

Refractive Indices and Surface Tensions of Binary Mixtures of Isoamyl Acetate, Ethyl Caproate, Ethyl Benzoate, Isoamyl Butyrate, Ethyl Phenylacetate, and Ethyl Caprylate with Ethanol at (288.15, 298.15, 308.15, and 318.15) K

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Refractive indices and surface tensions for six binary mixtures formed by the flavor esters (isoamyl acetate, ethyl caproate, ethyl benzoate, isoamyl butyrate, ethyl phenylacetate, and ethyl caprylate) with ethanol over the whole composition range were measured at four different temperatures (288.15, 298.15, 308.15, and 318.15) K and atmospheric pressure. Refractive indices were measured using a digital Abbe-type refractometer. Surface tensions were determined using the Wilhelmy-plate method. From experimental data, deviations in the refractive index Δn were calculated both on a mole fraction basis and on a volume fraction basis. The deviations in the molar refraction ΔR and the deviations in the surface tension $\Delta\sigma$ were calculated on a mole fraction basis. The binary data of these deviations were correlated as a function of the mole fraction using the Redlich–Kister equation.

Introduction

The thermophysical properties involving flavor esters are of increasing interest due to their wide usage in flavoring, perfumery, artificial essences, and cosmetics. Among these properties, surface tension is a basic property, related to vapor–liquid interfacial effects that must be known to analyze heat and mass transfer of working fluids. Several studies for binary or ternary mixtures on surface tension have been conducted recently, but there are few data in the literature for the mixtures involving such ester compounds.^{1,2}

In continuation of our previous work,³ we report in this paper the measurements of surface tension and refractive index data for the binary mixtures of six flavor esters: isoamyl acetate, ethyl caproate, ethyl benzoate, isoamyl butyrate, ethyl phenylacetate, and ethyl caprylate, respectively, with ethanol at temperatures of (288.15, 298.15, 308.15, and 318.15) K and at atmospheric pressure. From the experimental results, the deviations in the refractive index from the mole fraction average $\Delta_x n_D$ and from the volume fraction average $\Delta_\phi n_D$ have been calculated. The molar refraction deviations ΔR on a mole fraction basis were derived. The deviations in the surface tension from the mole fraction average $\Delta\sigma$ were calculated. To our knowledge, no such data in the literature are available for the binary systems studied here.

Experimental Section

Materials. The chemicals used were of analytical grade and were used without further purification. The purity of these chemicals was analyzed by gas chromatography (Perkin-Elmer Autosystem). The mass fraction purities as determined by the major peak areas on gas chromatography together with the sources and Chemical Abstract Service registration numbers (CASRN) of the chemicals are given in Table 1. The purity of solvents was further ascertained by comparing their densities, refractive indices, and surface tensions at a temperature of 298.15 K with the corresponding values reported in the literature

Table 1. Sources and Mass Fraction (*W*) Purities of the Esters Used in This Study

compounds	molecular formula	sources	CASRN	100 <i>W</i>
isoamyl acetate	C ₇ H ₁₄ O ₂	Tedia (USA)	123-92-2	99.3
ethyl caproate	C ₈ H ₁₆ O ₂	Acros (USA)	123-66-0	99.7
ethyl benzoate	C ₉ H ₁₀ O ₂	Acros (USA)	93-89-0	99.8
isoamyl butyrate	C ₉ H ₁₈ O ₂	Acros (USA)	106-27-4	99.4
ethyl phenylacetate	C ₁₀ H ₁₂ O ₂	Acros (USA)	101-97-3	99.4
ethyl caprylate	C ₁₀ H ₂₀ O ₂	Acros (USA)	106-32-1	99.5
ethanol	C ₂ H ₆ O	Merck (Germany)	64-17-5	99.9

