to estimate the binary interaction parameters and standard errors. The results of excess molar volume and deviations in viscosity are comparable with the available published results at 298.15 K. The sign and magnitude of the calculated excess quantities have been discussed in terms of the nature of the solvent−solvent interactions in binary mixtures.

Introduction

The nature and type of interactions in binary organic liquid mixtures have been studied in terms of the mixing parameters such as excess molar volume, V^E, deviations in viscosity, $\Delta\eta$, and deviations in refractivity, ΔR. These parameters can be calculated from the direct measurements of density, ρ, viscosity, η, and refractive index for the sodium D line, n_D, of the mixtures as well as pure components. The present study is undertaken in view of the nonavailability of the extensive physical property data on binary mixtures of cyclohexane with *n*-alkanes at different temperatures. Such data are useful in process engineering design applications and other related areas.

In continuation of our earlier research concerning the accumulation of the binary physical property data on organic liquid mixtures (Aminabhavi et al., 1993, 1994; Aminabhavi and Bindu, 1995), we present here the experimental results of ρ, η, and n_D for the mixtures of cyclohexane with hexane, heptane, octane, nonane, and decane at (298.15, 303.15, and 308.15) K over the whole range of mixture compositions at atmospheric pressure. These results are used to calculate V^E, $\Delta\eta$, and ΔR values which are then fitted to the Redlich−Kister relation (Redlich and Kister, 1948) to estimate the binary interaction parameters and standard errors. The calculated values of V^E and $\Delta\eta$ are comparable with the results of Awwad and Salman (1986) and of Martin (1993) at 298.15 K.

Experimental Section

Materials. Cyclohexane was purchased from BDH. The analytical grade hexane, heptane, octane, nonane, and decane obtained from S. D. Fine Chemicals Ltd. were used directly as supplied. The solvent purity was ascertained by comparing their density, viscosity, and refractive index values with the literature data (Table 1). The GLC analyses were performed using a flame ionization detector (Nucon series, model 5700/5765, with fused silica columns) with a sensitivity better than 10^{-8} g of fatty acid/μL of solvent. These data for each liquid are also included in Table 1.

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Binary mixtures were prepared by mixing the calculated volumes of liquid components in airtight glass bottles. The mass measurements were done on a single-pan Mettler balance (Model AE-240) to an accuracy of ±0.0001 g. The possible error in the mole fraction is less than ±0.0002.

Measurements. Densities of liquids and their mixtures were obtained by using a pycnometer having a bulb volume of 15 cm^3 and a capillary with an internal diameter of 1 mm. Densities are accurate to ±0.0002 g·cm^{-3}. An average of triplicate measurements was taken into account, and these were reproducible within ±0.1%. The V^E values are accurate to ±0.05 cm^3·mol^{-1}. For all mixture compositions and pure solvents, triplicate measurements were performed, and the average of these values was considered in all calculations.

Viscosities were measured with a Cannon Fenske viscometer (size 100, Industrial Research Glassware Ltd., Roselle, NJ). An electronic stopwatch having a precision of ±0.01 s was used for the measurements of flow times. The viscosity values are accurate to ±0.001 mPa·s. Triplicate measurements of flow times were reproducible within ±0.05%. Computation of the kinematic viscosity, ν, was done by using the relation $\nu = At - B/t$, where t is the flow time of liquid levels between two marks in the viscometer and A and B are constants of the viscometer determined by calibrating with pure water and pure benzene at the working temperatures. Absolute viscosities, η/mPa·s, were then calculated using $\eta = \nu\rho$. The estimated errors in the η values are approximately ±0.05%.

Refractive indices were measured using the sodium D line with a thermostated Abbe refractometer (Bellingham and Stanley Ltd., London). Calibration checks of the refractometer are done routinely with the help of the glass piece of known refractive index provided with the instrument. At least three independent readings were taken for each composition, and the average of these readings was used to calculate the refractive index. Measurements were made at different temperatures by circulating water through the refractometer. Refractive indices are accurate to ±0.0002 units.

