Density, Speed of Sound, and Refractive Index for Binary Mixtures Containing Cycloalkanes and Aromatic Compounds at $T = 313.15$ K

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In this work, densities, speeds of sound, and refractive indices of 12 binary systems of cycloalkanes with aromatics (cyclopentane (1), or cyclohexane (1), or methylcyclohexane (1) + benzene (2), + toluene (2), + ethylbenzene (2)) at $T = 313.15$ K and at atmospheric pressure were determined over the whole composition range. From the experimental results, the excess properties (excess molar volumes, isentropic compression, excess molar isentropic compression, and refractive deviations) at $T = 313.15$ K of all studied binary systems were calculated and satisfactorily fitted to the Redlich−Kister equation.

Introduction

Information about the density of pure liquids and mixtures and their dependence on composition and temperature is important basic data used in chemical engineering designs (in different applications for surface facilities, pipeline systems, and mass transfer operations), solution theory, and molecular thermodynamics.

In this article, as an extension of our work concerning the study of the behavior of cycloalkanes with different solvents,[1−4] we show the experimental density, speed of sound, and refractive index at $T = 313.15$ K of 9 binary systems containing cycloalkanes (1) and aromatic (2) compounds (cyclopentane (1), or cyclohexane (1), or methylcyclohexane (1) + benzene (2), + toluene (2), + ethylbenzene (2)). Experimental density, speed of sound, and refractive index data over the whole composition range for all binary systems have been determined at $T = 313.15$ K at atmospheric pressure. The results were used to calculate excess molar volumes, isentropic compression, excess molar isentropic compression, and refractive deviations. The aromatic compounds used in this study are very important in petrochemical industries. A review of the literature shows that only one author[5] has measured some of these binary systems at the study temperature, but the author has not determined the speed of sound and the refractive index.

Experimental Section

Chemicals. Cyclopentane, cyclohexane, and methylcyclohexane were supplied by Fluka with purity higher than 99.0 %, 99.8 %, and 99.0 %, respectively. Benzene was supplied by Merck with purity higher than 99.8 %, and toluene and ethylbenzene were supplied by Sigma-Aldrich with purity higher than 99.8 % for both pure liquids. They were degassed ultrasonically and dried over molecular sieves type 4 Å supplied by Aldrich and kept under an inert argon atmosphere. Table 1 shows a comparison between experimental and literature data of pure components at $T = 298.15$ K.

Apparatus and Procedure. Samples were prepared by syringing known masses of the pure liquids into stoppered bottles. For mass measurements, a Mettler AX-205 delta range balance with a precision of $\pm 10^{-5}$ g was used. Good mixing was ensured by magnetic stirring. All samples were prepared immediately prior to measurements to avoid variations in composition due to evaporation of solvent. The uncertainty in the composition of the samples is $\pm 3 \cdot 10^{-4}$.

Table 1. Comparison of Measured Pure Component Properties Data with Literature Values at $T = 298.15$ K

	$\rho/(g \cdot cm^{-3})$		n_D	
compd	exptl	lit.	exptl	lit.
cyclopentane	0.7400	0.74045[a]	1.40342	1.40363[a]
cyclohexane	0.7738	0.77389[a]	1.42353	1.42354[a]
methylcyclohexane	0.7649	0.76506[a]	1.42053	1.42058[a]
benzene	0.8735	0.87360[a]	1.49775	1.49792[a]
toluene	0.8621	0.86219[a]	1.49399	1.49413[a]
ethylbenzene	0.8625	0.86253[a]	1.49304	1.49320[a]

[a] From ref 6.

Densities and speeds of sounds were measured using an Anton Paar DSA-48 digital vibrating-tube densimeter. The uncertainty in experimental measurements has been found to be less than $\pm 10^{-4}$ g·cm^{-3} for the density and ± 1 m·s^{-1} for the speed of sound. The apparatus was calibrated by water and air according to the manual instruction. The calibration was checked with pure liquids shown in Table 1.

To measure refractive indices of pure components and their mixtures at $T = 313.15$ K, an automatic refractometer (Abbemat-HP, Dr. Kernchen) with an uncertainty in the experimental measurements of $\pm 4 \cdot 10^{-5}$ was used.

