Refractive Indices and Surface Tensions of Binary Mixtures of Ethyl Acetoacetate, Ethyl Isovalerate, Methyl Benzoate, Benzyl Acetate, Ethyl Salicylate, and Benzyl Propionate with Ethanol at (288.15, 298.15, 308.15, and 318.15) K

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Refractive indices and surface tensions for binary mixtures of six flavor esters (ethyl acetoacetate, ethyl isovalerate, methyl benzoate, benzyl acetate, ethyl salicylate, and benzyl propionate) 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. Deviations in the refractive index Δn_D and deviations in the surface tension $\Delta\sigma$ from volume fraction average for the binary mixtures were derived from experimental data. The binary data of Δn_D and $\Delta\sigma$ were correlated as a function of mole fraction using the Redlich−Kister equation.

Introduction

The surface tension of mixtures is an important thermophysical property in heat and mass transfer processes such as liquid−liquid extraction, gas absorption, and distillation. The surface tension is also a property that contains at least implicit information on molecular interaction and could thus be used as a qualitative guide to understand the behavior of mixtures. The studies dealing with surface tension of binary mixtures are extensive, but the systems that involve esters are of increasing interest due to their wide usage in flavoring, perfumery, artificial essences, and cosmetics.[1,2]

Among different types of flavor esters, ethyl acetoacetate, ethyl isovalerate, methyl benzoate, benzyl acetate, ethyl salicylate, and benzyl propionate are commonly used. On the other hand, ethanol is an important alcohol component in the flavor and fragrance process manufacturing industries. 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 these six flavor esters respectively with ethanol at the temperatures of (288.15, 298.15, 308.15, and 318.15) K and atmospheric pressure. The experimental results are used to calculate deviations in the refractive index Δn_D and deviations in the surface tension $\Delta\sigma$ from volume fraction average. To the best of our knowledge, no such data in the earlier literature are available for the above-mentioned mixtures.

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) using a FID detector. The mass fraction purities as determined by the major peak areas on gas chromatography together with the sources and Chemical Abstract Service Registry Numbers (CASRN) of 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 as shown in Table 2. Experimental densities were measured with an Anton Paar DMA-5000 vibrating-tube density meter (Anton-Paar, Graz, Austria) with an uncertainty of 5×10^{-6} g·cm^{-3}, which was thermostatically controlled to within ± 0.01 K.

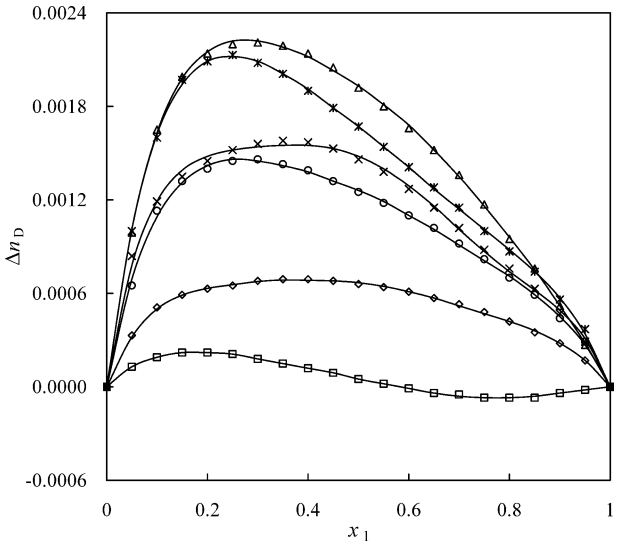

Figure 1. Change of refractive indices Δn_D with mole fraction x_1 at T = 298.15 K: ◇, ethyl acetoacetate + ethanol; □, ethyl isovalerate + ethanol; △, methyl benzoate + ethanol; ×, benzyl acetate + ethanol; ∗, ethyl salicylate + ethanol; ○, benzyl propionate + ethanol. Solid curves were calculated from the Redlich−Kister equation.

Apparatus and Procedure. Samples were prepared by mass in a 50 cm^3 Erlenmeyer flask provided with a ground glass joint stopper, using a Precisa 262SMA balance with an uncertainty of ± 3×10^{-5} g. Refractive indices n_D were measured with a digital Abbe refractometer RX-5000 (ATAGO, Tokyo, Japan), which works at a wavelength of 589 nm corresponding to the D-line of sodium. The temperature was controlled to ± 0.05 K with circulating thermostat water to a jacketed sample vessel. Calibration was performed periodically under atmospheric pressure using double-distilled water. The uncertainty of the

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

compounds	molecular formula	sources	CASRN[a]	100 *w*
ethyl acetoacetate	$C_6H_{10}O_3$	Acros (USA)	141-97-9	99.2
ethyl isovalerate	$C_7H_{14}O_2$	Acros (USA)	108-64-5	99.0
methyl benzoate	$C_8H_8O_2$	Lancaster (England)	93-58-3	99.2
benzyl acetate	$C_9H_{10}O_2$	Acros (USA)	140-11-4	99.6
ethyl salicylate	$C_9H_{10}O_3$	Acros (USA)	118-61-6	99.6
benzyl propionate	$C_{10}H_{12}O_2$	TCI (Japan)	122-63-4	99.4
ethanol	C_2H_6O	Merck (Germany)	64-17-5	99.9

[a] CASRN, Chemical Abstracts Service Registry Number.

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

		ρ/g·cm^{-3}		n_D		σ/mN·m^{-1}	
compounds	*T*/K	exptl	lit.	exptl	lit.	exptl	lit.
ethyl acetoacetate	298.15	1.02345	1.0208[4] 1.02126[5]	1.41658	1.4189[4]	32.30	31.3[6]
ethyl isovalerate	298.15	0.85978	0.86401[7]	1.39401		23.50	23.24[8] [a]
methyl benzoate	298.15	1.08392	1.0839[5] 1.08363[9] 1.0837[10]	1.51467	1.51457[4] 1.51466[9] 1.5152[10]	37.20	
benzyl acetate	298.15	1.05075		1.49982		36.40	
ethyl salicylate	298.15	1.12500		1.52022		36.30	
benzyl propionate	298.15	1.02760		1.49498		34.80	
ethanol	298.15	0.78500	0.78493[11] 0.78515[12] 0.78506[13]	1.35941	1.3593[12] 1.3595[13]	22.30	21.68[11] 21.74[12] 21.8[14]

[a] Data obtained from linear interpolation.