Table 2. Comparison of Measured Densities ρ , Refractive Indices n_D , and Surface Tensions σ of Pure Components with Literature Values at 298.15 K

compounds	ρ g·cm ⁻³		n_D		σ mN·m ⁻¹	
	exptl	lit	exptl	lit	exptl	lit
isoamyl acetate	0.86791	0.86621 ⁴ 0.87601 ⁵ 0.8664 ⁶	1.39836	1.3981 ⁶	24.3	24.24 ^{7 a}
ethyl caproate	0.86629	0.86664 ⁸	1.40504	1.4077 ^{9 b}	25.4	25.32 ^{7 a}
ethyl benzoate	1.04142	1.041 ¹⁰ 1.0421 ¹¹	1.50328	1.5046 ¹¹	34.6	34.8 ¹²
isoamyl butyrate	0.85869	0.86204 ⁵	1.40815	0.8600 ^{7 a}	24.7	24.92 ^{7 a}
ethyl phenylacetate	1.02696		1.49513		34.6	
ethyl caprylate	0.86215	0.86219 ¹³	1.41560	1.4150 ¹³	26.4	27.03 ^{7 a}
ethanol	0.78500	0.78493 ⁶ 0.78515 ¹⁴ 0.78506 ¹⁵	1.35941	1.3593 ¹⁴ 1.3595 ¹⁵	22.3	21.68 ⁶ 21.74 ¹⁴ 21.8 ¹⁶

^a Data obtained from linear interpolation. ^b Data at 20 °C.

as shown in Table 2. Experimental densities were measured with an Anton Paar DMA-5000 vibrating-tube density meter (Anton-Paar, Graz, Austria) with uncertainties of $5 \cdot 10^{-6}$ g·cm⁻³, which was thermostatically controlled to within ± 0.01 K in all measured variables included.

Apparatus and Procedure. Samples were prepared by mass in a 50 cm³ Erlenmeyer flask provided with a ground glass joint stopper, using a Precisa 262SMA balance with an uncertainty of $\pm 3 \cdot 10^{-5}$ g. Refractive indices (n_D) were measured with a digital Abbe refractometer RX-5000 (ATAGO, Tokyo, Japan),

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Table 9. Coefficients and Standard Deviations δ of $\Delta_v n_D$, $\Delta_\phi n_D$, ΔR , and $\Delta\sigma$ for the Binary Mixtures from $T = 288.15$ K to $T = 318.15$ K