Results and Discussion

The experimental data of density, speed of sound, refractive index, isentropic compression, excess molar volume, excess molar isentropic compression, and refractive index deviations for the binary systems (cyclopentane (1), or cyclohexane (1), or methylcyclohexane (1) + benzene (2), + toluene (2), + ethylbenzene (2)) at $T = 313.15$ K and atmospheric pressure are reported in Table 2. The excess molar volumes and refractive index deviation were calculated by the following equation

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Table 2. Densities (ρ), Refractive Indices (n_D), Speeds of Sound (u), and Isentropic Compression ($K_{S,m}$) of the Binary Mixtures at $T = 313.15$ K

x_1	ρ / g·cm^{-3}	n_D	u / m·s^{-1}	$K_{S,m}$ / m^3·TPa^{-1}·mol^{-1}	x_1	ρ / g·cm^{-3}	n_D	u / m·s^{-1}	$K_{S,m}$ / m^3·TPa^{-1}·mol^{-1}
Cyclopentane (1) + Benzene (2)									
0.0000	0.8575	1.48777	1230.5	0.07016	0.5917	0.7744	1.42988	1155.3	0.09169
0.0366	0.8520	1.48417	1224.4	0.07151	0.6893	0.7621	1.42110	1147.2	0.09499
0.0941	0.8434	1.47833	1215.7	0.07359	0.7983	0.7486	1.41158	1139.7	0.09855
0.1945	0.8286	1.46795	1200.6	0.07736	0.8971	0.7368	1.40315	1134.2	0.10160
0.2706	0.8178	1.46049	1190.2	0.08017	0.9471	0.7310	1.39904	1132.0	0.10303
0.3937	0.8007	1.44843	1175.5	0.08462	1.0000	0.7250	1.39456	1130.0	0.10449
0.4893	0.7878	1.43934	1165.0	0.08810					
Cyclopentane (1) + Toluene (2)									
0.0000	0.8482	1.48509	1240.9	0.08317	0.5862	0.7789	1.43503	1168.4	0.09567
0.0418	0.8436	1.48201	1235.6	0.08396	0.6895	0.7658	1.42526	1157.4	0.09797
0.0924	0.8380	1.47801	1228.7	0.08499	0.7951	0.7522	1.41512	1147.3	0.10022
0.1907	0.8268	1.46979	1215.8	0.08703	0.8958	0.7389	1.40537	1138.3	0.10238
0.2717	0.8173	1.46313	1205.2	0.08880	0.9456	0.7323	1.40037	1134.2	0.10339
0.3579	0.8070	1.45562	1194.8	0.09064	1.0000	0.7250	1.39456	1130.0	0.10449
0.4944	0.7904	1.44336	1178.6	0.09364					
Cyclopentane (1) + Ethylbenzene (2)									
0.0000	0.8493	1.48538	1257.3	0.09311	0.6281	0.7787	1.43421	1175.5	0.09970
0.0393	0.8455	1.48234	1251.9	0.09350	0.6870	0.7709	1.42853	1168.0	0.10042
0.0830	0.8412	1.47928	1246.4	0.09386	0.7988	0.7553	1.41702	1154.0	0.10186
0.1874	0.8305	1.47140	1232.9	0.09482	0.8980	0.7408	1.40651	1141.9	0.10314
0.2847	0.8200	1.46405	1219.9	0.09585	0.9475	0.7332	1.40072	1135.9	0.10383
0.3689	0.8105	1.45715	1209.3	0.09668	1.0000	0.7250	1.39456	1130.0	0.10449
0.5410	0.7899	1.44226	1186.8	0.09862					
Cyclohexane (1) + Benzene (2)									
0.0000	0.8575	1.48777	1230.5	0.07016	0.6058	0.7891	1.43724	1179.8	0.09435
0.0819	0.8462	1.47958	1218.9	0.07389	0.7000	0.7809	1.43118	1177.7	0.09736
0.1282	0.8402	1.47489	1213.2	0.07592	0.7989	0.7732	1.42538	1177.3	0.10010
0.2087	0.8301	1.46761	1204.5	0.07940	0.9059	0.7657	1.41966	1178.6	0.10264