In all physical property measurements, an INSERF model 016 AP thermostat was used at a constant digital temperature control of ±0.01 K at the desired temperature.

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Table 1. Comparison of Experimental Densities (ρ), Viscosities (η), and Refractive Indices (n_D) of Pure Liquids with Literature Values at 298.15 K

	ρ/g·cm^{-3}		η/mPa·s		n_D	
liquid (mol % purity)	expt	lit.[a]	expt	lit.[a]	expt	lit.[a]
cyclohexane (>99.6)	0.7740	0.7739	0.883	0.892	1.4235	1.4235
hexane (>99.6)	0.6549	0.6549	0.298	0.297	1.3714	1.3723
heptane (>99.5)	0.6794	0.6795	0.388	0.391	1.3853	1.3851
octane (>99.2)	0.6989	0.6985	0.505	0.513	1.3949	1.3951
nonane (>99.4)	0.7140	0.7138	0.652	0.661	1.4021	1.4031
decane (>99.8)	0.7264	0.7264	0.844	0.841	1.4099	1.4097

[a] Marsh, K. N. *TRC Data Bases for Chemistry and Engineering—TRC Thermodynamic Tables*; Texas A & M University System: College Station, TX, 1994.

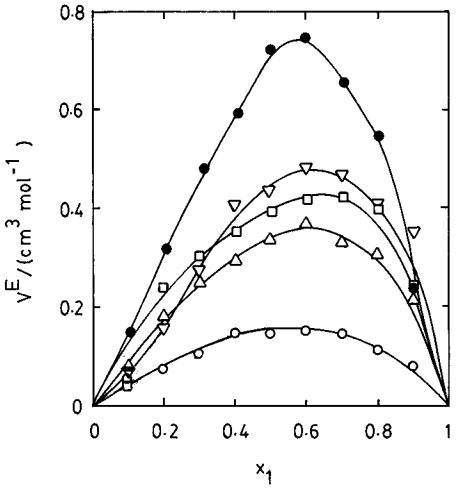

Figure 1. Excess molar volume vs mole fraction at 298.15 K for mixtures of cyclohexane with (○) hexane, (△) heptane, (□) octane, (▽) nonane, and (●) decane.

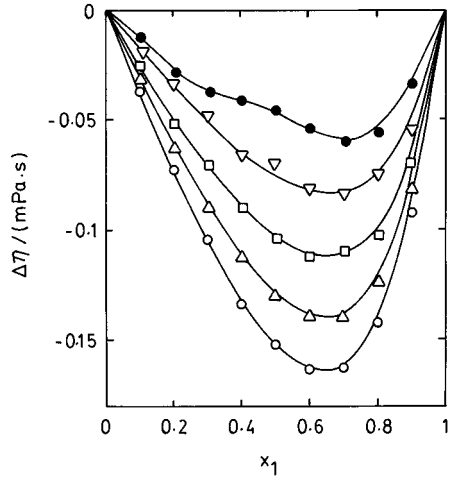

Figure 2. Deviations in viscosity vs mole fraction at 298.15 K for the binary mixtures given in Figure 1.

The experimental results of the binary mixtures compiled in Table 2 are the averages of at least three independent readings for each composition of the mixture.

Results and Discussion

The experimental values of density, viscosity, and refractive index presented in Table 2 are used to calculate V^E, $\Delta\eta$, and ΔR using the general equation of the type given earlier by Aminabhavi et al. (1993, 1994):

$$\Delta Y = Y_m - Y_1C_1 - Y_2C_2 \qquad (1)$$

where ΔY refers to V^E/cm^3·mol^{-1}, $\Delta\eta$/mPa·s, and ΔR/cm^3·mol^{-1}, respectively; Y_m is the measured mixture property