Y	T/K	a_0	a_1	a_2	a_3	a_4	a_5	$\delta \cdot 10^4$
Isoamyl Acetate (1) + Ethanol (2)								
$\Delta_v n_D$	288.15	0.03384	-0.01419	0.00888	-0.00519			1.12
	298.15	0.03291	-0.01318	0.01490	0.00464			1.85
	308.15	0.03215	-0.01378	0.00742	-0.00215	-0.00166		0.10
	318.15	0.03095	-0.01336	0.00699	-0.00063	-0.00296		0.12
$\Delta_\phi n_D$	288.15	-0.00041	0.00033	0.00057	-0.00056			0.10
	298.15	-0.00095	0.00069	0.00102	0.00018	-0.00190		0.08
	308.15	-0.00142	0.00084	0.00109	0.00119	-0.00317		0.11
	318.15	-0.00187	0.00096	0.00084	0.00254	-0.00446		0.15
$\Delta R/\text{cm}^3 \cdot \text{mol}^{-1}$	288.15	0.09420	0.03191	0.03994	-0.03050			6.20
	298.15	0.08796	0.04495	0.09049	-0.01381	-0.12788		5.03
	308.15	0.09524	0.05368	0.08460	0.00500	-0.15456		6.33
	318.15	0.10723	0.05632	0.08991	0.06900	-0.23872		7.77
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	288.15	0.45111	-0.31345	0.12900	-0.79861	1.20178		53.8
	298.15	0.01266	-0.04704	-0.38263	0.67654	1.17951	-1.70164	37.0
	308.15	-0.18703	0.12547	-0.55478	1.07098	0.72700	-1.57397	47.4
	318.15	-0.38966	0.40876	-0.62988	0.51681			33.2
Ethyl Caproate (1) + Ethanol (2)								
$\Delta_v n_D$	288.15	0.04394	-0.02076	0.01100	-0.00696	0.00411		0.10
	298.15	0.04293	-0.02033	0.01098	-0.00625	0.00332		0.12
	308.15	0.04214	-0.02003	0.01101	-0.00536	0.00179		0.10
	318.15	0.04096	-0.01969	0.01113	-0.00379			0.11
$\Delta_\phi n_D$	288.15	-0.00032	0.00022	0.00165	-0.00106			0.11
	298.15	-0.00078	0.00044	0.00134	-0.00041			0.09
	308.15	-0.00118	0.00055	0.00134	0.00040	-0.00127		0.09
	318.15	-0.00162	0.00047	0.00156	0.00204	-0.00280		0.15
$\Delta R/\text{cm}^3 \cdot \text{mol}^{-1}$	288.15	0.11483	0.03178	0.10486				6.59
	298.15	0.11417	0.04148	0.09308				9.58
	308.15	0.12699	0.03994	0.06269	0.03709			11.9
	318.15	0.13707	0.03764	0.14316	0.10510	-0.17537		11.0
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	288.15	2.47764	-1.90377	0.85328	0.05690	0.43251		39.6
	298.15	2.16931	-1.80638	0.77538	0.08821	0.42357		34.5
	308.15	2.02131	-1.73155	0.91882				26.3
	318.15	1.97276	-1.73155	0.85703				61.9
Ethyl Benzoate (1) + Ethanol (2)								
$\Delta_v n_D$	288.15	0.12823	-0.05612	0.02294	-0.01804	0.01463		0.38
	298.15	0.12700	-0.05571	0.02291	-0.01549	0.01038		0.27
	308.15	0.12571	-0.05516	0.02283	-0.01381	0.00680		0.19
	318.15	0.12380	-0.05461	0.02418	-0.01159			0.28
$\Delta_\phi n_D$	288.15	0.00636	-0.00485	0.00139	-0.00742	0.00997		0.33
	298.15	0.00559	-0.00516	0.00473	-0.00428			0.57
	308.15	0.00515	-0.00463	0.00155	-0.00348	0.00237		0.16
	318.15	0.00440	-0.00467	0.00086	-0.00130			0.24
$\Delta R/\text{cm}^3 \cdot \text{mol}^{-1}$	288.15	0.41779	-0.11979	0.01014	-0.12218	0.30846		12.4
	298.15	0.40843	-0.11925	0.02086	-0.05984	0.13223		8.12
	308.15	0.40919	-0.10836	0.02209	-0.04024			10.7
	318.15	0.40071	-0.10854	0.05448	0.01892	-0.21507		13.3
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	288.15	2.28621	-3.57438	-1.40988	-4.01779	3.08992	5.56058	93.4
	298.15	1.64256	-3.40470	-0.98967	-4.28467	1.09094	5.50462	78.4
	308.15	1.23204	-3.29476	-0.76888	-4.31770	-0.61115	5.51757	141.8
	318.15	1.03325	-3.36996	-0.75734	-4.05326	-1.94113	5.05729	146.4
Isoamyl Butyrate (1) + Ethanol (2)								
$\Delta_v n_D$	288.15	0.05077	-0.02474	0.01300	-0.00979	0.00572		0.09
	298.15	0.04986	-0.02396	0.01217	-0.00996	0.00512		0.39
	308.15	0.04909	-0.02368	0.01251	-0.00793	0.00350		0.08
	318.15	0.04790	-0.02249	0.01283	-0.01010	0.00054	0.00527	0.09
$\Delta_\phi n_D$	288.15	-0.00020	0.00137	0.00013	-0.00105			0.08
	298.15	-0.00057	0.00177	-0.00043	-0.00058			0.12
	308.15	-0.00093	0.00190	-0.00125	0.00079			0.18
	318.15	-0.00146	0.00213	-0.00004	0.00257	-0.00411		0.30
$\Delta R/\text{cm}^3 \cdot \text{mol}^{-1}$	288.15	0.13883	0.10643	0.03706	-0.16765	0.06288	0.21365	6.07
	298.15	0.14250	0.13421	0.04200	-0.19029	0.00954	0.22793	4.78
	308.15	0.15150	0.14744	0.04626	-0.21253	-0.06667	0.29692	5.94
	318.15	0.16356	0.17445	0.07921	-0.22743	-0.18942	0.39695	11.6
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	288.15	1.40303	-1.43060	1.34843	-0.69479			36.7
	298.15	1.06118	-1.13238	1.21290	-0.73059			52.9
	308.15	1.03615	-1.06733	1.29256	-0.85184			43.1
	318.15	0.97710	-1.02749	1.22291	-0.79935			68.7