0.2898	0.8205	1.46073	1197.0	0.08279	0.9505	0.7627	1.41753	1179.7	0.10359
0.4018	0.8085	1.45171	1188.6	0.08721	1.0000	0.7596	1.41513	1181.7	0.10445
0.5047	0.7983	1.44414	1183.3	0.09096					
Cyclohexane (1) + Toluene (2)									
0.0000	0.8482	1.48509	1240.9	0.08317	0.5965	0.7909	1.44073	1189.8	0.09868
0.0377	0.8444	1.48212	1236.8	0.08420	0.7019	0.7819	1.43356	1185.1	0.10079
0.0967	0.8382	1.47761	1230.0	0.08596	0.7958	0.7744	1.42760	1182.2	0.10236
0.1945	0.8285	1.47007	1220.1	0.08865	0.9011	0.7665	1.42097	1181.2	0.10363
0.2433	0.8234	1.46621	1215.4	0.09006	0.9500	0.7630	1.41807	1180.9	0.10416
0.3985	0.8086	1.45475	1202.3	0.09412	1.0000	0.7596	1.41513	1181.7	0.10445
0.5672	0.7934	1.44276	1191.4	0.09806					
Cyclohexane (1) + Ethylbenzene (2)									
0.0000	0.8493	1.48538	1257.3	0.09311	0.6009	0.7946	1.44349	1201.9	0.10190
0.0515	0.8447	1.48140	1251.8	0.09394	0.7005	0.7856	1.43614	1195.0	0.10297
0.0976	0.8405	1.47828	1247.0	0.09469	0.7976	0.7769	1.42925	1189.2	0.10382
0.2494	0.8267	1.46788	1231.8	0.09709	0.9017	0.7680	1.42208	1184.5	0.10431
0.3647	0.8162	1.45999	1221.3	0.09877	0.9504	0.7638	1.41865	1182.8	0.10445
0.3855	0.8143	1.45831	1219.3	0.09909	1.0000	0.7596	1.41513	1181.7	0.10445
0.5028	0.8036	1.45026	1209.4	0.10069					
Methylcyclohexane (1) + Benzene (2)									
0.0000	0.8575	1.48777	1230.5	0.07016	0.6120	0.7807	1.43297	1158.1	0.11059
0.0486	0.8493	1.48201	1220.3	0.07363	0.7053	0.7726	1.42729	1154.2	0.11604
0.0982	0.8413	1.47621	1211.2	0.07713	0.7994	0.7653	1.42214	1151.5	0.12125
0.1995	0.8267	1.46566	1195.3	0.08410	0.8991	0.7584	1.41728	1149.9	0.12644
0.2999	0.8135	1.45619	1182.5	0.09092	0.9530	0.7548	1.41480	1149.5	0.12918
0.4038	0.8013	1.44745	1172.0	0.09776	1.0000	0.7520	1.41272	1149.6	0.13138
0.4998	0.7913	1.44043	1164.6	0.10379					
Methylcyclohexane (1) + Toluene (2)									
0.0000	0.8482	1.48509	1240.9	0.08317	0.5986	0.7840	1.43702	1171.5	0.11352
0.0488	0.8421	1.48065	1233.3	0.08570	0.7022	0.7749	1.43008	1164.0	0.11848
0.0977	0.8361	1.47630	1226.0	0.08825	0.8016	0.7668	1.42397	1158.1	0.12299
0.1967	0.8246	1.46750	1212.4	0.09338	0.9003	0.7591	1.41817	1153.2	0.12735
0.2969	0.8136	1.45909	1200.1	0.09853	0.9487	0.7555	1.41549	1151.2	0.12940
0.3751	0.8055	1.45314	1191.7	0.10246	1.0000	0.7520	1.41272	1149.6	0.13138
0.5049	0.7927	1.44348	1179.2	0.10895					
Methylcyclohexane (1) + Ethylbenzene (2)									
0.0000	0.8493	1.48538	1257.3	0.09311	0.5977	0.7886	1.44003	1184.5	0.11621
0.0509	0.8438	1.48108	1249.9	0.09508	0.6968	0.7792	1.43303	1174.9	0.12004
0.0975	0.8389	1.47740	1243.4	0.09686	0.7988	0.7697	1.42601	1165.7	0.12396
0.1957	0.8286	1.46965	1230.3	0.10066	0.8973	0.7609	1.41941	1157.5	0.12764
0.2794	0.8200	1.46339	1219.8	0.10389	0.9483	0.7564	1.41609	1153.5	0.12952
0.4006	0.8079	1.45428	1205.4	0.10858	1.0000	0.7520	1.41272	1149.6	0.13138
0.4962	0.7984	1.44736	1194.9	0.11230					