Table 3. Refractive Indices n_D, Surface Tensions σ, Refractive Index Deviations Δn_D, and Surface Tension Deviations $\Delta\sigma$ for the Ethyl Acetoacetate (1) + Ethanol (2) System

x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}	x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}
				T = 288.15 K					
0.00	1.36333	23.00	0.00000	0.000	0.55	1.40575	28.92	0.00061	−1.558
0.05	1.36956	23.53	0.00033	−0.525	0.60	1.40793	29.46	0.00057	−1.418
0.10	1.37506	24.10	0.00051	−0.900	0.65	1.40999	30.02	0.00053	−1.231
0.15	1.37997	24.65	0.00059	−1.200	0.70	1.41190	30.54	0.00049	−1.060
0.20	1.38432	25.22	0.00063	−1.401	0.75	1.41370	31.05	0.00047	−0.878
0.25	1.38823	25.77	0.00065	−1.551	0.80	1.41541	31.53	0.00048	−0.705
0.30	1.39180	26.28	0.00068	−1.681	0.85	1.41698	32.02	0.00045	−0.505
0.35	1.39507	26.78	0.00069	−1.768	0.90	1.41843	32.47	0.00038	−0.328
0.40	1.39807	27.28	0.00069	−1.808	0.95	1.41975	32.91	0.00022	−0.146
0.45	1.40085	27.80	0.00068	−1.787	1.00	1.42091	33.30	0.00000	0.000
0.50	1.40339	28.35	0.00065	−1.699					
				T = 298.15 K					
0.00	1.35941	22.30	0.00000	0.000	0.55	1.40141	27.96	0.00051	−1.599
0.05	1.36544	22.77	0.00018	−0.554	0.60	1.40358	28.52	0.00047	−1.427
0.10	1.37088	23.31	0.00038	−0.930	0.65	1.40563	29.04	0.00044	−1.269
0.15	1.37574	23.83	0.00048	−1.236	0.70	1.40754	29.55	0.00041	−1.099
0.20	1.38006	24.38	0.00053	−1.434	0.75	1.40934	30.06	0.00040	−0.907
0.25	1.38398	24.89	0.00057	−1.604	0.80	1.41105	30.54	0.00042	−0.726
0.30	1.38756	25.38	0.00059	−1.735	0.85	1.41262	31.01	0.00040	−0.537
0.35	1.39081	25.88	0.00061	−1.805	0.90	1.41407	31.46	0.00034	−0.352
0.40	1.39378	26.36	0.00061	−1.849	0.95	1.41540	31.89	0.00020	−0.173
0.45	1.39654	26.86	0.00060	−1.833	1.00	1.41658	32.30	0.00000	0.000
0.50	1.39907	27.40	0.00056	−1.742					
				T = 308.15 K					
0.00	1.35531	21.50	0.00000	0.000	0.55	1.39719	26.84	0.00043	−1.627
0.05	1.36119	21.91	0.00006	−0.572	0.60	1.39935	27.38	0.00040	−1.460
0.10	1.36663	22.42	0.00023	−0.941	0.65	1.40139	27.90	0.00038	−1.288
0.15	1.37143	22.91	0.00035	−1.244	0.70	1.40332	28.40	0.00035	−1.114
0.20	1.37577	23.42	0.00041	−1.451	0.75	1.40513	28.89	0.00034	−0.929
0.25	1.37972	23.90	0.00046	−1.624	0.80	1.40685	29.36	0.00037	−0.746
0.30	1.38331	24.38	0.00050	−1.740	0.85	1.40844	29.81	0.00036	−0.566
0.35	1.38658	24.84	0.00052	−1.827	0.90	1.40990	30.26	0.00030	−0.371
0.40	1.38957	25.32	0.00053	−1.851	0.95	1.41124	30.68	0.00019	−0.192
0.45	1.39231	25.81	0.00051	−1.826	1.00	1.41246	31.10	0.00000	0.000
0.50	1.39485	26.32	0.00048	−1.747					
				T = 318.15 K					
0.00	1.35148	20.60	0.00000	0.000	0.55	1.39280	25.80	0.00035	−1.620
0.05	1.35715	20.98	0.00002	−0.581	0.60	1.39495	26.31	0.00031	−1.475
0.10	1.36248	21.46	0.00009	−0.961	0.65	1.39696	26.81	0.00028	−1.316
0.15	1.36720	21.94	0.00021	−1.256	0.70	1.39889	27.30	0.00028	−1.145
0.20	1.37154	22.43	0.00028	−1.468	0.75	1.40068	27.78	0.00028	−0.965
0.25	1.37546	22.90	0.00035	−1.637	0.80	1.40240	28.24	0.00032	−0.786
0.30	1.37902	23.37	0.00039	−1.751	0.85	1.40399	28.70	0.00032	−0.591
0.35	1.38226	23.83	0.00041	−1.827	0.90	1.40544	29.14	0.00027	−0.401
0.40	1.38523	24.30	0.00042	−1.850	0.95	1.40679	29.56	0.00017	−0.217
0.45	1.38796	24.79	0.00041	−1.816	1.00	1.40803	30.00	0.00000	0.000
0.50	1.39047	25.29	0.00039	−1.738					