Table 9 (Continued)

Y	T/K	a_0	a_1	a_2	a_3	a_4	a_5	$\delta \cdot 10^4$
Ethyl Phenylacetate (1) + Ethanol (2)								
$\Delta_{\phi}n_D$	288.15	0.13025	-0.06212	0.02559	-0.00957	0.01616	-0.01115	0.28
	298.15	0.12906	-0.06146	0.02560	-0.01097	0.01373	-0.00847	0.32
	308.15	0.12812	-0.06038	0.02507	-0.01683	0.01236		0.40
	318.15	0.12652	-0.05977	0.02521	-0.01631	0.00957		0.50
$\Delta_{\phi}n_D$	288.15	0.00374	-0.00263	-0.00115	-0.00325	0.00868		0.38
	298.15	0.00337	-0.00269	-0.00093	-0.00261	0.00638		0.35
	308.15	0.00301	-0.00288	-0.00138	-0.00213	0.00526		0.35
	318.15	0.00248	-0.00296	0.00054	-0.00162			0.50
$\Delta R/cm^3 \cdot mol^{-1}$	288.15	0.31528	-0.05673	-0.10927	0.02234	0.39728		17.1
	298.15	0.31514	-0.06250	-0.10365	0.01158	0.28363		17.7
	308.15	0.32503	-0.07281	-0.13054	0.00157	0.22431		16.6
	318.15	0.32897	-0.07655	-0.06499				21.8
$\Delta\sigma/mN \cdot m^{-1}$	288.15	1.01160	-0.75958	0.26447	1.30428	2.27879	-4.25214	132.3
	298.15	0.23154	-0.12920	0.78931	-1.95543			144.5
	308.15	-0.26715	0.03999	-0.16987	-2.18170			121.2
	318.15	-0.57368	0.07444	-1.25362	-2.39765			130.2
Ethyl Caprylate (1) + Ethanol (2)								
$\Delta_{\phi}n_D$	288.15	0.06122	-0.03346	0.01828	-0.00947	0.00670	-0.00378	0.06
	298.15	0.06049	-0.03295	0.01796	-0.00993	0.00590	-0.00231	0.03
	308.15	0.06000	-0.03259	0.01785	-0.01074	0.00465		0.07
	318.15	0.05912	-0.03212	0.01734	-0.00980	0.00338		0.08
$\Delta_{\phi}n_D$	288.15	-0.00058	0.00032	-0.00014	0.00052			0.08
	298.15	-0.00088	0.00030	0.00013	0.00128	-0.00150		0.09
	308.15	-0.00118	0.00039	0.00011	0.00198	-0.00278		0.15
	318.15	-0.00145	0.00046	-0.00010	0.00285	-0.00403		0.21
$\Delta R/cm^3 \cdot mol^{-1}$	288.15	0.13401	0.03493	0.03346	-0.09497	-0.09597		6.32
	298.15	0.14299	0.02289	0.00599	-0.05130	-0.09574		8.92
	308.15	0.16074	0.02761	-0.00388	-0.02980	-0.12600		12.9
	318.15	0.18711	0.03527	-0.01814	-0.01441	-0.19003		12.4
$\Delta\sigma/mN \cdot m^{-1}$	288.15	3.87051	-2.19294	1.16288	-0.82642	0.52225		27.6
	298.15	3.72614	-2.07023	0.94213	-0.82452	0.52666		57.1
	308.15	3.58066	-2.03152	1.10774	-0.73883			34.8
	318.15	3.59324	-2.02934	1.05296	-0.66499			58.9