$$V^{E} = \sum_{i=1}^{N} x_i M_i(\rho^{-1} - \rho_i^{-1}) \quad (1)$$

$$\Delta n_D = n_D - \sum_{i}^{N} x_i n_{D,i} \quad (2)$$

where ρ and ρ_i are the density of the mixture and the density of the pure components, respectively; x_i represents the mole fraction of the component i, and n_D and $n_{D,i}$ are the refractive index of the mixture and the pure component, respectively. The uncertainties for the excess molar volume and refractive index deviation are $\pm\ 2\cdot10^{-2}$ cm^3·mol^{-1} and $4\cdot10^{-5}$, respectively.

The speed of sound, u, is related to the isentropic compressibility, κ_s, by the Laplace equation

$$\kappa_s = -V_m^{-1}(\partial V_m/\partial\rho)_S = \rho^{-1}u^{-2} = V_m/(M_m u^2) \quad (3)$$

where V_m is the molar volume and M_m is the molar mass of the mixture.

To achieve agreement with the other thermodynamic quantities, it is appropriate to shift from the volume-intensive κ_s to the mole-intensive quantity $K_{S,m}$[7,8]

$$K_{S,m} = -(\partial V_m/\partial\rho)_S = V_m\kappa_s = V_m^{\ 2}/(M_m u^2) \quad (4)$$

where $K_{S,m}$ is the molar isentropic compressibility.

Experimental values for the speed of sound are reported in Table 2 together with derived values of the isentropic compression and its excess, $K^E_{S,m}$. The excess molar isentropic compressibility, $K^E_{S,m}$, is calculated by the following equation

Table 3. Fitting Parameters and Root-Mean-Square Deviations (rmsd) for Binary Mixtures at $T = 313.15$ K

	B_0	B_1	B_2	B_3	rmsd
Cyclopentane (1) + Benzene (2)					
V^E/cm^3·mol^{-1}	1.3697	0.1632			0.005
Δn_D	−0.01137	−0.00079	0.00258		0.00006
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	0.0046	0.0024			0.00002
Cyclopentane (1) + Toluene (2)					
V^E/cm^3·mol^{-1}	0.2432	0.2875			0.004
Δn_D	0.01210	−0.00053	0.0362		0.00008
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	−0.0002	0.0017			0.00002
Cyclopentane(1) + Ethylbenzene(2)					
V^E/cm^3·mol^{-1}	0.0708	0.1338			0.003
Δn_D	0.02397	0.00521	−0.00101		0.00008
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	−0.0025	0.0004			0.00003
Cyclohexane (1) + Benzene (2)					
V^E/cm^3·mol^{-1}	2.5564	0.1446	0.1407		0.008
Δn_D	−0.02791	0.00266	−0.00185		0.00006
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	0.0141	0.0026			0.00004
Cyclohexane (1) + Toluene(2)					
V^E/cm^3·mol^{-1}	2.3060	0.4877	−0.0207	0.2174	0.008
Δn_D	−0.01065	−0.00178	0.00062		0.00004
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	0.0111	0.0040			0.00004
Cyclohexane (1) + Ethylbenzene (2)					
V^E/cm^3·mol^{-1}	1.9768	0.5894			0.006
Δn_D	0.00091	−0.00080	−0.00414	0.00453	0.00009
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	0.0077	0.0037			0.00004
Methylcyclohexane (1) + Benzene (2)					
V^E/cm^3·mol^{-1}	2.3030	0.0228	0.1555		0.007
Δn_D	−0.03956	0.00846	−0.00087		0.00005
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	0.0122	0.0019			0.00003
Methylcyclohexane (1) + Toluene (2)					
V^E/cm^3·mol^{-1}	1.639	0.2006			0.008
Δn_D	−0.02040	0.00120	0.00040		0.00008
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	0.9834	0.2386	0.2810		0.00003
Methylcyclohexane (1) + Ethylbenzene (2)					
V^E/cm^3·mol^{-1}	0.9834	0.2386	0.2810		0.004
Δn_D	−0.00797	0.00091	−0.00236		0.00004
$K^E_{S,m}$/m^3·TPa^{-1}·mol^{-1}	0.0046	0.0024			0.00002