Table 4. Refractive Indices n_D, Surface Tensions σ, Refractive Index Deviations Δn_D, and Surface Tension Deviations $\Delta\sigma$ for the Ethyl Isovalerate (1) + Ethanol (2) System

x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}	x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}
				$T = 288.15$ K					
0.00	1.36333	23.00	0.00000	0.000	0.55	1.39028	23.93	0.00002	−0.133
0.05	1.36770	23.14	0.00013	−0.027	0.60	1.39151	23.99	−0.00001	−0.122
0.10	1.37142	23.26	0.00019	−0.052	0.65	1.39264	24.04	−0.00004	−0.118
0.15	1.37464	23.37	0.00022	−0.068	0.70	1.39369	24.09	−0.00005	−0.110
0.20	1.37745	23.46	0.00022	−0.089	0.75	1.39467	24.15	−0.00007	−0.090
0.25	1.37993	23.55	0.00021	−0.097	0.80	1.39559	24.20	−0.00007	−0.076
0.30	1.38213	23.62	0.00018	−0.115	0.85	1.39646	24.25	−0.00007	−0.060
0.35	1.38410	23.69	0.00015	−0.124	0.90	1.39729	24.30	−0.00004	−0.042
0.40	1.38587	23.76	0.00012	−0.125	0.95	1.39807	24.35	−0.00002	−0.022
0.45	1.38748	23.82	0.00009	−0.130	1.00	1.39880	24.40	0.00000	0.000
0.50	1.38894	23.88	0.00005	−0.129					
				$T = 298.15$ K					
0.00	1.35941	22.30	0.00000	0.000	0.55	1.38564	23.05	−0.00004	−0.161
0.05	1.36363	22.40	0.00008	−0.043	0.60	1.38684	23.10	−0.00007	−0.154
0.10	1.36724	22.50	0.00012	−0.067	0.65	1.38795	23.15	−0.00009	−0.143
0.15	1.37038	22.58	0.00014	−0.095	0.70	1.38897	23.20	−0.00011	−0.129
0.20	1.37312	22.66	0.00014	−0.111	0.75	1.38993	23.26	−0.00012	−0.103
0.25	1.37554	22.73	0.00013	−0.125	0.80	1.39083	23.31	−0.00012	−0.084
0.30	1.37769	22.79	0.00011	−0.140	0.85	1.39169	23.35	−0.00011	−0.073
0.35	1.37961	22.85	0.00008	−0.148	0.90	1.39250	23.40	−0.00008	−0.050
0.40	1.38134	22.91	0.00005	−0.149	0.95	1.39328	23.45	−0.00004	−0.026
0.45	1.38291	22.96	0.00002	−0.154	1.00	1.39401	23.50	0.00000	0.000
0.50	1.38434	23.00	−0.00001	−0.165					
				$T = 308.15$ K					
0.00	1.35531	21.50	0.00000	0.000	0.55	1.38102	22.04	−0.00011	−0.179
0.05	1.35942	21.57	0.00004	−0.050	0.60	1.38220	22.08	−0.00013	−0.165
0.10	1.36295	21.64	0.00006	−0.083	0.65	1.38329	22.13	−0.00016	−0.157
0.15	1.36603	21.70	0.00008	−0.113	0.70	1.38430	22.17	−0.00017	−0.138
0.20	1.36872	21.76	0.00007	−0.132	0.75	1.38524	22.22	−0.00018	−0.116
0.25	1.37110	21.81	0.00006	−0.153	0.80	1.38613	22.27	−0.00018	−0.102
0.30	1.37321	21.86	0.00004	−0.165	0.85	1.38698	22.31	−0.00015	−0.076
0.35	1.37510	21.91	0.00002	−0.172	0.90	1.38779	22.36	−0.00012	−0.049
0.40	1.37680	21.96	−0.00001	−0.172	0.95	1.38857	22.41	−0.00006	−0.030
0.45	1.37834	22.00	−0.00004	−0.179	1.00	1.38931	22.45	0.00000	0.000
0.50	1.37974	21.50	−0.00008	−0.181					
				$T = 318.15$ K					
0.00	1.35148	20.60	0.00000	0.000	0.55	1.37644	21.17	−0.00018	−0.189
0.05	1.35545	20.65	0.00001	−0.070	0.60	1.37759	21.21	−0.00020	−0.185
0.10	1.35888	20.72	0.00002	−0.103	0.65	1.37865	21.26	−0.00022	−0.167
0.15	1.36186	20.78	0.00002	−0.133	0.70	1.37964	21.30	−0.00023	−0.158
0.20	1.36447	20.83	0.00000	−0.162	0.75	1.38056	21.35	−0.00024	−0.136
0.25	1.36679	20.88	−0.00001	−0.183	0.80	1.38143	21.40	−0.00023	−0.112
0.30	1.36884	20.94	−0.00003	−0.185	0.85	1.38226	21.45	−0.00020	−0.086
0.35	1.37068	20.99	−0.00005	−0.192	0.90	1.38306	21.50	−0.00015	−0.059
0.40	1.37233	21.03	−0.00009	−0.203	0.95	1.38384	21.55	−0.00008	−0.030
0.45	1.37383	21.08	−0.00012	−0.199	1.00	1.38458	21.60	0.00000	0.000
0.50	1.37519	21.12	−0.00015	−0.201					

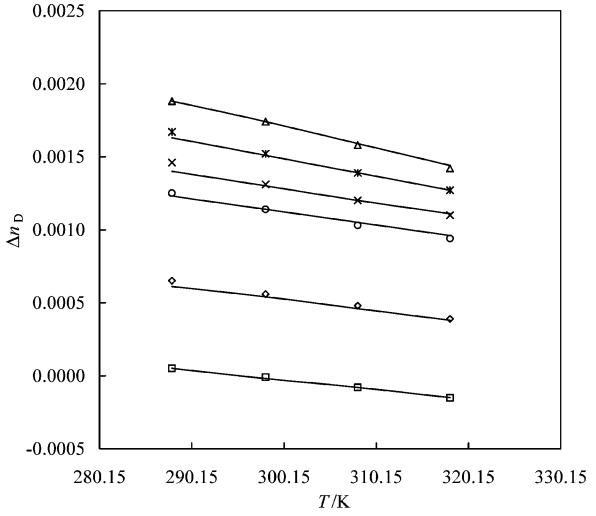

Figure 2. Variation of refractive indices Δn_D at $x = 0.5$ from $T = 288.15$ K to $T = 318.15$ K: ◇, ethyl acetoacetate + ethanol; □, ethyl isovalerate + ethanol; △, methyl benzoate + ethanol; ×, benzyl acetate + ethanol; ∗, ethyl salicylate + ethanol; ○, benzyl propionate + ethanol. Solid lines were calculated from the Redlich−Kister equation.

refractive index measurement was estimated to be less than ± 0.00002.

Surface tensions σ were measured with an automatic surface tension meter model CBVP-A3 (Kyowa, Japan), which works with the Wilhelmy-plate method. The platinum plate was thoroughly cleaned and flame-dried before each measurement. Calibration was performed periodically under atmospheric pressure, in accordance with specifications, using two 200 mg of calibration masses. All liquids were thermostatically controlled to within ± 0.05 K with circulating thermostat water to a jacketed sample vessel. The uncertainty of surface tension measurement was estimated to be within ± 0.2 mN·m^{-1}.