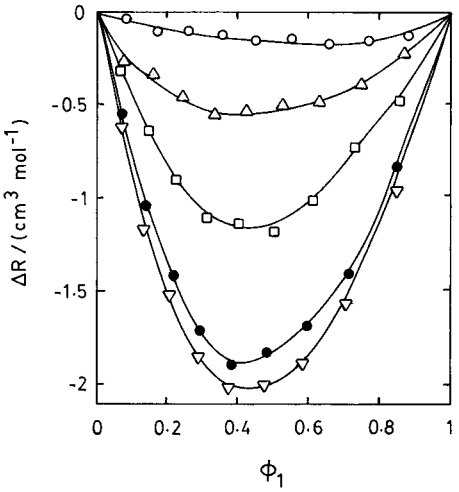

Figure 3. Deviations in molar refractivity vs volume fraction at 298.15 K for the binary mixtures given in Figure 1.

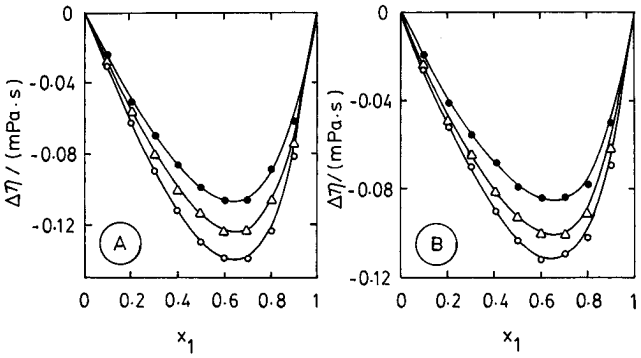

Figure 4. Deviations in viscosity vs mole fraction for binary mixtures of cyclohexane with (A) heptane and (B) octane at (○) 298.15 K, (△) 303.15 K, and (●) 308.15 K.

under question, and Y_i refers to the respective property of the pure component of the mixture. The symbols C_1 and C_2 are the mixture compositions expressed in mole fraction, x_i, of the ith component for the calculation of V^E and $\Delta\eta$.

The volume fraction, ϕ_i of the ith component is used to compute ΔR values. The volume fraction was calculated by using

$$\phi_i = x_iV_i/\sum_{i=1}^{2} x_iV_i \qquad (2)$$

To calculate the values of ΔR, Lorentz–Lorenz and Eykman equations have been used for the molar refractivity values, R_i and R_m, of the pure components and of the mixtures.

The calculated values of V^E, $\Delta\eta$, and ΔR have been fitted to the Redlich–Kister equation (Redlich and Kister, 1948) to estimate the coefficients, A_i by the method of least

Table 2. Experimental Densities (ρ), Viscosities (η), and Refractive Indices (n_D) of Binary Mixtures at Different Temperatures