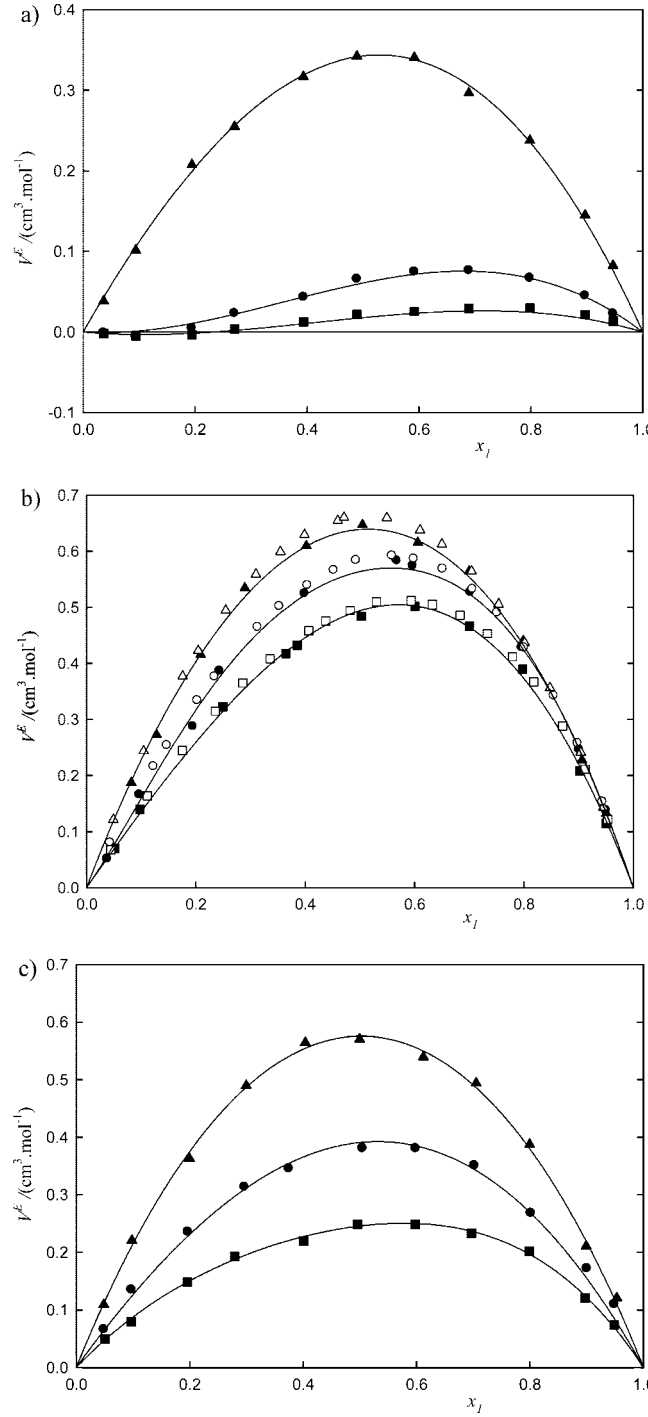

Figure 1. Excess molar volumes, V^E, from the Redlich−Kister equation plotted against mole fraction for the binary mixtures: cycloalkane (1) + aromatic (2) at $T = 313.15$ K for (a) cyclopentane, (b) cyclohexane (from ref 5), and (c) methylcyclohexane. Closed symbols correspond to experimental data: ▲, benzene; ●, toluene; ■, ethylbenzene, and open symbols correspond to literature data.

$$K^{E}_{S,m} = K_{S,m} - K^{id}_{S,m} \quad (5)$$

where $K^{id}_{S,m}$ is defined by the approach developed by Benson and Kiyohara[9]

$$K^{id}_{S,m} = \sum_i x_i \left[K^*_{S,i} + T\frac{(E^*_{p,i})^2}{C^*_{p,i}} \right] - T\left[\frac{\left(\sum_i x_i E^*_{p,i}\right)^2}{\sum_i x_i C^*_{p,i}} \right] \quad (6)$$

where $K^*_{S,i}$ is the product of the molar volume, V^*_i, and the isentropic compressibility, $\kappa^*_{S,i}$, of the pure component, i. The molar isobaric expansion of pure component i, $E^*_{p,i}$, is the product of the molar volume and the isobaric expansibility $\alpha^*_{p,i}$ ($\alpha^*_{p,i} = -(1/\rho)(\partial\rho/\partial T)_P$), and $C^*_{p,i}$ is the molar isobaric heat capacity of the pure component, i. The uncertainty for this excess property is $\pm\ 7\cdot10^{-5}$ $m^3\cdot TPa^{-1}\cdot mol^{-1}$. The values of $\alpha^*_{p,i}$ and $C^*_{p,i}$ are obtained from the literature.[10]