The refractive indexes and surface tensions of the binary mixtures were measured at $T =$ (288.15, 298.15, 308.15, and 318.15) K. A set with the compositions varying from 0.05 to 0.95 mole fractions was prepared for each binary system. The uncertainty in the liquid composition was estimated to be ± 1 × 10^{-4}. All measurements described above were performed at least three times at atmospheric pressure (100.5 ± 0.3) kPa, and an average of at least three measurements was calculated for each temperature.

Table 5. Refractive Indices n_D, Surface Tensions σ, Refractive Index Deviations Δn_D, and Surface Tension Deviations $\Delta\sigma$ for the Methyl Benzoate (1) + Ethanol (2) System

x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}	x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}
				$T = 288.15$ K					
0.00	1.36333	23.00	0.00000	0.000	0.55	1.47785	30.97	0.00180	−3.323
0.05	1.38013	23.80	0.00099	−0.783	0.60	1.48384	31.62	0.00166	−3.282
0.10	1.39497	24.63	0.00165	−1.373	0.65	1.48938	32.29	0.00152	−3.180
0.15	1.40825	25.40	0.00199	−1.884	0.70	1.49451	33.01	0.00136	−2.992
0.20	1.41993	26.18	0.00214	−2.266	0.75	1.49936	33.78	0.00117	−2.722
0.25	1.43052	26.92	0.00220	−2.584	0.80	1.50381	34.60	0.00095	−2.372
0.30	1.44018	27.67	0.00221	−2.803	0.85	1.50803	35.60	0.00076	−1.814
0.35	1.44896	28.37	0.00219	−2.992	0.90	1.51200	36.60	0.00052	−1.232
0.40	1.45703	29.04	0.00214	−3.141	0.95	1.51569	37.60	0.00027	−0.626
0.45	1.46450	29.69	0.00205	−3.248	1.00	1.51915	38.60	0.00000	0.000
0.50	1.47142	30.33	0.00181	−3.310					
				$T = 298.15$ K					
0.00	1.35941	22.30	0.00000	0.000	0.55	1.47332	29.79	0.00163	−3.289
0.05	1.37591	23.00	0.00086	−0.809	0.60	1.47930	30.41	0.00151	−3.251
0.10	1.39068	23.78	0.00144	−1.383	0.65	1.48487	31.05	0.00139	−3.155
0.15	1.40383	24.51	0.00180	−1.875	0.70	1.49000	31.75	0.00121	−2.964
0.20	1.41549	25.25	0.00193	−2.244	0.75	1.49482	32.50	0.00105	−2.692
0.25	1.42606	25.96	0.00199	−2.544	0.80	1.49926	33.30	0.00083	−2.342
0.30	1.43570	26.66	0.00201	−2.769	0.85	1.50347	34.20	0.00065	−1.865
0.35	1.44448	27.30	0.00199	−2.978	0.90	1.50743	35.20	0.00044	−1.265
0.40	1.45255	27.90	0.00194	−3.160	0.95	1.51112	36.20	0.00021	−0.642
0.45	1.46000	28.55	0.00185	−3.234	1.00	1.51467	37.20	0.00000	0.000
0.50	1.46691	29.16	0.00174	−3.295					
				$T = 308.15$ K					
0.00	1.35531	21.50	0.00000	0.000	0.55	1.46859	28.60	0.00150	−3.239
0.05	1.37154	22.10	0.00072	−0.845	0.60	1.47455	29.20	0.00138	−3.198
0.10	1.38617	22.87	0.00121	−1.372	0.65	1.48010	29.80	0.00125	−3.120
0.15	1.39926	23.57	0.00158	−1.844	0.70	1.48520	30.50	0.00108	−2.910
0.20	1.41088	24.28	0.00174	−2.197	0.75	1.49002	31.20	0.00092	−2.669
0.25	1.42138	24.94	0.00182	−2.506	0.80	1.49445	32.00	0.00072	−2.301
0.30	1.43104	25.59	0.00184	−2.743	0.85	1.49866	32.80	0.00056	−1.908
0.35	1.43980	26.20	0.00182	−2.948	0.90	1.50260	33.80	0.00036	−1.293
0.40	1.44784	26.80	0.00176	−3.100	0.95	1.50631	34.80	0.00016	−0.656
0.45	1.45528	27.40	0.00168	−3.195	1.00	1.50993	35.80	0.00000	0.000
0.50	1.46220	28.00	0.00158	−3.239					
				$T = 318.15$ K					
0.00	1.35148	20.60	0.00000	0.000	0.55	1.46384	27.60	0.00133	−3.260
0.05	1.36738	21.20	0.00054	−0.832	0.60	1.46979	28.20	0.00123	−3.215
0.10	1.38191	21.90	0.00100	−1.418	0.65	1.47528	28.80	0.00112	−3.135
0.15	1.39488	22.60	0.00137	−1.880	0.70	1.48039	29.46	0.00098	−2.962
0.20	1.40643	23.30	0.00152	−2.235	0.75	1.48515	30.16	0.00081	−2.719
0.25	1.41686	23.95	0.00161	−2.546	0.80	1.48961	30.92	0.00063	−2.389
0.30	1.42643	24.60	0.00164	−2.777	0.85	1.49378	31.76	0.00047	−1.954
0.35	1.43518	25.20	0.00163	−2.986	0.90	1.49773	32.70	0.00028	−1.396
0.40	1.44318	25.80	0.00157	−3.133	0.95	1.50144	33.80	0.00010	−0.658
0.45	1.45059	26.40	0.00151	−3.223	1.00	1.50514	34.80	0.00000	0.000
0.50	1.45746	27.00	0.00142	−3.264					