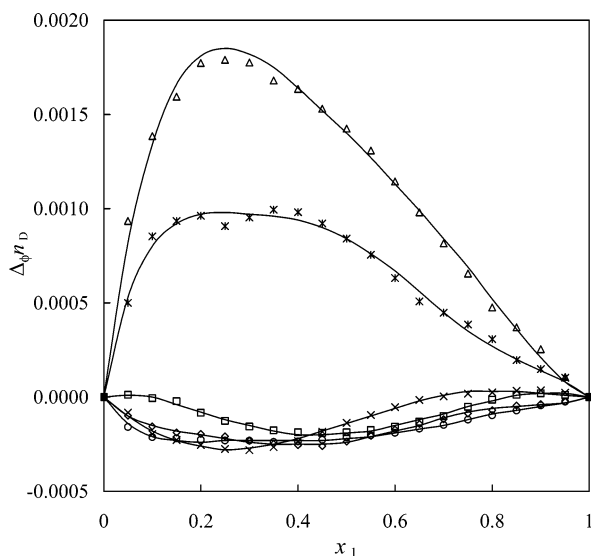


Figure 1. Change of surface tensions $\Delta_{\phi}n_D$ with mole fraction x_1 at $T = 298.15$ K: \diamond , isoamyl acetate + ethanol; \square , ethyl caproate + ethanol; \triangle , ethyl benzoate + ethanol; \times , isoamyl butyrate + ethanol; $*$, ethyl phenylacetate + ethanol; \circ , ethyl caprylate + ethanol. Solid curves were calculated from the Redlich–Kister equation.

where n is the number of experimental points and p is the number of adjustable parameters.

For the whole composition range, the $\Delta_{\phi}n_D$ values are positive for the mixtures of ethyl benzoate + ethanol and ethyl phenylacetate + ethanol, while the negative values were found for the mixtures of isoamyl acetate + ethanol and ethyl caprylate

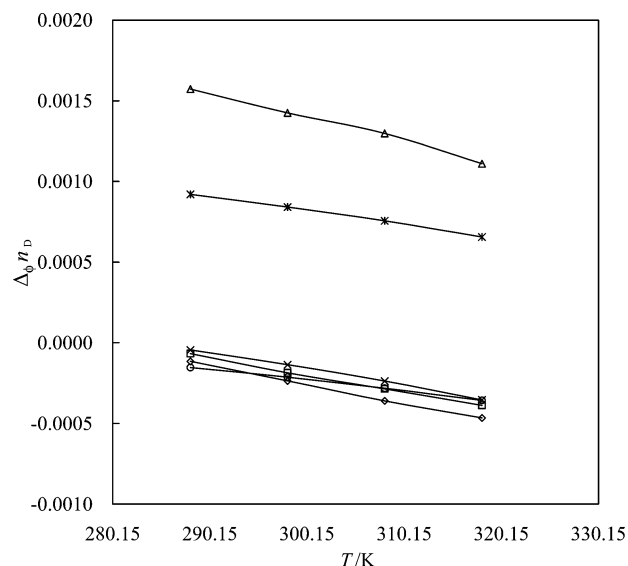


Figure 2. Variation of refractive indices $\Delta_{\phi}n_D$ at $x = 0.5$ from $T = 288.15$ K to $T = 318.15$ K: \diamond , isoamyl acetate + ethanol; \square , ethyl caproate + ethanol; \triangle , ethyl benzoate + ethanol; \times , isoamyl butyrate + ethanol; $*$, ethyl phenylacetate + ethanol; \circ , ethyl caprylate + ethanol.

+ ethanol. The $\Delta_{\phi}n_D$ values are negative for the mixtures ethyl caproate + ethanol and isoamyl butyrate + ethanol over the whole composition range except that some positive values were found at the temperatures of 288.15 K and 298.15 K. Figure 1 shows the results of $\Delta_{\phi}n_D$ for the six binary systems at $T = 298.15$ K. The values of $\Delta_{\phi}n_D(x = 0.5)$ at 298.15 K show the