x	ρ/g·cm^{-3}	η/(mPa·s)	n_D	x	ρ/g·cm^{-3}	η/(mPa·s)	n_D	x	ρ/g·cm^{-3}	n/(mPa·s)	n_D	x	ρ/g·cm^{-3}	η/(mPa·s)	n_D	x	ρ/g·cm^{-3}	η/(mPa·s)	n_D
Cyclohexane (1) + Hexane (2)				Cyclohexane (1) + Heptane (2)				Cyclohexane (1) + Octane (2)				Cyclohexane (1) + Nonane (2)				Cyclohexane (1) + Decane (2)			
298.15 K				298.15 K				298.15 K				298.15 K				298.15 K			
0.0000	0.6549	0.298	1.3714	0.0000	0.6794	0.388	1.3853	0.0000	0.6989	0.505	1.3949	0.0000	0.7140	0.652	1.4021	0.0000	0.7264	0.844	1.4099
0.0958	0.6642	0.317	1.3753	0.1013	0.6863	0.406	1.3864	0.0972	0.7037	0.516	1.3969	0.1060	0.7179	0.658	1.4035	0.1037	0.7289	0.836	1.4109
0.1981	0.6748	0.341	1.3792	0.2027	0.6935	0.425	1.3904	0.2042	0.7086	0.530	1.3985	0.1999	0.7212	0.664	1.4036	0.2065	0.7315	0.824	1.4115
0.2966	0.6850	0.367	1.3841	0.3040	0.7012	0.448	1.3930	0.3014	0.7142	0.548	1.4001	0.2987	0.7250	0.672	1.4055	0.3077	0.7343	0.818	1.4122
0.3986	0.6961	0.397	1.3888	0.4037	0.7094	0.475	1.3952	0.4044	0.7205	0.568	1.4019	0.3971	0.7291	0.677	1.4064	0.4003	0.7380	0.818	1.4130
0.4972	0.7075	0.436	1.3935	0.4998	0.7178	0.505	1.4000	0.5017	0.7270	0.591	1.4051	0.4952	0.7341	0.696	1.4081	0.5005	0.7407	0.818	1.4142
0.5972	0.7194	0.484	1.3988	0.6018	0.7272	0.546	1.4040	0.6029	0.7344	0.620	1.4070	0.5978	0.7397	0.708	1.4105	0.6000	0.7450	0.814	1.4154
0.6988	0.7321	0.544	1.4039	0.6985	0.7372	0.594	1.4073	0.7041	0.7425	0.661	1.4110	0.6994	0.7463	0.730	1.4125	0.7045	0.7507	0.812	1.4169
0.7993	0.7453	0.623	1.4097	0.8024	0.7485	0.661	1.4117	0.8014	0.7513	0.705	1.4154	0.7989	0.7537	0.762	1.4157	0.8020	0.7568	0.819	1.4181
0.8977	0.7588	0.730	1.4157	0.9002	0.7604	0.751	1.4169	0.8971	0.7615	0.774	1.4182	0.8991	0.7621	0.805	1.4180	0.9001	0.7649	0.845	1.4204
1.0000	0.7740	0.883	1.4235	1.0000	0.7740	0.883	1.4235	1.0000	0.7740	0.883	1.4235	1.0000	0.7740	0.883	1.4235	1.0000	0.7740	0.883	1.4235
303.15 K				303.15 K				303.15 K				303.15 K				303.15 K			
0.0000	0.6502	0.284	1.3682	0.0000	0.6751	0.368	1.3820	0.0000	0.6948	0.479	1.3930	0.0000	0.7100	0.612	1.3997	0.0000	0.7225	0.786	1.4080
0.0958	0.6595	0.301	1.3722	0.1013	0.6819	0.386	1.3841	0.0972	0.6995	0.487	1.3946	0.1060	0.7139	0.617	1.4011	0.1037	0.7247	0.780	1.4084
0.1981	0.6699	0.324	1.3767	0.2027	0.6891	0.402	1.3880	0.2042	0.7043	0.498	1.3960	0.1999	0.7171	0.623	1.4019	0.2065	0.7275	0.766	1.4089
0.2966	0.6804	0.349	1.3810	0.3040	0.6968	0.422	1.3899	0.3014	0.7100	0.515	1.3979	0.2987	0.7206	0.632	1.4032	0.3077	0.7303	0.761	1.4097