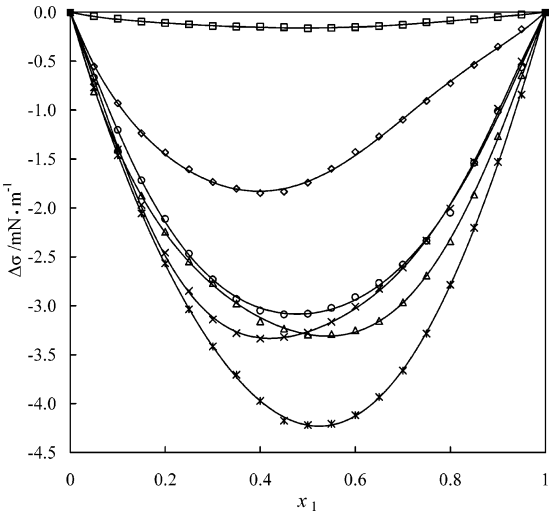

Figure 3. Change of surface tensions $\Delta\sigma$ with mole fraction x_1 at T = 298.15 K: ◇, ethyl acetoacetate + ethanol; □, ethyl isovalerate + ethanol; △, methyl benzoate + ethanol; ×, benzyl acetate + ethanol; ∗, ethyl salicylate + ethanol; ○, benzyl propionate + ethanol. Solid curves were calculated from the Redlich−Kister equation.

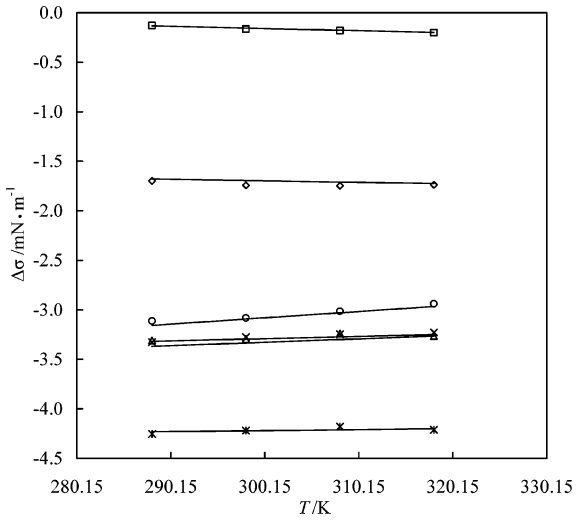

Figure 4. Variation of surface tensions $\Delta\sigma$ at $x = 0.5$ from $T = 288.15$ K to $T = 318.15$ K: ◇, ethyl acetoacetate + ethanol; □, ethyl isovalerate + ethanol; △, methyl benzoate + ethanol; ×, benzyl acetate + ethanol; ∗, ethyl salicylate + ethanol; ○, benzyl propionate + ethanol. Solid lines were calculated from the Redlich−Kister equation.

Table 6. Refractive Indices n_D, Surface Tensions σ, Refractive Index Deviations Δn_D, and Surface Tension Deviations $\Delta\sigma$ for the Benzyl Acetate (1) + Ethanol (2) System

x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}	x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}
				$T = 288.15$ K					
0.00	1.36333	23.00	0.00000	0.000	0.55	1.47020	30.80	0.00138	−3.209
0.05	1.38022	24.00	0.00084	−0.673	0.60	1.47521	31.50	0.00127	−3.046
0.10	1.39462	24.80	0.00119	−1.336	0.65	1.47987	32.20	0.00115	−2.843
0.15	1.40725	25.50	0.00135	−1.925	0.70	1.48418	32.90	0.00102	−2.604
0.20	1.41836	26.10	0.00145	−2.470	0.75	1.48819	33.62	0.00088	−2.313
0.25	1.42813	26.70	0.00152	−2.894	0.80	1.49191	34.40	0.00076	−1.934
0.30	1.43696	27.30	0.00156	−3.215	0.85	1.49539	35.20	0.00063	−1.509
0.35	1.44489	28.00	0.00158	−3.347	0.90	1.49871	36.10	0.00047	−0.960
0.40	1.45211	28.70	0.00157	−3.404	0.95	1.50170	36.90	0.00029	−0.490
0.45	1.45868	29.40	0.00153	−3.394	1.00	1.50439	37.70	0.00000	0.000
0.50	1.46470	30.10	0.00146	−3.327					
				$T = 298.15$ K					
0.00	1.35941	22.30	0.00000	0.000	0.55	1.46565	29.69	0.00123	−3.164
0.05	1.37602	23.20	0.00066	−0.702	0.60	1.47065	30.36	0.00113	−3.010
0.10	1.39033	23.90	0.00101	−1.403	0.65	1.47531	31.02	0.00102	−2.827
0.15	1.40288	24.57	0.00122	−1.968	0.70	1.47958	31.68	0.00089	−2.610
0.20	1.41395	25.18	0.00134	−2.456	0.75	1.48358	32.37	0.00077	−2.332
0.25	1.42372	25.77	0.00142	−2.848	0.80	1.48729	33.08	0.00065	−2.007
0.30	1.43255	26.36	0.00145	−3.141	0.85	1.49076	33.92	0.00053	−1.527
0.35	1.44048	27.02	0.00147	−3.280	0.90	1.49408	34.80	0.00040	−0.985
0.40	1.44767	27.69	0.00145	−3.336	0.95	1.49709	35.60	0.00024	−0.502
0.45	1.45420	28.37	0.00140	−3.318	1.00	1.49982	36.40	0.00000	0.000
0.50	1.46016	29.02	0.00131	−3.276					
				$T = 308.15$ K					
0.00	1.35531	21.50	0.00000	0.000	0.55	1.46119	28.67	0.00112	−3.154
0.05	1.37164	22.30	0.00045	−0.765	0.60	1.46619	29.30	0.00101	−3.029
0.10	1.38588	23.00	0.00084	−1.434	0.65	1.47082	29.94	0.00089	−2.857
0.15	1.39843	23.63	0.00108	−2.012	0.70	1.47508	30.58	0.00077	−2.651
0.20	1.40945	24.23	0.00121	−2.485	0.75	1.47907	31.24	0.00066	−2.395
0.25	1.41929	24.83	0.00130	−2.846	0.80	1.48279	31.93	0.00055	−2.083
0.30	1.42810	25.43	0.00136	−3.110	0.85	1.48626	32.80	0.00043	−1.566
0.35	1.43604	26.07	0.00137	−3.252	0.90	1.48959	33.70	0.00032	−0.997
0.40	1.44323	26.72	0.00134	−3.313	0.95	1.49262	34.50	0.00021	−0.507
0.45	1.44974	27.38	0.00129	−3.302	1.00	1.49539	35.30	0.00000	0.000
0.50	1.45571	28.03	0.00120	−3.247					
				$T = 318.15$ K					
0.00	1.35148	20.60	0.00000	0.000	0.55	1.45653	27.56	0.00101	−3.133
0.05	1.36738	21.30	0.00027	−0.827	0.60	1.46154	28.17	0.00091	−3.018
0.10	1.38161	22.00	0.00061	−1.465	0.65	1.46613	28.78	0.00079	−2.866
0.15	1.39409	22.60	0.00092	−2.045	0.70	1.47036	29.41	0.00068	−2.662
0.20	1.40506	23.20	0.00108	−2.494	0.75	1.47434	30.06	0.00057	−2.408
0.25	1.41487	23.80	0.00119	−2.833	0.80	1.47804	30.75	0.00046	−2.088
0.30	1.42364	24.40	0.00125	−3.079	0.85	1.48148	31.60	0.00036	−1.584
0.35	1.43155	25.03	0.00128	−3.214	0.90	1.48481	32.45	0.00026	−1.058
0.40	1.43870	25.66	0.00125	−3.280	0.95	1.48783	33.30	0.00016	−0.513
0.45	1.44517	26.30	0.00120	−3.275	1.00	1.49064	34.10	0.00000	0.000
0.50	1.45110	26.93	0.00110	−3.227					