The excess and deviation properties at studied temperature were fitted to a Redlich−Kister-type[11] equation

$$\Delta Q_{12} = x_1 x_2 \sum_{p=0}^{M} B_p (x_1 - x_2)^p \quad (7)$$

where ΔQ_{12} is the excess property, B_p is the fitting parameter, and M is the degree of the polynomic expansion, which was optimized using the F test.[12] The fitting parameters are given in Table 3 together with the root-mean-square deviations, σ

$$\sigma = \left\{ \frac{\sum_i^{n_{dat}} (z_{exptl} - z_{calcd})^2}{n_{dat}} \right\}^{1/2} \quad (8)$$

where z_{exptl}, z_{calcd}, and n_{dat} are the values of the experimental and calculated properties and the number of experimental data, respectively.

Figure 1 shows the fitted curve of excess molar volume values of the 9 binary systems containing cycloalkanes and aromatic compounds (cyclopentane (1), or cyclohexane (1), or methylcyclohexane (1) + benzene (2), + toluene (2), + ethylbenzene (2)) at $T = 313.15$ K. The systems present a similar behavior for this physical property.

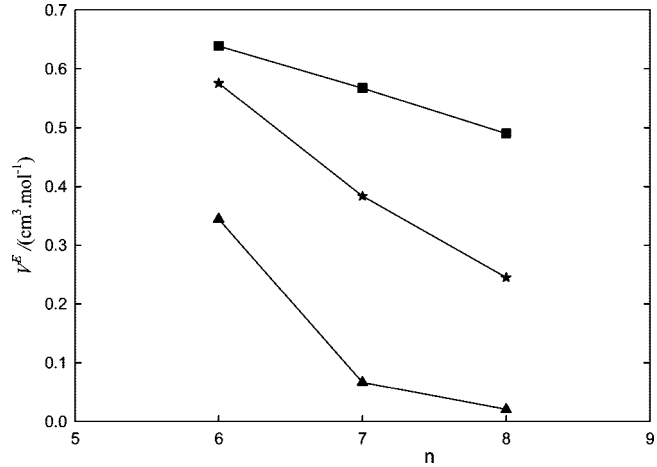

Figure 2. Variation of equimolar excess molar volumes, V^E, for the binary mixtures cycloalkane (1) + aromatic (2) against the number of carbon atoms, n, in the aromatic compound at $T = 313.15$ K: ▲, cyclopentane; ■, cyclohexane; ★, methylcyclohexane.