0.3986	0.6915	0.378	1.3859	0.4037	0.7049	0.447	1.3931	0.4044	0.7162	0.533	1.3997	0.3971	0.7249	0.633	1.4046	0.4003	0.7339	0.761	1.4106
0.4972	0.7028	0.412	1.3908	0.4998	0.7132	0.477	1.3980	0.5017	0.7227	0.554	1.4029	0.4952	0.7298	0.648	1.4055	0.5005	0.7366	0.760	1.4120
0.5972	0.7147	0.454	1.3959	0.6018	0.7226	0.512	1.4005	0.6029	0.7299	0.581	1.4052	0.5978	0.7354	0.659	1.4080	0.6000	0.7409	0.756	1.4131
0.6988	0.7275	0.511	1.4013	0.6985	0.7326	0.556	1.4043	0.7041	0.7382	0.614	1.4081	0.6994	0.7421	0.679	1.4099	0.7045	0.7458	0.754	1.4143
0.7993	0.7405	0.580	1.4068	0.8024	0.7438	0.619	1.4088	0.8014	0.7466	0.656	1.4113	0.7989	0.7492	0.706	1.4118	0.8020	0.7524	0.759	1.4148
0.8977	0.7540	0.672	1.4132	0.9002	0.7557	0.694	1.4139	0.8971	0.7568	0.717	1.4150	0.8991	0.7573	0.743	1.4166	0.9001	0.7596	0.780	1.4184
1.0000	0.7692	0.813	1.4217	1.0000	0.7692	0.813	1.4217	1.0000	0.7692	0.813	1.4217	1.0000	0.7692	0.813	1.4217	1.0000	0.7692	0.813	1.4217
308.15 K				308.15 K				308.15 K				308.15 K				308.15 K			
0.0000	0.6456	0.271	1.3662	0.000	0.6707	0.349	1.3803	0.0000	0.6906	0.450	1.3908	0.000	0.7061	0.574	1.3978	0.0000	0.7186	0.733	1.4053
0.0958	0.6548	0.288	1.3695	0.1013	0.6775	0.365	1.3810	0.0972	0.6954	0.459	1.3919	0.1060	0.7098	0.576	1.3985	0.1037	0.7208	0.728	1.4058
0.1981	0.6651	0.307	1.3739	0.2027	0.6847	0.379	1.3855	0.2042	0.7002	0.469	1.3935	0.1999	0.7130	0.582	1.3987	0.2065	0.7236	0.713	1.4064
0.2966	0.6758	0.330	1.3784	0.3040	0.6924	0.399	1.3870	0.3014	0.7058	0.483	1.3955	0.2987	0.7165	0.589	1.4005	0.3077	0.7263	0.708	1.4071
0.3986	0.6868	0.357	1.3832	0.4037	0.7004	0.422	1.3904	0.4044	0.7119	0.501	1.3974	0.3971	0.7208	0.591	1.4018	0.4003	0.7298	0.710	1.4082
0.4972	0.6981	0.389	1.3882	0.4998	0.7088	0.448	1.3948	0.5017	0.7184	0.518	1.4005	0.4952	0.7255	0.602	1.4032	0.5005	0.7325	0.710	1.4092
0.5972	0.7100	0.428	1.3932	0.6018	0.7181	0.481	1.3978	0.6029	0.7257	0.543	1.4023	0.5978	0.7311	0.616	1.4049	0.6000	0.7342	0.702	1.4104
0.6988	0.7227	0.479	1.3986	0.6985	0.7279	0.520	1.4014	0.7041	0.7337	0.573	1.4057	0.6994	0.7374	0.631	1.4075	0.7045	0.7415	0.699	1.4112
0.7993	0.7340	0.543	1.4043	0.8024	0.7392	0.577	1.4059	0.8014	0.7422	0.608	1.4086	0.7989	0.7447	0.658	1.4092	0.8020	0.7479	0.703	1.4134
0.8977	0.7492	0.623	1.4107	0.9002	0.7509	0.643	1.4110	0.8971	0.7522	0.664	1.4123	0.8991	0.7527	0.687	1.4130	0.9001	0.7550	0.716	1.4155
1.0000	0.7642	0.774	1.4175	1.0000	0.7642	0.744	1.4175	1.0000	0.7642	0.744	1.4175	1.0000	0.7642	0.744	1.4175	1.0000	0.7642	0.744	1.4175