Results and Discussion

Tables 3 to 8 list the experimental refractive indices, surface tensions, deviations in the refractive index from volume fraction average, and deviations in the surface tension from volume fraction average for six binary systems: ethyl acetoacetate, ethyl isovalerate, methyl benzoate, benzyl acetate, ethyl salicylate, or benzyl propionate + ethanol at $T = (288.15, 298.15, 308.15,$ and 318.15) K. The values of surface tension and refractive index decrease systematically from 288.15 K to 318.15 K through the whole range of mole fractions.

The deviation in the refractive index Δn_D was calculated from the volume fraction average as suggested by Brocos et al.[15] and is given as

$$\Delta n_D = n_D - \sum_{i=1}^{2} \phi_i n_{D_i} \tag{1}$$

where n_D, n_{D_i}, and ϕ_i are the refractive index of the mixture, the refractive index of pure component i, and the volume fraction of pure component i, respectively. The volume fraction ϕ_i of component i of a mixture is defined by

$$\phi_i = \frac{x_i V}{\sum_{j=1}^{2} x_j V_j} \tag{2}$$

where V and V_j are the molar volume of the mixture and the molar volume of pure component j, respectively. The molar volume V of the mixture was calculated from the following equation:

$$V = \frac{\sum_{i=1}^{2} x_i M_i}{\rho} \tag{3}$$

where M_i is the molecular weight of component i. ρ is the mixture density and was obtained from the previous study.[3] The deviation in the surface tension from volume

Table 7. Refractive Indices n_D, Surface Tensions σ, Refractive Index Deviations Δn_D, and Surface Tension Deviations $\Delta\sigma$ for the Ethyl Salicylate (1) + Ethanol (2) System

x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}	x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}
				$T = 288.15$ K					
0.00	1.36333	23.00	0.00000	0.000	0.55	1.48670	29.71	0.00154	−4.239
0.05	1.38326	24.00	0.00100	−0.700	0.60	1.49239	30.33	0.00141	−4.139
0.10	1.40028	24.80	0.00160	−1.375	0.65	1.49763	31.00	0.00128	−3.949
0.15	1.41504	25.50	0.00197	−1.967	0.70	1.50246	31.70	0.00115	−3.694
0.20	1.42788	26.10	0.00209	−2.508	0.75	1.50695	32.50	0.00100	−3.308
0.25	1.43914	26.65	0.00213	−2.973	0.80	1.51112	33.40	0.00087	−2.793
0.30	1.44918	27.20	0.00208	−3.333	0.85	1.51498	34.40	0.00074	−2.152
0.35	1.45820	27.70	0.00201	−3.652	0.90	1.51852	35.40	0.00056	−1.488
0.40	1.46635	28.10	0.00190	−3.993	0.95	1.52184	36.40	0.00037	−0.804
0.45	1.47375	28.60	0.00179	−4.168	1.00	1.52481	37.50	0.00000	0.000
0.50	1.48051	29.13	0.00167	−4.254					
				$T = 298.15$ K					
0.00	1.35941	22.30	0.00000	0.000	0.55	1.48212	28.66	0.00140	−4.206
0.05	1.37908	23.17	0.00086	−0.768	0.60	1.48782	29.25	0.00129	−4.118
0.10	1.39599	23.90	0.00144	−1.460	0.65	1.49304	29.90	0.00117	−3.933
0.15	1.41072	24.55	0.00184	−2.056	0.70	1.49784	30.60	0.00104	−3.663
0.20	1.42347	25.14	0.00196	−2.567	0.75	1.50232	31.38	0.00092	−3.283
0.25	1.43468	25.65	0.00198	−3.037	0.80	1.50649	32.25	0.00080	−2.785
0.30	1.44465	26.15	0.00193	−3.415	0.85	1.51035	33.18	0.00066	−2.203
0.35	1.45364	26.65	0.00185	−3.706	0.90	1.51391	34.18	0.00049	−1.528
0.40	1.46177	27.10	0.00173	−3.972	0.95	1.51723	35.17	0.00030	−0.843
0.45	1.46918	27.55	0.00162	−4.174	1.00	1.52022	36.30	0.00000	0.000
0.50	1.47593	28.10	0.00152	−4.219					
				$T = 308.15$ K					
0.00	1.35531	21.50	0.00000	0.000	0.55	1.47760	27.68	0.00130	−4.153
0.05	1.37478	22.27	0.00075	−0.829	0.60	1.48328	28.26	0.00119	−4.066
0.10	1.39159	22.94	0.00129	−1.548	0.65	1.48852	28.90	0.00108	−3.881
0.15	1.40628	23.55	0.00171	−2.156	0.70	1.49331	29.56	0.00095	−3.642
0.20	1.41902	24.13	0.00185	−2.653	0.75	1.49779	30.30	0.00084	−3.294
0.25	1.43020	24.67	0.00185	−3.072	0.80	1.50196	31.13	0.00073	−2.829
0.30	1.44012	25.17	0.00179	−3.431	0.85	1.50582	32.03	0.00060	−2.270
0.35	1.44908	25.62	0.00170	−3.755	0.90	1.50938	33.03	0.00043	−1.589
0.40	1.45719	26.08	0.00160	−3.996	0.95	1.51272	34.07	0.00027	−0.849
0.45	1.46462	26.58	0.00150	−4.134	1.00	1.51573	35.20	0.00000	0.000
0.50	1.47139	27.12	0.00139	−4.178					
				$T = 318.15$ K					
0.00	1.35148	20.60	0.00000	0.000	0.55	1.47306	26.80	0.00118	−4.201
0.05	1.37068	21.30	0.00061	−0.907	0.60	1.47873	27.38	0.00108	−4.118
0.10	1.38739	21.97	0.00116	−1.634	0.65	1.48397	28.00	0.00099	−3.958
0.15	1.40202	22.57	0.00159	−2.259	0.70	1.48874	28.67	0.00088	−3.713
0.20	1.41470	23.17	0.00172	−2.743	0.75	1.49321	29.40	0.00077	−3.379
0.25	1.42580	23.70	0.00173	−3.178	0.80	1.49738	30.20	0.00065	−2.947
0.30	1.43567	24.20	0.00166	−3.543	0.85	1.50123	31.10	0.00053	−2.391
0.35	1.44456	24.67	0.00156	−3.853	0.90	1.50479	32.18	0.00038	−1.634
0.40	1.45266	25.17	0.00145	−4.060	0.95	1.50814	33.20	0.00023	−0.916
0.45	1.46008	25.70	0.00137	−4.173	1.00	1.51116	34.40	0.00000	0.000
0.50	1.46685	26.25	0.00127	−4.211					