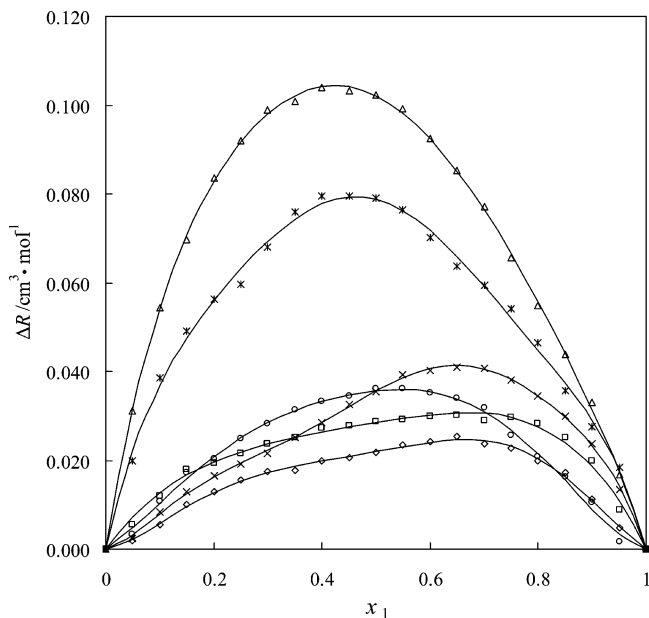


Figure 3. Change of ΔR with mole fraction x_1 at $T = 298.15$ K: \diamond , isoamyl acetate + ethanol; \square , ethyl caproate + ethanol; \triangle , ethyl benzoate + ethanol; \times , isoamyl butyrate + ethanol; $*$, ethyl phenylacetate + ethanol; \circ , ethyl caprylate + ethanol. Solid curves were calculated from the Redlich–Kister equation.

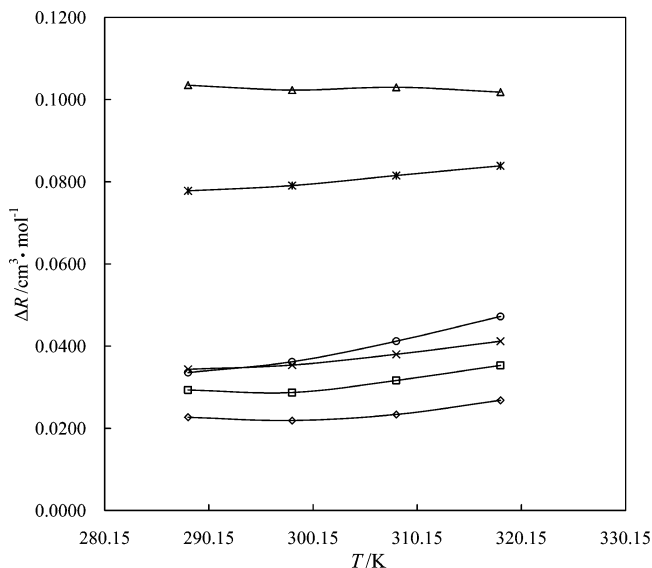


Figure 4. Variation of refractive indices ΔR at $x = 0.5$ from $T = 288.15$ K to $T = 318.15$ K: \diamond , isoamyl acetate + ethanol; \square , ethyl caproate + ethanol; \triangle , ethyl benzoate + ethanol; \times , isoamyl butyrate + ethanol; $*$, ethyl phenylacetate + ethanol; \circ , ethyl caprylate + ethanol.

order as ethyl benzoate + ethanol > ethyl phenylacetate + ethanol > isoamyl butyrate + ethanol > ethyl caproate + ethanol > ethyl caprylate + ethanol > isoamyl acetate + ethanol. Figure 2 plots the values of $\Delta\phi_{nD}(x = 0.5)$ from $T = 288.15$ K to $T = 318.15$ K for all of the systems. The values of $\Delta\phi_{nD}(x = 0.5)$ decrease systematically with a rise in temperature. The values of $\Delta\phi_{nD}(x = 0.5)$ vary from -0.00047 to 0.00159 .

The values of molar refraction deviation ΔR are positive for all of the systems. The ΔR values are graphically represented as a function of mole fraction at 298.15 K in Figure 3. The values of $\Delta R(x = 0.5)$ at 298.15 K show the order as ethyl benzoate + ethanol > ethyl phenylacetate + ethanol > ethyl caprylate + ethanol > isoamyl butyrate + ethanol > ethyl caproate + ethanol > isoamyl acetate + ethanol. Figure 4 plots the values of $\Delta R(x = 0.5)$ from $T = 288.15$ K to $T = 318.15$