Table 3. Estimated Parameters of Excess Functions for Mixtures

function	temp/K	A_0	A_1	A_2	σ
		Cyclohexane (1) + Hexane (2)			
$V^E/10^6(m^3 \cdot mol^{-1})$	298.15	0.662	−0.300	0.273	0.010
	303.15	0.588	−0.259	0.381	0.012
	308.15	0.540	−0.178	0.561	0.015
$\Delta\eta$/mPa·s	298.15	−0.612	0.347	−0.172	0.001
	303.15	−0.538	0.318	−0.187	0.002
	308.15	−0.469	0.259	−0.127	0.002
$\Delta R_{LL}/10^6(m^3 \cdot mol^{-1})$	298.15	−0.511	−0.145	−0.376	0.012
	303.15	−0.589	−0.386	−0.418	0.009
	308.15	−0.265	−0.003	−1.079	0.076
$\Delta R_{EYK}/10^6(m^3 \cdot mol^{-1})$	298.15	−1.163	−0.367	−0.898	0.028
	303.15	−1.348	−0.933	−1.007	0.020
	308.15	−0.578	−0.016	−2.541	0.178
		Cyclohexane (1) + Heptane (2)			
$V^E/10^6(m^3 \cdot mol^{-1})$	298.15	1.278	−0.567	0.277	0.012
	303.15	1.254	−0.536	0.326	0.013
	308.15	1.212	−0.536	0.266	0.011
$\Delta\eta$/mPa·s	298.15	−0.517	0.321	−0.173	0.002
	303.15	−0.457	0.277	−0.158	0.003
	308.15	−0.396	0.224	−0.122	0.002
$\Delta R_{LL}/10^6(m^3 \cdot mol^{-1})$	298.15	−2.185	0.445	−0.934	0.048
	303.15	−2.248	−0.127	−0.793	0.044
	308.15	−2.273	0.401	−0.959	0.054
$\Delta R_{EYK}/10^6(m^3 \cdot mol^{-1})$	298.15	−4.810	0.966	−2.192	0.113
	303.15	−4.960	−0.380	−1.873	0.104
	308.15	−5.008	0.860	−2.242	0.126
		Cyclohexane (1) + Octane (2)			
$V^E/10^6(m^3 \cdot mol^{-1})$	298.15	1.546	−0.715	0.485	0.042
	303.15	1.527	−0.643	0.567	0.043
	308.15	1.403	−0.606	0.422	0.043
$\Delta\eta$/mPa·s	298.15	−0.410	0.260	−0.179	0.002
	303.15	−0.369	0.222	−0.177	0.001
	308.15	−0.313	0.190	−0.131	0.002
$\Delta R_{LL}/10^6(m^3 \cdot mol^{-1})$	298.15	−4.677	1.161	−0.330	0.039
	303.15	−4.799	0.733	−0.730	0.022
	308.15	−4.698	1.015	−0.620	0.023
$\Delta R_{EYK}/10^6(m^3 \cdot mol^{-1})$	298.15	−10.28	2.562	0.799	0.091
	303.15	−10.56	1.547	−1.708	0.050
	308.15	−10.31	2.214	−1.439	0.054
		Cyclohexane (1) + Nonane (2)			
$V^E/10^6(m^3 \cdot mol^{-1})$	298.15	1.846	−1.514	0.373	0.049
	303.15	1.785	−1.437	0.336	0.067
	308.15	1.805	−1.313	0.624	0.053
$\Delta\eta$/mPa·s	298.15	−0.294	0.213	−0.148	0.003
	303.15	−0.262	0.199	−0.127	0.003
	308.15	−0.223	0.144	−0.099	0.003
$\Delta R_{LL}/10^6(m^3 \cdot mol^{-1})$	298.15	−7.956	1.967	−0.966	0.025
	303.15	−8.102	1.745	−0.279	0.033
	308.15	−8.001	2.173	−0.921	0.034
$\Delta R_{EYK}/10^6(m^3 \cdot mol^{-1})$	298.15	−17.52	4.275	−2.202	0.059
	303.15	−17.86	3.752	−5.589	0.077
	308.15	−17.62	4.765	−2.104	0.079
		Cyclohexane (1) + Decane (2)			
$V^E/10^6(m^3 \cdot mol^{-1})$	298.15	1.986	−1.155	−0.372	0.075
	303.15	1.911	−1.359	0.576	0.073
	308.15	2.373	−1.873	−0.966	0.193
$\Delta\eta$/mPa·s	298.15	−0.195	0.143	−0.150	0.003
	303.15	−0.169	0.126	−0.135	0.004
	308.15	−0.131	0.102	−0.139	0.004
$\Delta R_{LL}/10^6(m^3 \cdot mol^{-1})$	298.15	−11.58	3.251	−1.399	0.023
	303.15	−11.72	3.341	−1.839	0.041
	308.15	−11.37	3.596	−1.792	0.051
$\Delta R_{EYK}/10^6(m^3 \cdot mol^{-1})$	298.15	−25.48	7.116	−3.080	0.051
	303.15	−25.81	7.309	−4.145	0.095
	308.15	−25.00	7.925	−3.940	0.114