fraction average $\Delta\sigma$, is given by

$$\Delta\sigma = \sigma - \sum_{i=1}^{2} \phi_i \sigma_i \tag{4}$$

where σ and σ_i are the surface tension of the mixture and the surface tension of pure component i, respectively.

The values of Δn_D decrease with a rise in temperature for all of the mixtures. For the whole composition range, the Δn_D values are positive for all of the mixtures except for the system of ethyl isovalerate + ethanol, which shows some negative Δn_D values. The Δn_D values are also graphically represented as a function of mole fraction for $T = 298.15$ K in Figure 1. The values of Δn_D ($x = 0.5$) show the sequence as follows: ethyl isovalerate + ethanol < ethyl acetoacetate + ethanol < benzyl propionate + ethanol < benzyl acetate + ethanol < ethyl salicylate + ethanol < methyl benzoate + ethanol. The values of Δn_D ($x = 0.5$) at 298.15 K vary from −0.00001 to 0.00174. Figure 2 plots the values of Δn_D ($x = 0.5$) from $T = 288.15$ K to $T = 318.15$ K for all of the systems.

The values of surface tension deviation $\Delta\sigma$ are negative over the whole composition range for all of the mixtures. The $\Delta\sigma$ values are also graphically represented as a function of mole fraction for $T = 298.15$ K in Figure 3. The values of $\Delta\sigma$ ($x = 0.5$) increase as the sequence: ethyl salicylate + ethanol < benzyl acetate + ethanol ≈ methyl benzoate + ethanol < benzyl propionate + ethanol < ethyl acetoacetate + ethanol < ethyl isovalerate + ethanol. Figure 4 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 $\Delta\sigma$ ($x = 0.5$) show no clear temperature dependence on temperature. The values of $\Delta\sigma$ ($x = 0.5$) at $T = 298.15$ K vary from −4.254 mN·m^{-1} to −0.129 mN·m^{-1}.

The mixing functions Δn_D and $\Delta\sigma$ were represented mathematically by the Redlich−Kister equation for correlating the experimental data:[16]

$$Y = x_1 x_2 \sum_{k=0}^{m} a_k (x_1 - x_2)^k \tag{5}$$

where Y refers to Δn_D or $\Delta\sigma$/mN·m^{-1}, and a_k is a coefficient.

Table 8. Refractive Indices n_D, Surface Tensions σ, Refractive Index Deviations Δn_D, and Surface Tension Deviations $\Delta\sigma$ for the Benzyl Propionate (1) + Ethanol (2) System