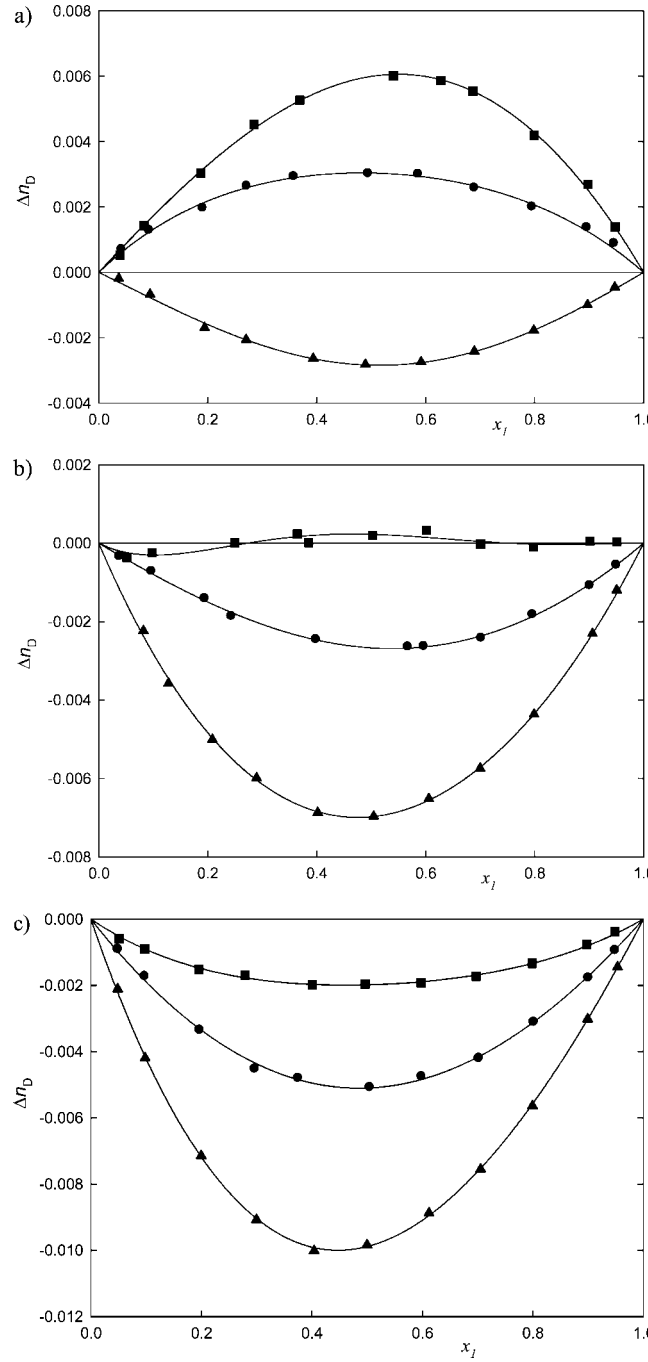

Figure 3. Refractive index deviations, Δn_D, from the Redlich−Kister equation plotted against mole fraction for the binary mixtures: cycloalkane (1) + aromatic (2) at $T = 313.15$ K for (a) cyclopentane, (b) cyclohexane, and (c) methylcyclohexane. Closed symbols correspond to experimental data: ▲, benzene; ●, toluene; ■, ethylbenzene.

As can be observed, in Figure 1, the excess molar volume is positive over the entire composition range for all investigated mixtures, with a maximum between $x_1 = 0.5$ and $x_1 = 0.6$, except for the systems cyclopentane (1) + toluene (2) and cyclopentane (1) + ethylbenzene (2). The excess molar volumes of these two systems show a sigmoid kind of behavior. Such behavior is the result of several opposing effects. For the systems containing benzene, the excess presents a maximum over $x_1 = 0.5$; for those systems containing toluene, the maximum is over