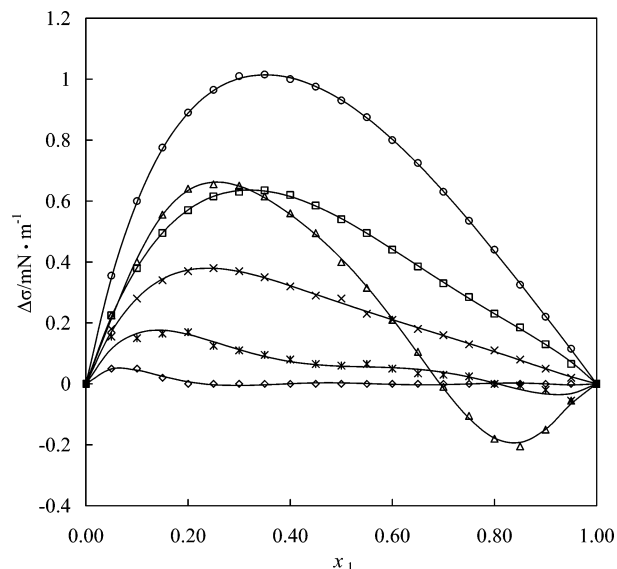


Figure 5. Change of surface tensions $\Delta\sigma$ with mole fraction x_1 at $T = 298.15$ K: \diamond , isoamyl acetate + ethanol; \square , ethyl caproate + ethanol; \triangle , ethyl benzoate + ethanol; \times , isoamyl butyrate + ethanol; $*$, ethyl phenylacetate + ethanol; \circ , ethyl caprylate + ethanol. Solid curves were calculated from the Redlich–Kister equation.

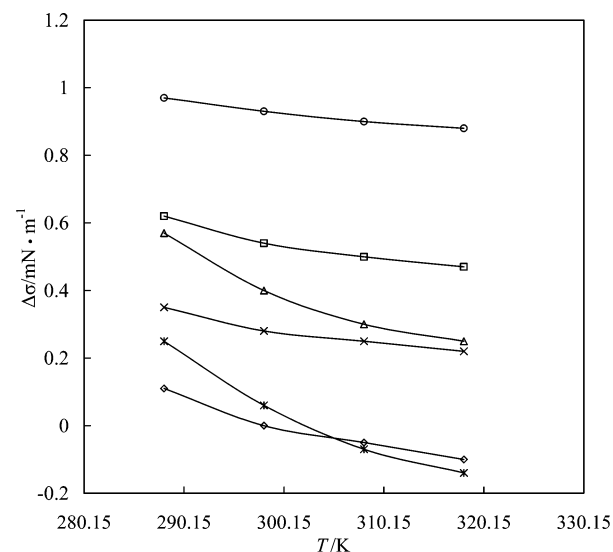


Figure 6. Variation of refractive indices $\Delta\sigma$ at $x = 0.5$ from $T = 288.15$ K to $T = 318.15$ K: \diamond , isoamyl acetate + ethanol; \square , ethyl caproate + ethanol; \triangle , ethyl benzoate + ethanol; \times , isoamyl butyrate + ethanol; $*$, ethyl phenylacetate + ethanol; \circ , ethyl caprylate + ethanol.

K for all of the systems. The values of $\Delta R(x = 0.5)$ show no clear temperature dependence. The values of $\Delta R(x = 0.5)$ vary from 0.0219 $\text{cm}^3\cdot\text{mol}^{-1}$ to 0.1035 $\text{cm}^3\cdot\text{mol}^{-1}$.

The values of surface tension deviation $\Delta\sigma$ are graphically represented as a function of mole fraction at 298.15 K in Figure 5. The values of $\Delta\sigma$ are positive for all of the systems except for some negative values that were found for the mixtures of ethyl benzoate + ethanol and ethyl phenylacetate + ethanol in the region of higher ester concentrations. The values of $\Delta\sigma(x = 0.5)$ at 298.15 K show the order as isoamyl acetate + ethanol < ethyl phenylacetate + ethanol < isoamyl butyrate + ethanol < ethyl benzoate + ethanol < ethyl caproate + ethanol < ethyl caprylate + ethanol. Figure 6 plots the values of $\Delta\sigma(x = 0.5)$ from $T = 288.15$ K to $T = 318.15$ K for all of the systems. The values of $\Delta\sigma$ decrease with increasing temperature for all of the mixtures. The values of $\Delta\sigma(x = 0.5)$ vary from -0.14 $\text{mN}\cdot\text{m}^{-1}$ to 0.97 $\text{mN}\cdot\text{m}^{-1}$.

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