x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}	x_1	n_D	σ/mN·m^{-1}	Δn_D	$\Delta\sigma$/mN·m^{-1}
				$T = 288.15$ K					
0.00	1.36333	23.00	0.00000	0.000	0.55	1.46918	29.95	0.00118	−3.051
0.05	1.38108	24.00	0.00065	−0.632	0.60	1.47384	30.50	0.00110	−2.947
0.10	1.39613	24.80	0.00113	−1.224	0.65	1.47807	31.06	0.00102	−2.797
0.15	1.40891	25.50	0.00132	−1.725	0.70	1.48193	31.62	0.00092	−2.615
0.20	1.42001	26.13	0.00140	−2.141	0.75	1.48549	32.23	0.00082	−2.354
0.25	1.42966	26.70	0.00145	−2.491	0.80	1.48877	32.88	0.00070	−2.029
0.30	1.43817	27.23	0.00146	−2.777	0.85	1.49183	33.70	0.00059	−1.510
0.35	1.44572	27.76	0.00143	−2.974	0.90	1.49464	34.50	0.00044	−0.991
0.40	1.45248	28.30	0.00139	−3.088	0.95	1.49721	35.20	0.00029	−0.554
0.45	1.45858	28.85	0.00132	−3.128	1.00	1.49950	36.00	0.00000	0.000
0.50	1.46414	29.40	0.00125	−3.113					
				$T = 298.15$ K					
0.00	1.35941	22.30	0.00000	0.000	0.55	1.46464	28.89	0.00108	−3.022
0.05	1.37691	23.20	0.00051	−0.667	0.60	1.46929	29.43	0.00100	−2.911
0.10	1.39185	24.00	0.00095	−1.203	0.65	1.47352	29.97	0.00090	−2.766
0.15	1.40454	24.64	0.00118	−1.717	0.70	1.47737	30.52	0.00082	−2.580
0.20	1.41559	25.25	0.00128	−2.113	0.75	1.48092	31.10	0.00072	−2.337
0.25	1.42524	25.78	0.00134	−2.467	0.80	1.48419	31.70	0.00061	−2.049
0.30	1.43375	26.30	0.00135	−2.731	0.85	1.48725	32.50	0.00051	−1.539
0.35	1.44128	26.80	0.00133	−2.931	0.90	1.49006	33.30	0.00037	−1.010
0.40	1.44801	27.31	0.00129	−3.050	0.95	1.49265	34.00	0.00022	−0.563
0.45	1.45408	27.84	0.00122	−3.087	1.00	1.49498	34.80	0.00000	0.000
0.50	1.45962	28.36	0.00114	−3.082					
				$T = 308.15$ K					
0.00	1.35531	21.50	0.00000	0.000	0.55	1.46031	27.85	0.00096	−2.950
0.05	1.37266	22.30	0.00041	−0.714	0.60	1.46494	28.37	0.00088	−2.846
0.10	1.38756	23.10	0.00082	−1.206	0.65	1.46917	28.88	0.00081	−2.719
0.15	1.40020	23.70	0.00105	−1.721	0.70	1.47302	29.40	0.00072	−2.551
0.20	1.41123	24.30	0.00117	−2.094	0.75	1.47658	29.96	0.00062	−2.318
0.25	1.42091	24.82	0.00125	−2.430	0.80	1.47986	30.55	0.00053	−2.030
0.30	1.42944	25.32	0.00127	−2.690	0.85	1.48292	31.30	0.00043	−1.562
0.35	1.43696	25.80	0.00125	−2.887	0.90	1.48574	32.10	0.00030	−1.025
0.40	1.44370	26.30	0.00119	−2.996	0.95	1.48834	32.80	0.00017	−0.570
0.45	1.44977	26.82	0.00112	−3.026	1.00	1.49074	33.60	0.00000	0.000
0.50	1.45530	27.33	0.00103	−3.015					
				$T = 318.15$ K					
0.00	1.35148	20.60	0.00000	0.000	0.55	1.45569	26.78	0.00087	−2.884
0.05	1.36853	21.34	0.00028	−0.733	0.60	1.46029	27.28	0.00079	−2.791
0.10	1.38322	22.10	0.00066	−1.231	0.65	1.46451	27.78	0.00071	−2.664
0.15	1.39583	22.70	0.00089	−1.718	0.70	1.46833	28.30	0.00062	−2.489
0.20	1.40687	23.30	0.00105	−2.066	0.75	1.47188	28.83	0.00053	−2.278
0.25	1.41651	23.80	0.00114	−2.401	0.80	1.47516	29.40	0.00044	−2.004
0.30	1.42500	24.30	0.00117	−2.641	0.85	1.47824	30.10	0.00035	−1.579
0.35	1.43250	24.78	0.00115	−2.822	0.90	1.48106	30.90	0.00023	−1.035
0.40	1.43922	25.28	0.00109	−2.916	0.95	1.48366	31.60	0.00011	−0.575
0.45	1.44524	25.78	0.00102	−2.953	1.00	1.48611	32.40	0.00000	0.000
0.50	1.45072	26.28	0.00094	−2.940					

Table 9. Coefficients and Standard Deviations δ of Δn_D and $\Delta\sigma$ for the Binary Mixtures from $T = 288.15$ K to $T = 318.15$ K

Y	c_0·10^2	d_0·10^4	c_1·10^2	d_1·10^4	c_2·10^2	d_2·10^4	δ·10^4
			Ethyl Acetoacetate (1) + Ethanol (2)				
Δn_D	0.2927	−0.3142	−0.1519	0.3347	0.4291	−0.7659	0.46
$\Delta\sigma$/mN·m^{-1}	−662.69	−60.55	391.42	−37.15	65.28	−304.41	282
			Ethyl Isovalerate (1) + Ethanol (2)				
Δn_D	0.0584	−0.2589	−0.1621	0.0968	0.1374	−0.3739	0.07
$\Delta\sigma$/mN·m^{-1}	−40.58	−86.78	−6.01	67.15	16.15	−71.32	48
			Methyl Benzoate (1) + Ethanol (2)				
Δn_D	0.8423	−0.5891	−0.6919	0.5422	0.8421	−1.3179	0.64
$\Delta\sigma$/mN·m^{-1}	−1340.59	88.84	−31.53	−62.00	−189.03	−411.31	629
			Benzyl Acetate (1) + Ethanol (2)				
Δn_D	0.6191	−0.3927	−0.4120	0.2027	0.6440	−1.2241	0.53
$\Delta\sigma$/mN·m^{-1}	−1367.97	137.21	322.64	−170.88	107.88	−823.39	573
			Ethyl Salicylate (1) + Ethanol (2)				
Δn_D	0.7218	−0.4765	−0.6770	0.3696	0.9164	−0.7041	0.53
$\Delta\sigma$/mN·m^{-1}	−1699.12	41.59	−184.99	254.41	337.55	−1274.02	358
			Benzyl Propionate (1) + Ethanol (2)				
Δn_D	0.5451	−0.3537	−0.3895	0.1693	0.6762	−1.0708	0.33
$\Delta\sigma$/mN·m^{-1}	−1301.32	255.31	107.36	−53.20	41.66	−556.38	397

Since the coefficient a_k is a function of temperature, we propose a linear dependence on temperature for those coefficients in the studied temperature range. Using this temperature dependence, eq 5 can be written as follows:

$$Y = x_1 x_2 \sum_{k=0}^{m} (c_k + d_k t)(x_1 - x_2)^k \tag{6}$$

where t is the temperature in °C. The values of coefficients c_k and d_k were determined by a nonlinear regression analysis based on the least-squares method and are summarized along with the standard deviations between the experimental and fitted values of the respective functions in Table 9. The standard deviation (δ) is defined by

$$\delta = \left[\sum_{i=1}^{n} \frac{(Y_i^{\text{exptl}} - Y_i^{\text{calcd}})^2}{n - p} \right]^{1/2} \tag{7}$$

where n is the number of experimental points and p is the number of adjustable parameters. The δ values lie between 7.0×10^{-6} and 6.4×10^{-5} and between 0.0048 mN·m^{-1} and 0.063 mN·m^{-1} for Δn_D and $\Delta\sigma$, respectively.

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