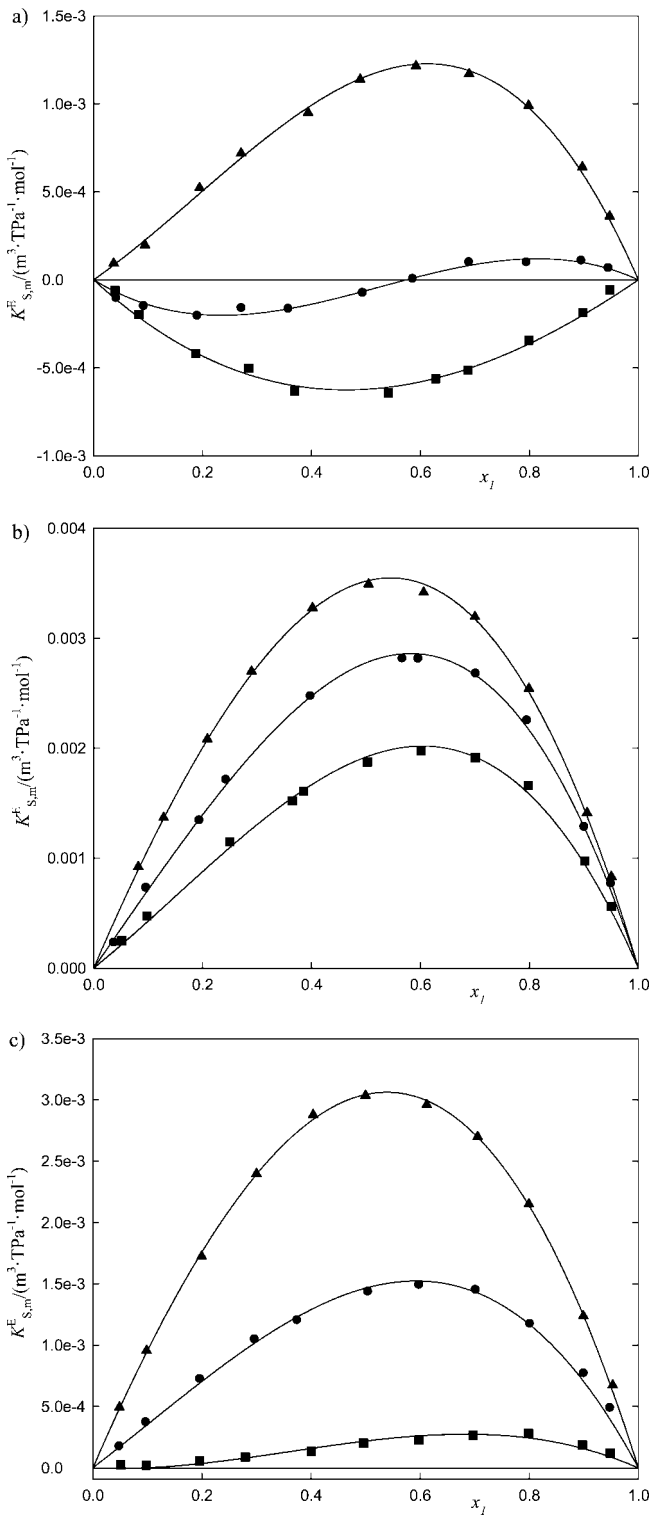

Figure 4. Variation of deviation in isentropic compressibility, Δk_s, from the Redlich−Kister equation plotted against mole fraction for the binary mixtures: cycloalkane (1) + aromatic (2) at $T = 313.15$ K for (a) cyclopentane, (b) cyclohexane and, (c) methylcyclohexane. Closed symbols correspond to experimental data: ▲, benzene; ●, toluene; ■, ethylbenzene.

$x_1 = 0.55$, except for the system with cyclopentane, which presents a minimum over $x_1 = 0.1$ and a maximum over $x_1 = 0.7$. For the binary systems with ethylbenzene, the excess molar volume presents a maximum over $x_1 = 0.6$, except for the system cyclopentane (1) + ethylbenzene (2), which presents a minimum over $x_1 = 0.1$ and a maximum over $x_1 = 0.7$. The comparison with available literature data[5,6] was shown in this Figure, and very good results were obtained. When the aliphatic chain of the aromatic compounds increases, the excess molar volume decreases (Figure 2) for all studied binary systems.

Figure 3 shows the refractive index deviation for all studied systems over the entire composition range at $T = 313.15$ K. In this excess property, all systems present a small deviation from ideal behavior, above all of the binary system cyclohexane (1) + ethylbenzene (2). All systems present a minimum at approximately $x_1 = 0.5$, except for the binary systems cyclopentane (1) + toluene (2), + ethylbenzene (2), which present a maximum at about $x_1 = 0.5$.

The excess molar isentropic compressions are plotted in Figure 4 over the entire composition range at $T = 313.15$ K. As in case of the refractive index deviation, this excess presents a small deviation from ideal behavior for all binary systems, above all the systems cyclopentane (1) + toluene (2) and the system methylcyclohexane (1) + ethylbenzene (2).

Conclusions

In this article, we present the densities, speeds of sound, and refractive indices of 9 binary systems containing cycloalkanes with aromatic compounds (cyclopentane (1), or cyclohexane (1), or methylcyclohexane (1) + benzene (2), + toluene (2), + ethylbenzene (2)) at $T = 313.15$ K and at atmospheric pressure over the whole composition range.

The excess properties (excess molar volume, refractive index deviations, and excess molar isentropic compressions) have been determined from experimental data for all binary systems, and these data have been fitted to the Redlich−Kister equation.

The systems cyclopentane (1) + toluene (2), and + ethylbenzene (2) show a sigmoid kind of behavior for the excess molar volume. The other studied systems present a maximum for this excess. It was possibly observed that when the aliphatic chain of the aromatic compounds increases, the excess molar volume decreases for all studied binary systems.

The excess properties of the refractive index and of the isentropic compression show a small deviation from ideal behavior for all systems.

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