squares using the Marquardt algorithm (Marquardt, 1963). It was found that the number of coefficients, A_i, required for the best fit (i.e., to produce minimum error) in all the cases was three, viz. A_0, A_1, and A_2. The standard error, σ, values have been calculated by using the relation given in our earlier paper (Aminabhavi et al., 1993, 1994). The estimated results of A_i and σ are presented in Table 3.

The calculated V^E results of the binary mixtures at 298.15 K are presented in Figure 1. The smooth lines are drawn through these points. For all the mixtures, the values of V^E are positive. The positive V^E values increase systematically from hexane to decane, showing the effect of an increase of V^E with the increasing number of carbon atoms in *n*-alkanes. The plot of $\Delta\eta$ as a function of x_1 at 298.15 K presented in Figure 2, shows negative values

Table 4. Comparison of Equimolar Values of V^E and $\Delta\eta$ with the Literature Values at 298.15 K

cyclohexane with	property				
	V^E/cm^3·mol^{-1}			$\Delta\eta$/mPa·s	
	present	*a*	*b*	present	*a*
hexane	0.163	0.160	0.148	−0.152	−0.137
heptane	0.318	0.325	0.300	−0.130	−0.137
octane	0.374	0.408	0.384	−0.104	−0.104
nonane	0.452	0.449		−0.070	−0.084
decane	0.554	0.537		−0.045	−0.047

[a] Interpolated values of Awwad and Salman, 1986. [b] Martin, 1993.

for all the mixtures, and these values decrease systematically from decane to hexane. Similarly, the Lorentz−Lorenz ΔR values of the mixtures presented in Figure 3 at 298.15 K are also negative and generally show a decrease from hexane to octane. The results of ΔR calculated from the Eykman equation also follow the same trend but are not depicted graphically. The ΔR values of mixtures of cyclohexane with decane are slightly higher than those of the mixtures of cyclohexane and nonane.

In the literature, Awwad and Salman (1986) presented the values of V^E and $\Delta\eta$ for the binary mixtures of cyclohexane with *n*-alkanes at 298.15 K. Comparison of our V^E and $\Delta\eta$ values at 298.15 K suggests a reasonably good agreement for all the systems. The shapes of the curves also remain identical to our data. In Table 4, we have compared the equimolar values of V^E and $\Delta\eta$ with the available literature data of Awwad and Salman (1986) and of Martin (1993) at 298.15 K. Our data agree closely with their published results.

The V^E and ΔR results of the mixtures of cyclohexane with *n*-alkanes do not exhibit a systematic effect on temperature over the temperature range (298.15−308.15) K. However, the $\Delta\eta$ values for the majority of the mixtures increase systematically with increasing temperature, as shown typically in Figure 4 for mixtures of cyclohexane with heptane and octane. No experimental data of ρ, η, and n_D on these mixtures are available in the literature at higher temperatures with which we can compare the present results.

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